GREEN SYNTHESIS, BIOLOGICAL EVALUATION, AND DENSITY FUNCTIONAL THEORY CALCULATIONS OF THIAZOLIDINONE DERIVATIVES – A REVIEW

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ABSTRACT

Objective: Green chemistry articulates an area of research, developing from scientific discoveries about pollution awareness and it exploits a set of principles that reduces or eliminates the usage or generation of hazardous materials in all steps of synthetic progression. Hence, successful introduction of microwave technology is the current exhilarating field in green chemistry, generally classified as microwave-assisted organic synthesis of heterocyclic compounds exclusively thiazolidinone derivatives. Thiazolidinone nucleus especially the 4-thiazolidinone moiety has engaged a distinctive place in the field of medicinal chemistry due to widespread range of biological activities. This variety in the biological response profile has fascinated the consideration of many researchers to discover this skeleton to its manifold potential against numerous activities. This review is complementary to earlier reviews and aims to review the work reported on various biological activities of thiazolidinone derivatives from the year 1991 to the beginning of 2017. Statistics are presented for active compounds, some of which have passed the preclinical testing stage.

Methods: An easy and efficient microwave-assisted protocol has been developed for the green synthesis of thiazolidinone derivatives, and reviewed their inhibitory effects on the activity of various pathogens and was optimized by density functional theory.

Results: All compounds were found to possess various biological activities such as antibacterial, antitubercular, anticancer, antifungal and activities, respectively.

Conclusion: These thiazolidinone derivatives can be believable as new candidates for the treatment of various diseases. Final thoughts attracted from this analysis can perform crucial function shaping the way our team believes concerning existing as well as future projects.

Keywords: Green synthesis, Microwave irradiation, Biological evaluation, Thiazolidinone derivatives

INTRODUCTION

In the new millennium, we have novel biological targets which are defined at the molecular level, which have incredible success in comprehending human illness. On the other hand, the drug design processes are largely including pharmaceutical research study, scientific intuition, reaction, and expertise to drive. With its origins rooted in natural synthesis as well as medicinal chemistry, heterocyclic compounds are existed themselves as a fundamental department of organic chemistry. International Union of Pure and Applied Chemistry defined "cyclic compounds possessing ring members with atoms of at least two distinct elements" [1]. Heterocyclic ring structures are core substances made up of components aside from carbon, where one of the most usual substituents is oxygen, nitrogen as well as sulfur [2,3]. According to the heteroatom[s] existing in the ring structure, heterocyclic could be recorded as oxygen, nitrogen, or sulfur based and within each and every class of compound is based on the size of the ring structure, determined by the total number of atoms [4]. The type and size of the ring structures, as one with the substituent groups of the core scaffold, impact are strongly on the physicochemical properties [5].

Heating responses with conventional devices, such as oil bath and also home heating mantles, are not only slow but also, additionally, it produces a hot surface area on the reaction vessel where reagents, items, and substrates are frequently disintegrate after at some time. Microwave power alternatively is brought right into the chemical reactor from another location and travels through the walls of the response vessel, warming the catalysts, and solvents directly. Microwave dielectric heating drives chain reactions by manipulating the benefit of the potential of some solids and liquids to change electromagnetic radiation into warm. Recently, a new technique has actually concerned the leading edge of chemical investigation, that is, microwave dielectric heating. In a similar method to the introduction of the isomantle, this technical advancement will no question need an adjustment in the drug store's mindset. In the future, the drug store will make use of fast blasts of microwave power to heat and also increase chemical reactions, as opposed to an alternate application in the very first circumstances for the mantle or hot plate. High-speed synthesis with microwaves has actually attracted a significant quantity of rate of interest in most recent years [6]. Considering that, the very first reports on the usage of microwave home heating by Gedye and also Giguere/Majetich groups in speeding up organic chemical conversion in 1986, far more than 2000 write-ups have been released in the area of microwave-assisted organic synthesis [7,8].

Thiazole is heterocyclic derivatives highlight both nitrogen in addition to sulfur atom as significant aspect of the aromatic five-membered ring. Thiazole, derivative 1 was first reported by Hantzsch [9] in 1887. They are usually isomeric with 1, 2-azoles. The derivative with nitrogen and sulfur are referred as isothiazole 2. A thiazole ring is discovered naturally inside the essential vitamin thiamine 3.
Typically, the chemistry of thiazole draws the attention of synthetic organic chemists due to their varied biological activities [10-13] such as antibacterial, antitubercular, anticancer, and antifungal activities. Recently, the usage of thiazole has been uncovered in drug development for the treatment of allergy [14], hypertension [15], inflammation [16], schizophrenia [17], bacterial [18], HIV infections [19], hypnotics [20], and more recently for the treatment of pain [21], as fibrinogen receptor with antithrombotic activity and as new inhibitors of bacterial DNA gyrase B [22].

**DISCOVERY OF THIAZOLE**

The initial synthesis of thiazole has been described by Hantzsch and Weber [23] in 1887. This five membered ring system comprising sulfur and nitrogen hetero atoms at positions-1 and -3, is involved in many of the natural products. In its most basic form, the reaction is provided in Scheme 1 which involves utilization of α-halocarbonyl compound with reactants composed of N-C-S linkage.

![Scheme 1: Synthesis of thiazole](image)

The reaction may also be carried out in the presence of all three carbon atoms being substituted with suitable alkyl or aryl groups. Besides, it reacts with thiourea rather than thioamide to yield the related thiazole 4.

![Scheme 1: Synthesis of thiazole](image)

This specific review deals with typically the primary innovations regarding nitrogen, oxygen, and sulfur-centered heterocyclic scaffolds stressing out there their main roles inside pharmaceutical compounds. To meet typically the pressure on the requirement, within this endeavor, we prepared to synthesize thiazole that contains heterocyclic compounds by microwave condition.

**ADVANTAGE OF MICROWAVE IRRADIATION**

In the past decade, microwave irradiation has grabbed hugeness as a powerful tool for quick and efficient synthesis of numerous compounds due to selective absorption of microwave energy by the polar molecules [25]. The application of microwave irradiation to provide improved reaction rate and increased product in the field of chemical synthesis plus, it is quite successfully used in the organization of a variety of carbon-heteroatom bonds. During modern times, microwaves have been extensively utilized to carry out chemical reactions and possess become a beneficial nonconventional energy source for executing organic synthesis [26,27]. This is maintained an awesome number of journals in recent years especially in the year 2003, related to the application of microwaves as a result of an incredible accessibility of reliable microwave instrumentation [7,28-32].

Al-Shamkhani and Al-Hazam [33] reported a series of substituted 2-amino thiazole 5, by the reaction of acetophenone with thiourea and iodine in microwave oven.

![Al-Shamkhani and Al-Hazam](image)

Samadhiya et al. [34] made an attempt to design by conventional as well as microwave strategies from 2-amino-5-nitrothiazole as a starting material to synthesize N-[2-(2-(substituted phenyl)-4-oxo-5-((substituted benzylidene)-1,3-thiazolidineiminoethyl]-2-amino-5-nitrothiazole.

![Samadhiya et al.](image)
Al-Shamkhani et al. [35] synthesized novel heterocyclic compounds with thiazolidinone derivative 7.

![Image of 7]

Desai et al. [36] synthesized 5-arylidene derivatives 8, which bear a fluorine atom in the 4th site of typically the benzoyl group as starting compound, by the condensation method using conventional and microwave strategies.

![Image of 8]

Dubey et al. [37] synthesized new varieties of thiazolidine derivatives of benzotriazole 9. The reaction was carried out there by both conventional in addition to microwave strategies.

![Image of 9]

Nikalje et al. [38] synthesized a series of new 2-(3-(2-(1,3-dioxoisoxindolin-2-yl)acetamido)-4-oxo-2-phenylthiazolidin-5-yl) acetic acid 10 using microwave irradiation.

![Image of 10]

Mahmoodi et al. [39] synthesized several 1,3-thiazolidine-4-ones 11 by cycloaddition reaction of N-aryl, N’-acyl thiourea with acetylenic esters under microwave irradiation in solvent free conditions.

![Image of 11]

LITERATURE REVIEW OF THIAZOLIDINE-4-ONE DERIVATIVES

Atobe et al. [40] reported as noncompetitive inhibitors of (ADAMTS-5) A disintegrin and metalloproteinase with thrombospondin motifs – 5 for several thiazole bearing thiazolidin-4-ones 12. Compound 12 is apparently the best ADAMTS-5 inhibited and good selectivity over other metalloproteases.

![Image of 12]

Sala et al. [41] produced a library of 2, 3-thiazolidin-4-one derivatives 13 inside which thiazolidinone nucleus attaches two aromatic rings. Several of these compounds revealed strong inhibitory effects on breast cancer cell growth.

![Image of 13]
Pansare et al. [42] synthesized one-pot, three-component, and microwave-assisted preparation of new 3-(4-chloro-2-hydroxyphenyl)-2-(substituted)thiazolidin-4-one 14. This research exhibits that all these compounds were non-cytotoxic in nature and established for their antimicrobial specificity separate from any standard cytotoxicity.

\[
\begin{align*}
\text{14} & \quad \text{O} & \quad \text{N} & \quad \text{Ar} & \quad \text{N} & \quad \text{O} & \quad \text{Cl} & \quad \text{OH}
\end{align*}
\]

Nikalje et al. [43] synthesized N-(2-oxo-2((4-oxo-2-substituted thiazolidin-3-yl)amino)ethyl)benzamide derivatives 15 under microwave irradiation. Typically, the synthesized compound was found to be the most active in maximal electroshock (MES) type. The anticonvulsant screening data exhibit that 65% regarding the compounds were found for their activity in against to MES model when compared to 35% sc-Pentilene - Tetrazol (PTZ) type.

\[
\begin{align*}
\text{15} & \quad \text{O} & \quad \text{N} & \quad \text{Ar} & \quad \text{N} & \quad \text{O} & \quad \text{N} & \quad \text{S}
\end{align*}
\]

Pires Gouvea et al. [46] synthesized 4-thiazolidinones 18 bearing the morpholine moiety. Thiazolidin-4-ones in vivo anti-inflammatory actions were determined using a croton oil-induced ear edema model of inflammation in BALB C sufferers. Best results were found for the synthesized compounds.

\[
\begin{align*}
\text{18} & \quad \text{O} & \quad \text{N} & \quad \text{S} & \quad \text{R}
\end{align*}
\]

Nikalje et al. [43] synthesized N-(2-oxo-2((4-oxo-2-substituted thiazolidin-3-yl)amino)ethyl)benzamide derivatives 15 under microwave irradiation. Typically, the synthesized compound was found to be the most active in maximal electroshock (MES) type. The anticonvulsant screening data exhibit that 65% regarding the compounds were found for their activity in against to MES model when compared to 35% sc-Pentilene - Tetrazol (PTZ) type.

\[
\begin{align*}
\text{15} & \quad \text{O} & \quad \text{N} & \quad \text{Ar} & \quad \text{N} & \quad \text{O} & \quad \text{N} & \quad \text{S}
\end{align*}
\]

Verma et al. [47] synthesized a series of 1,3-thiazolidin-4-ones derivatives 19. The attractive characteristics of this strategy include metal-free mild reaction conditions, fast reaction time and efficiency of forming consecutive C-S and C-N bonds, and a single ring in one synthetic operation.

\[
\begin{align*}
\text{19} & \quad \text{O} & \quad \text{S} & \quad \text{CH}_3 & \quad \text{CH}_3
\end{align*}
\]

Ostapiuk [48] synthesized 2-[[5-benzyl-1, 3-thiazol-2-yl]imino]-1,3-thiazolidin-4-ones 20, 21, and 22 derivatives. The investigation of antibacterial and antifungal verification data revealed that all the tested compounds showed moderate to higher activity in comparison with standard drugs.

\[
\begin{align*}
\text{20} & \quad \text{HN} & \quad \text{N} & \quad \text{O} & \quad \text{CO}
\end{align*}
\]

\[
\begin{align*}
\text{21} & \quad \text{HN} & \quad \text{N} & \quad \text{O} & \quad \text{CO}
\end{align*}
\]

\[
\begin{align*}
\text{22} & \quad \text{HN} & \quad \text{N} & \quad \text{O} & \quad \text{CO}
\end{align*}
\]

Gidaro et al. [45] synthesized N-substituted 1,3-thiazolidin-4-ones derivatives 17. These compounds were evaluated to found their lowest inhibitory concentration (minimum inhibitory concentration) against several clinical Candida spp. with respect to topical and systemic reference drugs.

\[
\begin{align*}
\text{17} & \quad \text{Ar} & \quad \text{N} & \quad \text{N} & \quad \text{S}
\end{align*}
\]

Ali et al. [44] synthesized 2-imino-4-thiazolidinone derivatives 16 and evaluated their in vivo anti-inflammatory activity and their effect on ex vivo cyclooxygenase-2 (COX-2) and tumor necrosis factor. The synthesized derivatives revealed a reduction of 68.32% in typically the level of COX-2 in comparison with the indomethacin which displayed 66.23% inhibition.

\[
\begin{align*}
\text{16} & \quad \text{Cl} & \quad \text{Cl} & \quad \text{HN} & \quad \text{N} & \quad \text{S} & \quad \text{N} & \quad \text{R}
\end{align*}
\]

Gidaro et al. [45] synthesized N-substituted 1,3-thiazolidin-4-ones derivatives 17. These compounds were evaluated to found their lowest inhibitory concentration (minimum inhibitory concentration) against several clinical Candida spp. with respect to topical and systemic reference drugs.
Gilani et al. [49] synthesized several novel thiazolidin-4-one derivatives 23 of the benzothiazole moiety. An antimicrobial property of the derivatives was investigated against bacteria and fungus. Typically, the investigation of antibacterial and antifungal screening data says that all the tested compounds showed reasonable to higher inhibition.

Raza et al. [50] synthesized thiazolidin-4-one derivatives 24. In \textit{in vitro} and \textit{in vivo} assay systems, these were evaluated for their antihyperglycemic activity. Most of the compounds with thiazolidin-4-one moieties exhibited higher antihyperglycemic activity.

Raza et al. [51] synthesized pyrimidine derivatives clubbed with thiazolidin-4-one 25 and their \textit{in vitro} anticancer activities were screened. The produced compound exhibited amazing growth inhibition at single dosage.

D'Ascenzio et al. [52] have incorporated innumerable novel thiazolidin-4-one subsidiaries 26 for the assessment of their anti-toxoplasma gondii action. The results showed that thiazole-based compounds were assessed favorably to control parasite growth.

Patel et al. [53] synthesized thiazolidin-4-one fused with s-triazines 27. The synthesized analogs were further screened for their \textit{in vitro} antibacterial as well as anticancer efficacy against prostate cancer PC3 cells. Some derivatives possessed impressive antimicrobial activity and noticeable anticancer activity.

Zarghi et al. [55] reported an enormous number of 2,3-diaryl-1,3-thiazolidine-4-ones 30 possessing a methyl sulfonyl pharmacophore and analyzed their biological activities for COX-2 inhibitory activity.

Jackson et al. [54] synthesized thiazolidinones 28 and 29 derivatives in a 3-component one-pot reaction with mercaptoacetic acid, phenethylamine, and aryl aldehyde.

Pan et al. [56] synthesized 2-arylimino-3-aryl-thiazolidine-4-ones 31 and 32. All these consequences revealed that the thiazolidinone scaffold was noticeably an exceptional chemotype for the searching of antibiofilm drugs.
Li et al. [57] synthesized a 2-phenylsulfonylhydrazono-3-(2',3',4',6'-tetra-O-acetyl-D-glucopyranosyl)thiazolidine-4-one 33. Bioassay results indicated that this compound exhibits good fungicidal and herbicidal activities.

Gürsoy et al. [58] reported 4-thiazolidinones 34. It was reported that existence of phenolic, hydroxyl, and aryl-substituted methoxy groups improved antibacterial activity.

Sattigeri et al. [59] synthesized thiazolidin-2-one derivatives 35 and antimicrobial activities of the two new thiazole derivatives were also reported.

Dash et al. [60] synthesized 2-substituted-phenyl-3-(1-cyclopropyl-6-fluoro-7-[4-(2,3-dichlorophenylpiperazin-1-yl]-1,4-dihydroquinoine carboxamido -1, 3-thiazolidin-4-ones 36. Compounds with 2-Cl, 3-Cl and 4-Cl were found to have better bacterial activity compared with standard drug ampicillin.

Vicini et al. [61] reported new 2-thiazolylimino-5-arylidene-4-thiazolidinones 37 and indicating that the substituted and unsubstituted 5-arylidene moiety plays a crucial role in improving antimicrobial properties of this class of derivatives.

Jayachandran et al. [62] synthesized 3-[6'-fluoro-7-chloro[13'] benzothiazol-2'-yl]-m-nitrophenyl thiazolidine-4-one 38. A few of these derivatives indicated shows potential antimicrobacterial activity.

Chatrabhuji et al. [63] synthesized 4-thiazolidinones 39. These bitheterocycles and their precursors were examined for antimicrobial activity toward specific strains and they also screened for their antitubercular activity.

Alizadeh et al. [64] synthesized 1,3-thiazolidine-2-thiones 40 for their possible synthetic and pharmacological importance by utilizing simple and affordable starting materials.

BIOLOGICAL REVIEW OF THIAZOLIDINE DERIVATIVES

Several new derivatives N-[(4-oxo-2-substituted aryl-1,3-thiazolidine)-acetamidyl]-5-nitroindazoles 41 and 42 prepared by Upadhay et al. [65]. About three potent compounds 43, 44, and 45 were acknowledged as effective in killing prostatic cancer cells with increased selectivity compared to serine amide phosphates.
Taranalli et al. [66] synthesized thiazolidine-4-one derivatives 46 and evaluated their anti-inflammatory, analgesic in addition to antiulcer activities. The produced compound was discovered as the most active for each activity.

Balzarini et al. [67] synthesized thiazolidin-4-ones 48. Several derivatives of those compounds exhibited modest anti-HIV-1 activity. The anti-HIV process of 2,3-diaryl-1,3-thiazolidin-4-ones derivatives 49 was studied by Abhinit et al. [68] and reported as a new class of antiviral agent with minimal cytotoxicity.

Solomon et al. [69] reported typically the synthesis 1,3-thiazolidin-4-one nucleus at the terminal chain amino group regarding 4-aminoquinoline 50. All the particular synthesized compounds were analyzed for their antimalarial activity and some compounds to be able to have shown outstanding activity compared to the reference drugs.

Amin et al. [70] reported activity of coumarinyl thiazolidin-4-ones 51 and 52 and evaluated their anticonvulsant activity. These derivatives were found to have better activity against PTZ-induced seizures.

Agarwal et al. [71] synthesized 5-[[2-phenyl-4-oxo-thiazolidin-3-yl]amino]-2-oxo-thio barbituric acids derivatives 53 and Archana et al. [72] synthesized 3-[[4-[2-alkylphenyl]-4-oxo-1,3-thiazolidin-3-yl]-1,3,4-thiadiazol-2-yl]methylamino)-2-methyl-6-monoquinazolin-4(3H)-one 54 and screened in vivo for their anticonvulsant activity.
Shih and Ke [73] synthesized sydnonyl moiety substituted thiazolidinone in addition to thiazoline derivatives 55 and evaluated for their antioxidant activity. The antioxidant action of these derivatives has been identified to exhibit a new significant 1,1-diphenyl-2-picrylhydrazyl radical scavenging activity equivalent to that of Vitamin E.

Rosy et al. [74] synthesized 4-(4-hydrazinylbenzyl) 1, 3-oxazolidin-2-one derivative, and characterized by Fourier-transform infrared (IR) spectroscopy and 1H nuclear magnetic resonance spectral analysis and further studied for their interactions with topoiso merase II DNA gyrase enzymes by molecular docking protocol 4-(4-hydrazinylbenzyl) 1 and 3-oxazolidin-2-one compound and reported a good glide score value and glide energy.

Naraboli and Biradar [75] synthesized and evaluated antimicrobial and antioxidant activity of N-phenylpropyl-3-substituted indoline-2-one derivatives and showed that some of the synthesized compounds exhibited promising results.

Walmik et al. [76] studied indole derivatives and evaluated for their antimicrobial activity against bacterial strains such as Escherichia coli (MTCC723), Staphylococcus aureus (ATCC-29513), Klebsiella pneumoniae (NCTC-13 368), and Pseudomonas aeruginosa (MTCC-1688) and the fungal strains such as Aspergillus oryzae (MTCC-3567T), Aspergillus niger (MTCC-281), Aspergillus flavus (MTCC-1973), and Aspergillus terreus (MTCC-1782) and reported most of the compounds showed appreciable antimicrobial activity against the tested bacteria and fungi and emerged as potential molecules for further development.

Chaubey [77] reported pyridine is found to have a large number of biological activities those including antiviral, anticancer, antimicrobial, antidiabetic, and antitubercular and its derivatives are very much used as anticancer, antimicrobial, antiviral, antidiabetic, and antithrombotic agents.

**DENSITY FUNCTIONAL THEORY (DFT) REVIEW OF THIAZOLIDINONE**

Parthiban et al. [78] synthesized and study the docking pattern and anti-inflammatory activities of some novel analogs of imidazo [1, 2-a] pyridines by means of the protein sequences for prostaglandin reductase. It was found that all the synthesized derivatives possessed very good binding energy, bringing into concern that the compounds are good inhibitors of prostaglandin reductase and hence are vested with anti-inflammatory properties.

Rosy et al. [79] have reported that the metal ion interactions of 2-Thu are consistent with the bonding of ligands through sulfur in all the complexes of Cd (II), Hg (II), Cu (II), and Zn (II) bromides by IR spectroscopy.

Pîrna et al. [80] have reported molecular structure and relative energies of the three possible tautomers of thiazolidine by DFT calculations and it was reported that will form the thione 56 tautomer is most stable in gas-phase and also inside water and dimethyl sulfoxide.

Meng et al. [81] synthesized 2-substituted imino-3-substituted-5-heteroarylidene-1,3-thiazolidine-4-ones 57 as the potent bidentate PTP1B inhibitor. Biological screening test against PTP1B revealed that most of these compounds have positive inhibitory activity against PTP1B.

Mistry et al. [82] synthesized 4-oxo-thiazolidine 58 and their biological activities were evaluated. They will show comparatively excellent antibacterial as well as antifungal and antitubercular activities.

Adki et al. [83] reported synthesis of 1,3-benzothiazol-2-y)l)-1,3-thiazolan-4-one 59 and evaluated their antimicrobial activity. More number of compounds showed a good degree of antimicrobial activity.

Desai and Mistry [84] performed microwave-assisted synthesis of thiazolidinone 60. They finalized that the percentage yield with microwave-irradiated synthesis was good than the conventional process.
Cyclization of compounds can be supported by using thioglycolic acid and chloroacetyl chloride. By heterocyclization of Quinoline-imines with thioglycolic acid using zeolite 5A° under microwaves afforded 2-(2-chloroquinoline-3-yl)-3-substituted phenyl thiazolidin-4-one. Thiazolidin-4-one have a broad band of pharmacological properties i.e., Antifungal, Anti-tubercular, Antimicrobial, Antioxidant, Cytotoxic, Anti-inflammatory. Analytic, Anti YFV (yellow fever virus) activities. Antimicrobial is the most effective activity of the thiazolidine-4-ones. Anticancer and anti HIV are most promising activities of thiazolidin-4-ones for the researchers for the improvement of novel anticancer and anti HIV agents, which is prerequisite of currently medicinal field.

CONCLUSION

This review shows that microwave irradiation method is fastest approach of synthesis with very much better yield than typically the conventional technique.

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