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(*Mini Review*)

4-Thiazolidinones: A structural motif of great synthetic and biological activities

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ABSTRACT

Thiazolidinone heterocyclic-based compounds have been extensively explored for their potent pharmacological activities. Among several existing isomers of thiazolidinone according to the position of the carbonyl group, the 4-thiazolidinones privileged scaffolds showed significant importance in modern medicinal chemistry. The current minireview highlights the various involvement of 4-thiazolidinones in different diseases as bioactive candidates. This minireview summarises recent developments in the chemical and biological activities of 4-thiazolidinones, which have proven to be extremely significant in the production of numerous compounds and, consequently, the treatment of numerous diseases. Additionally, the resulting enhancement of activity due to certain molecular hybridization of 4-thiazolidinones with other moieties was illustrated. The 4-thiazolidinones and their derivatives are the most significant and well-researched chemicals. A range of biological actions of this family of chemicals are gathered and addressed in this systematic minireview. This minireview discusses the impact of various substituents on molecules' biological activity. The examples given here can serve as a useful guide for developing novel tiny molecules with high biological activity.

Keywords : Thiazolidinones, Anti-cancer, Anti-bacterial, Anti-tuberculosis.

1. Introduction

Among the different chemical skeletons, Thiazolidinone is a wonderful nucleus with great synthetic and biological importance. Thiazolidinone moiety has been incorporated into a broad range of biologically active compounds (1, 2). It has been considered a magic moiety that possesses a wide range of biological activities as shown in **Figure 1** such as anticancer (3), antiviral (4), anti-HIV (5), anti-inflammatory (6), antidiabetic activity (7), antibacterial (8), antifungal (9), and antitubercular activities (10). This broad diversity in biological activities gives more attention to synthesizing more new thiazolidinone derivatives and to studying mechanisms of action.

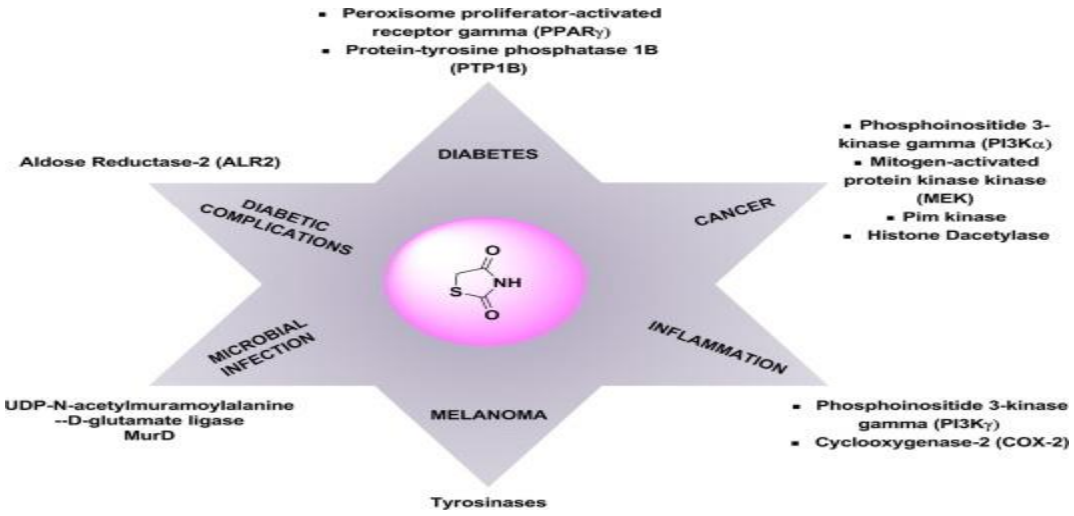
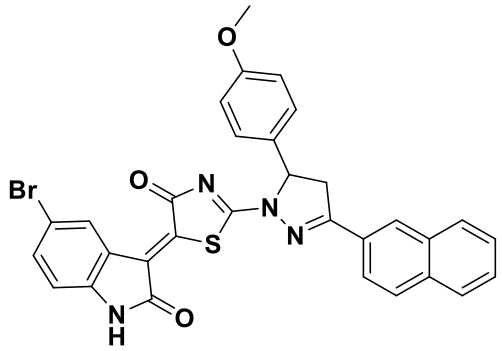


Figure.1. Different pharmacological activities of thiazolidinone ring (11).

2. Insight into different biological activities

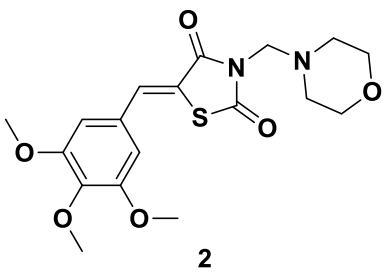
2.1. Anticancer activity

Kobylinska *et al* investigated the anticancer activity of les-3833 1, a thiazolidinone-based lead compound. This compound demonstrated tumor-suppressing activity *in vitro* and *in vivo*. It was tested against the human glioma cell line. It revealed IC_{50}: 0.84 µg/mL which is better than doxorubicin with IC_{50}: 0.90 µg/mL and GI_{50} and TGI values of 0.071 µM and 0.76 µM, respectively (12).



Ies-3833
1

Several thiazolidinone derivatives were designed, synthesized, and revealed good anti-cancer activity. In 2020, El-Kashef *et al* synthesized a set of thiazolidinone derivatives bearing a 5-(3, 4, 5-tri methoxy) benzylidene moiety. The synthesized derivatives were investigated for their anti-breast cancer activity against human breast cancer cell lines MCF-7 and MDA-MB-231. It was shown that compounds 2, 3, and 4 have the greatest anticancer activity with IC_{50} = 1.27, 1.50, and 1.31 µM respectively. They induced apoptosis of human breast cancer without affecting the normal non-cancerous breast cells (13).



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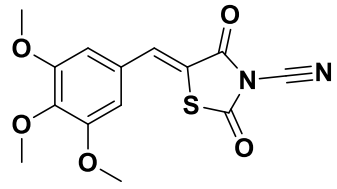

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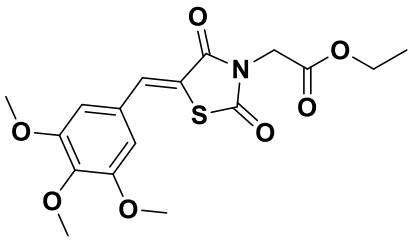
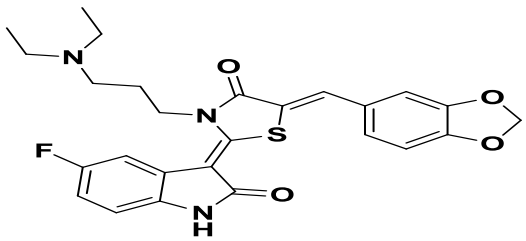


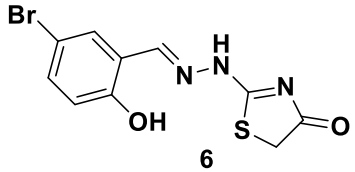
4

Compound **5** was tested against four human cancer cell lines (HT-29, H460, MDA-MB-231, and SMMC-7721) using MTT assay and exhibited IC_{50} values: 0.025, 0.075, 0.77, and 1.95µM respectively, compared with reference sunitinib with IC_{50} values 1.30, 2.70, 3.70 and 6.47µM respectively (14).



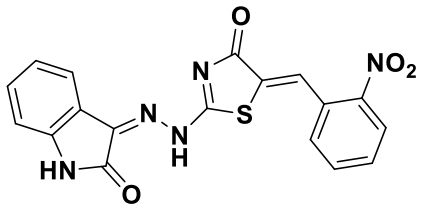
5

According to LV *et al*, compound **6** possessed a good kinase inhibitory activity, with IC_{50} values of 0.09 µM and 0.42 µM against endothelial growth factor receptor (EGFR) and human epidermal growth factor receptor 2 (HER-2), respectively. Compounds containing aryl hydrazones moieties were found to have more inhibitory activity than those containing aliphatic hydrazones (15).



6

In another study, the synthesis of some isatin-thiazolidinone hybrids and their cytotoxic activity against several cancer cell lines *in vitro* were reported. Among these compounds, 5-(2-nitrobenzylidene)-2-(isatin-3-azino)-thiazolidin-4-one **7**, which is proved to be the most active derivative, induced S phase arrest of the cell cycle in a time-dependent manner (16).

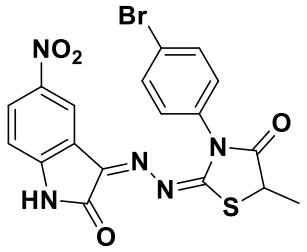


7

2.2. Antiviral activity

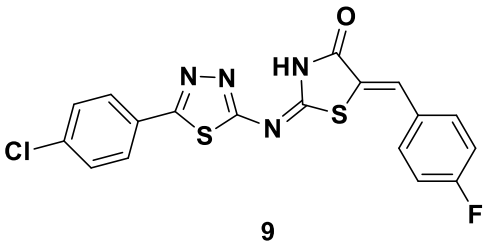
Numerous contagious diseases have been spread during the past few decades, posing major risks to global health. Examples of zoonoses that have spread over the world include Ebola, dengue fever, yellow fever, swine flu, severe acute respiratory syndrome (SARS), and the most recent coronavirus illness (COVID-19) (17).

A literature review revealed that several thiazolidinone derivatives display promising antiviral activity against different pathogens that cause serious illness. A series of hydrazinylidene thiazolidin-4-one derivatives were tested against the yellow fever virus and the bovine viral diarrhea virus (BVDV). The introduction of a methyl group at the 5-position of the thiazolidinone ring was responsible for detectable antiviral activity. The IC_{50} value of compound **8** against (BVDV) was reported to be 13 µg/mL (18).



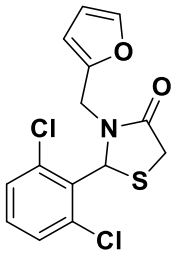
8

Compound **9** is a thiazolidinone derivative which is a non-nucleoside inhibitor of the non-structural protein 5B (NS5B) of dengue virus (DENV). It was found to play an important role in the viral replication cycle (19).



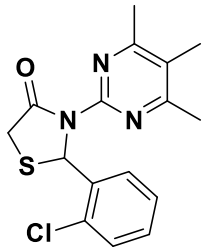
9

Rawal *et al* reported the anti-HIV activity of 2,3-diaryl substituted 4-thiazolidinone derivative (**10**). The results revealed that it is effective against HIV-1 reverse transcriptase enzyme at 0.20 µM concentration, with low cytotoxicity (20).



10

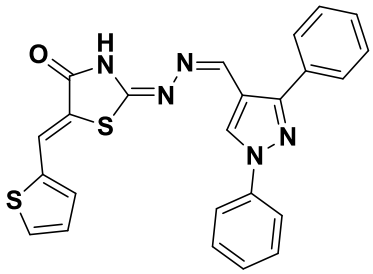
Additionally, a series of 2-aryl-3-(4,5,6-trimethylpyrimidin-2yl)-thiazolidin-4-ones was synthesized. Compound **11** displayed outstanding anti-HIV-RT activity (IC_{50}: 2.91 µM). Compared to (2-(2,6-dichlorophenyl)-3-(4,5,6-trimethylpyrimidin-2yl)-4-oxothiazlidin-5-yl acetate), that is used as a positive control (IC_{50}: 15.86 µM) (21).



11

2.3. Anti-inflammatory and analgesic activity:

A series of 5-arylidenethiazolidin-4-ones hybridized with the pyrazole moiety was tested for its anti-inflammatory activity using carrageenan-induced paw edema test in rats and the ability to inhibit the production of the inflammatory cytokine TNF α in serum was assessed. Compounds **12** and **13** had the highest potency with edema inhibition = 98.16 % and 96.73 %, respectively, compared to indomethacin with 96.94 % inhibition (22).



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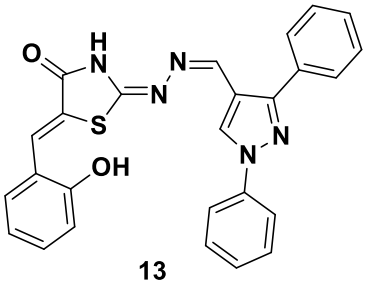
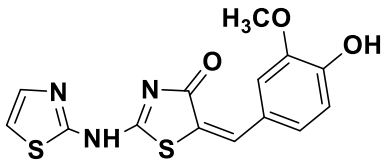


13

In a series of 2-thiazolylamino-5-arylidene-4-thiazolidinones, compound **14** showed the best inhibitory activity on carrageenin–induced rat paw edema (% inhibition = 72.7 %) (23).

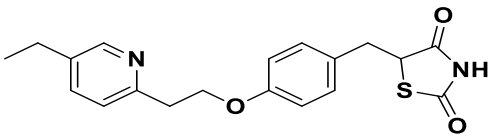


14

2.4. Antidiabetic activity

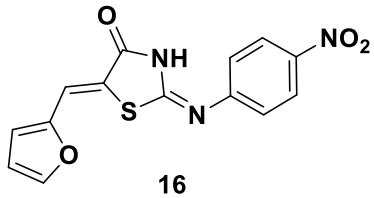
Thiazolidinone is a magic moiety used in the design of valuable anti-diabetic drugs. The 2, 4 thiazolidinedione pioglitazone **15** is a well-known oral drug used for the management of type 2 diabetes mellitus (24).

Pioglitazone is a selective agonist at peroxisome proliferator-activated receptor gamma (PPARγ) in target tissues for insulin action such as adipose tissue and the liver. Its activation increases the transcription of insulin-responsive genes involved in the control of glucose and lipid production, transport, and utilization. So, it promotes both tissue sensitivity to insulin and reduces the hepatic production of glucose (25).



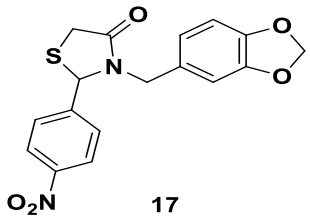
pioglitazone
15

Several thiazolidinone derivatives with potential antidiabetic activity were designed and synthesized. The inhibitory activity was tested against protein tyrosine phosphatase 1B (PTP1B) measuring the effect of each compound on phosphate production. Compound **16** showed maximum inhibition IC_{50}: 5.88 µM, compared to suramin with IC_{50}: 10.98 µM (26).



16

On the other hand, in 2020 Bilgicli *et al* reported that, the piperonyl-based thiazolidine-4-one derivative, namely: 3-(benzo[d][1,3]dioxol-5-ylmethyl)-2-(4-nitrophenyl) thiazolidine-4-one (**17**) acted by inhibiting of α-glucosidase enzyme at minor concentration with IC_{50}: 5.9 nM (27).

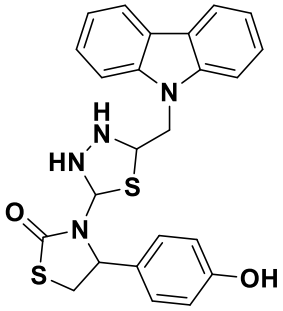


17

2.5. Anticonvulsant and antidepressant activity

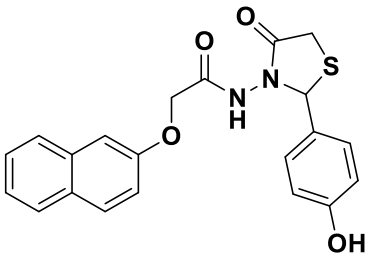
Several substituted thiazolidine-carbazole compounds are considered to be effective antipsychotic and anticonvulsant agents. In this study, derivatives having a thiazolidinone ring shows powerful antipsychotic and anticonvulsant properties. Among these, compound **18** shows a

very positive reaction to psychotic illnesses, where it exhibited very good response against stereotyped behavior induced by amphetamine (28).



18

Moreover, some novel 4-thiazolidinone derivatives were synthesized and their antidepressant activity was evaluated through a forced swim test (**FST**). It is a common test used for evaluation of the efficiency of antidepressants. Results showed that compound **19** has good antidepressant activity with 88.8 sec. duration of immobility, when compared to imipramine with 74.6 sec. duration of immobility (29).

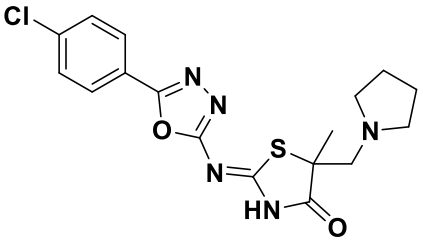


19

2.6. Antibacterial and antifungal activity:

Despite the wide variety of chemical compounds used against microbial and fungal infections, diseases continue to grow in strength. Extensive use of antibiotics contributed to the development of bacterial resistance, so there is an urgent need to synthesize new antimicrobial drugs with different mechanisms of action to overcome antibiotic resistance (30).

Kocabalkanli *et al* synthesized several derivatives of 2,5-disubstituted 4- thiazolidinones and tested their antimicrobial activity. It was found that **20** was more potent than **21** (31).



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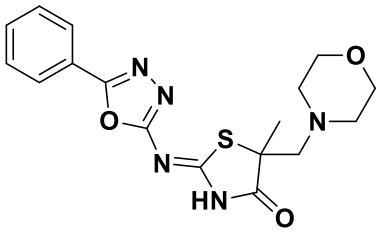
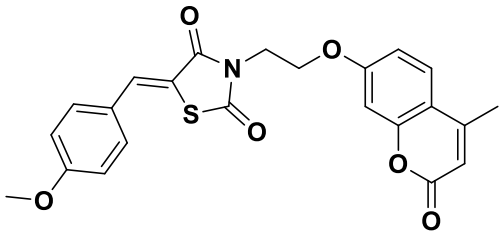


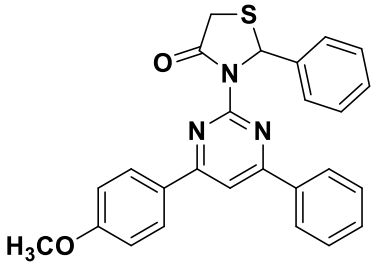
21

Additionally, compound **22** exerted good antibacterial and antifungal activities. This derivative was active against Gram-negative bacteria *E. coli* and *P. aeruginosa* at MIC: 1 µg/mL. It also showed significant antifungal activity at a concentration 1 µg/mL against *A. flavus, T. harzianum, C. albicans,* and *P. chrysogenum.* All these results were higher than the reference drugs ciprofloxacin (MIC: 2 µg/mL) and fluconazole (MIC: 2 µg/mL) (32).



22

Gopalakrishnan *et al* synthesized 2-phenyl-3-(4,6 diarylpyrimidin-2-yl) thiazolidin-4-ones **23, 24** and evaluated their antibacterial activity against *S. aureus, K. Kneumonia*, *P. aeruginosa*, and *E. coli* using ciprofloxacin as standard drug. The results confirmed that the presence of the 4-OCH_3 group at the phenyl ring exerts the maximum antibacterial activity against all the tested bacterial strains, while compounds with electron-withdrawing (4-chloro and 4-fluoro) did not improve the antibacterial activity (33).



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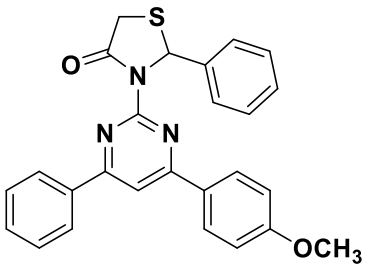
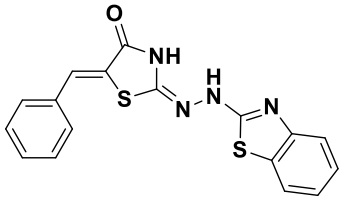


24

Compounds **25-27** and their insertion complex with β- cyclodextrin **28** were tested by S. Dash *et al* in 2020 for their antibacterial activity against *E. coli*, *S. aureus*, and *P. vulgaris*. The results showed that the insertion complex with compounds **25-27** has higher activity than the simple small molecules **25-27**. Inhibition zones of compound **27** were in the range of 15-16 mm, while in insertion complex it showed an increase in the zones of inhibition of 19-21mm (34).



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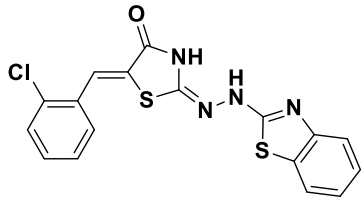

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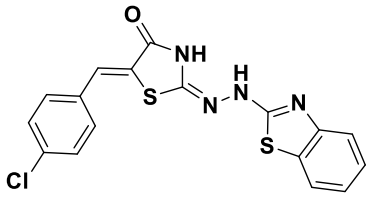
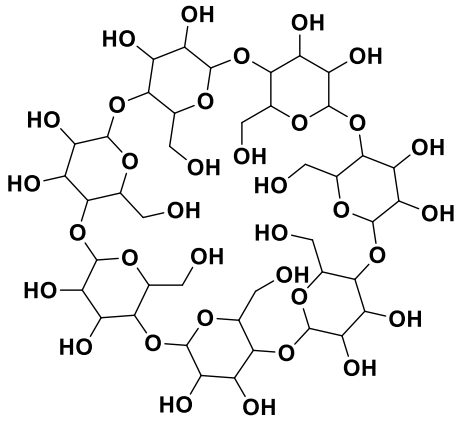


27

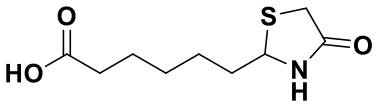


β-cyclodextrin
28

2.7. Antitubercular activity

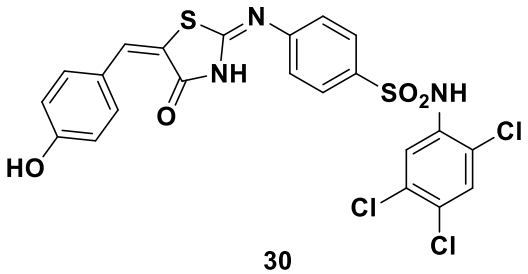
Tuberculosis is a deadly disease that appears as one of the leading causes of infectious disease mortality worldwide. Incomplete drug treatment gives rise to multidrug-resistant and extensively drug-resistant forms of tuberculosis. Therefore, there is a deep need to develop new antituberculosis drugs, particularly against the hard-to-kill multidrug-resistant and other latent forms. 4-Thiazolidinone scaffold has provided to be the cornerstone for the discovery of a number of novel antitubercular agents in recent years (35).

Acidomycin **29**, featuring the thiazolidin-4-one moiety, was found to exert a potent anti-tubercular activity against several MDR and XDR *Mtb* strains (MIC = 0.096-6.2 μM) (36).



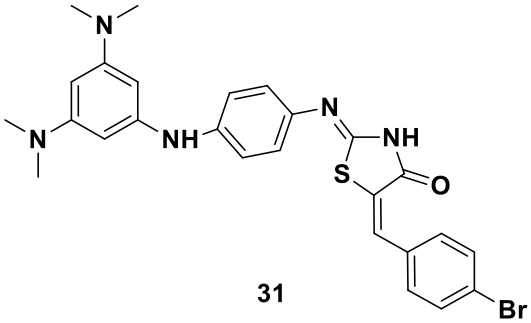
Acidomycin
29

The benzene sulfonamide hybrid with the 2-iminothiazolidin-4-one motif **30** led to an efficient anti-TB activity (H37Rv *Mtb* MIC = 3.12 µg/mL) over streptomycin (6.25 µg/mL) and a comparable activity to ciprofloxacin (3.12 µg/ mL) (37).



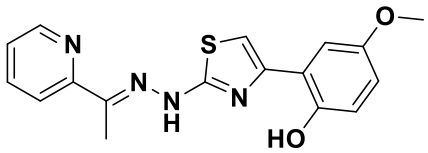
30

Compound **31** was found to be as potent as pyrazinamide with MIC = 32 µg/mL. It was shown that the presence of an electron-withdrawing group at position 4 of the benzylidene ring improved activity (38).



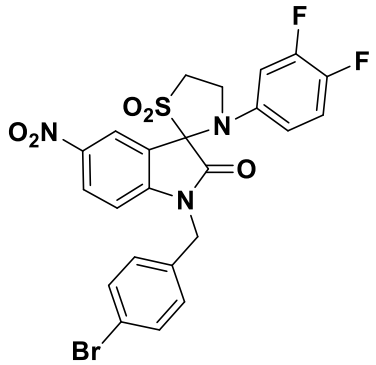
31

N-Pyridyl-*N*-thiazolyl-hydrazine derivatives were synthesized by Zitouni *et al*. Compound **32** revealed a potent antitubercular activity with IC_{50}: 6.22 µg/mL, and its structural details showed that the 2-pyridyl and 2-hydroxy-5-methoxyphenyl groups are required for antimycobacterial activity (39).



32

Additionally, M-tuberculosis protein tyrosine phosphatases A and B (Mptp A and B) secreted into the host cell by growing M. tuberculosis, is considered an important target for treatment of the disease. Vintonyak *et al* designed a series of indoline-2-one-3-spirothiazolidines as a new class of potent and selective inhibitors of MptpB. Compound **33** was found to have high inhibitory activity against *M-tuberculosis* protein tyrosine phosphatase B enzyme (IC_{50}: 1.1 µM) (40).



33

3. Conclusion

The 4-thiazolidinones scaffold was introduced as a successful platform to develop anti-TB, anticancer, antiviral, antidiabetic, anti-inflammatory, and antibacterial compounds. Further investigation regarding particular pharmacophoric features covering more chemical spaces around the 4-thiazolidinone core and their effect on certain phenotypic biological abnormalities was needed. Utilizing 4-thiazolidinone moiety potentiates fruitfully the pharmacological activities, particularly, regarding the development of cancer-targeted therapeutics, which encourages researchers to design new compounds containing thiazolidin-4-one nucleus.

- **Conflict of Interest**

The authors declare no conflict of interest.

4. References

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