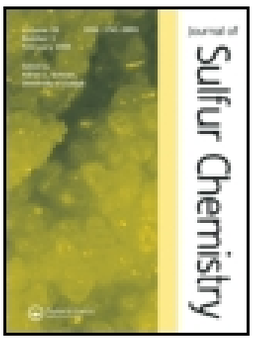

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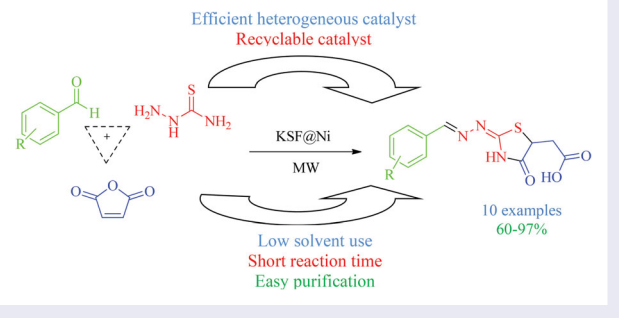
Microwave-assisted one-pot three-component synthesis of thiazolidinones using KSF@Ni as an efficient heterogeneous catalyst

Nosrat O. Mahmoodi, Samane Mohammadgholipour and Fateme Ghanbari Pirbasti

Department of Chemistry, Faculty of Science, University of Guilan, Rasht, Iran

ABSTRACT

Convenient, one-pot three-component reaction for the synthesis of thiazolidinones from aldehydes, thiosemicarbazide, and maleic anhydride in the presence of KSF@Ni as heterogeneous catalyst under microwave irradiation was developed. Products were obtained in reasonable yield in short reaction time.



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KSF@Ni; multi-component reaction; microwave irradiation; antibacterial; thiazolidinone

1. Introduction

The existence of heterocyclic compounds in pharmaceutical and natural products has attracted great interest of organic chemists. Thiazolidinone derivatives are a class of heterocyclic compound with diverse biological and pharmaceutical activities such as cytotoxicity [1], antiproliferative and tumor inhibitory [2], antihyperglycemic [3], antifungal [4], antitoxoplasma gondii [5], antibacterial [6], anti-urease [7], antiviral [8], antischistosomal [9], antimalarial [10], herbicidal [11], antidiabetic [12], anti-candida, and antioxidant activities [13]. Besides, thiazolidinones moieties are present in some drugs such as ralitoline (anticonvulsant), etozoline (diuretic), and thiazolidomycin (active against streptomyces species).

CONTACT Nosrat O. Mahmoodi mahmoodi@guilan.ac.ir Department of Chemistry, Faculty of Science, University of Guilan, P.O. Box 41335-1914, Rasht, Iran

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Multicomponent reactions have gained a great deal of attention as they provide easy and rapid access to a large, complex and diverse library of compounds without isolation of any intermediate and have other advantages such as productivity, simple procedures, convergence, time and energy saving, and high product yields. A survey of the literature showed that a number of research efforts have been devoted to multi-component synthesis of thiazolidinones, *e.g.* synthesis of 2-hydrazolyl-4-thiazolidinones [14], 2-imino-4-thiazolidinone [15], and spiro[indole-thiazolidinones] [16].

Recently, microwave irradiation has been used as an efficient energy source in many chemical reactions [17–20]. The advantages of microwave-assisted synthesis are rate accelerations, high selectivity, improved yields, less by products, shorter reaction times, and easier work-up.

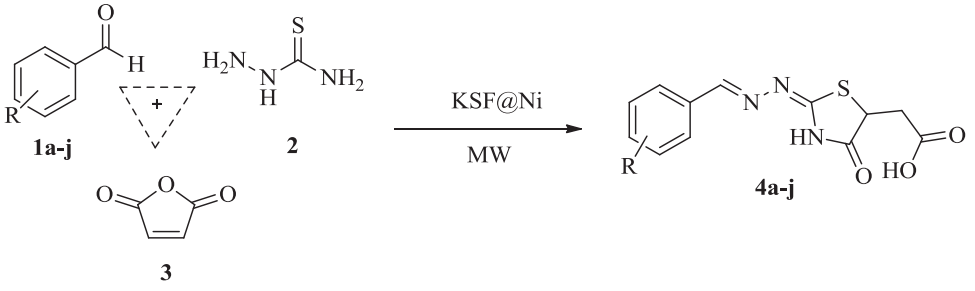
Heterogeneous catalysts are widely used over homogenous catalysts due to their advantages such as non-toxicity, easy of handling, storage safety, long-life times, safe, and easy disposal, easy recovery by filtration and tolerance of a wide range of temperatures and pressures. Montmorillonite KSF is a heterogeneous solid acid catalyst which has been used widely in synthesis of coumarin-3-carboxylic acids [21], in the conversion of amines to *N*-tert-butylcarbamates [22], in aza-Diels–Alder reactions of methylenecyclopropanes with arenecarbaldehydes and arylamines [23], in the synthesis of coumarins via Pechmann reaction [24], in the synthesis of 1,4-dioxo-3,4-dihydrophthalazine-2(1*H*)-carboxamides and carbothioamides [25], and in the synthesis of 1,5-diaryl pyrazoles [26].

2. Results and discussion

2.1. Chemistry

Recently, we report various multi-component reactions efficiently catalyzed by metals supported on KSF [27–35]. In continuation of our previous reports, we investigated in this report one-pot three-component synthesis of thiazolidinones catalyzed by the presence of premade KSF@Ni (Scheme 1).

The FT-IR spectra of KSF and KSF@Ni are depicted in Figure 1. Ni–OH vibration appeared at 3629 cm^{-1} and other vibrations, *e.g.* O–H, H–OH, Si–O, Si–O–Si appeared at 3433, 1637, 1044, 526 and 466 cm^{-1}, respectively [36] (Figure 1). The XRD patterns of KSF@Ni indicated the diffraction peaks at $2\theta = 18°$, 35°, 45° and 55° that confirmed translocation of Ni on KSF surface [37,38] (Figure 2).



Scheme 1. Synthesis of products **4a**–**j**.

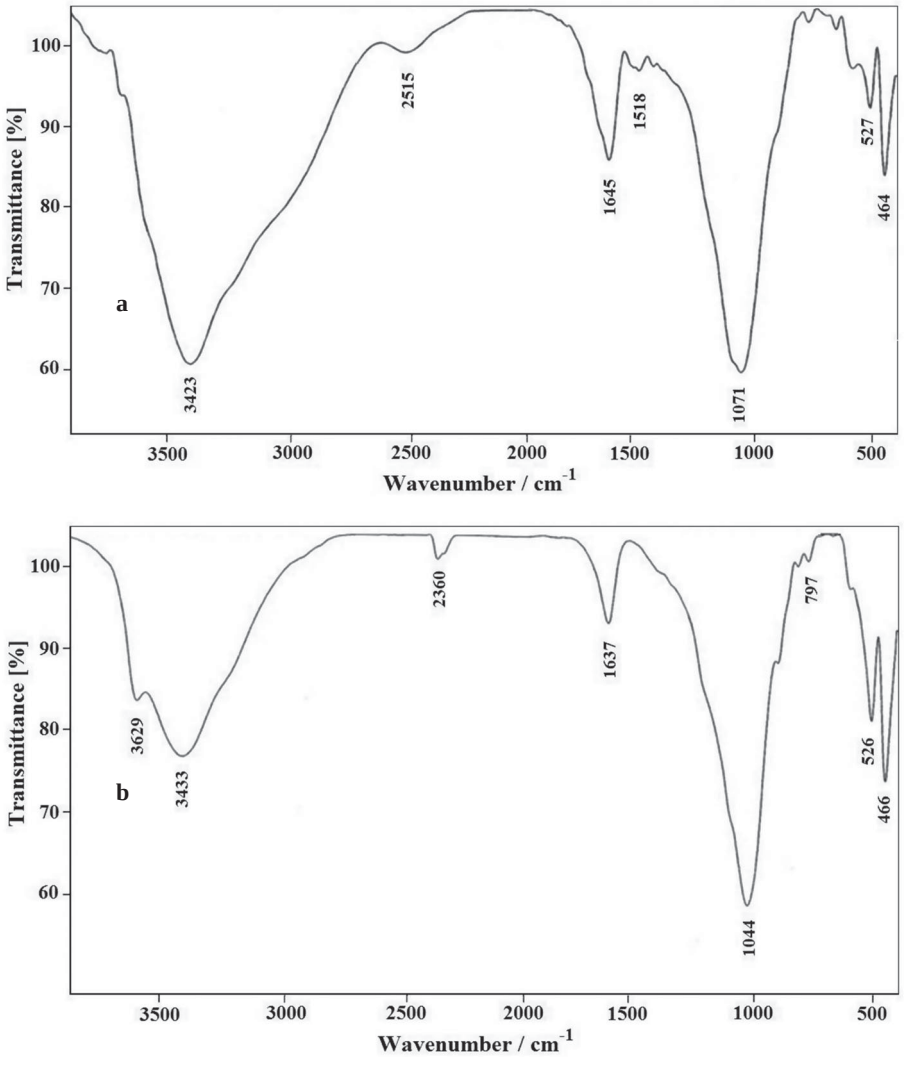


Figure 1. FT-IR spectra of KSF (a) and KSF@Ni (b).

The energy-dispersive X-ray (EDX) spectroscopy of KSF@Ni catalyst confirmed the presence of the expected elements in the structure of the catalyst (Figure 3). As expected, Si (66.26%) and Al (12.59%) are major elements that are the part of KSF and Ni (4.71%) and confirms incorporation or adsorption of Ni on KSF.

To find the appropriate reaction conditions for synthesis of products **4a–j** in the presence of KSF@Ni, the one-pot three-component reaction of benzaldehyde **1i**, thiosemicarbazide **2** and maleic anhydride **3** was selected as the model reaction. According to Table 1 entries 1–3, the model reaction preceded slowly under reflux condition in polar solvents, *e.g.* EtOH, DMF, and MeOH. However, the product was obtained in shorter reaction time in DMF (entry 2). The reaction occurred under microwave irradiation (70°C, 50 psi) in DMF and toluene separately and also in mixtures of these solvents (entries 4–8). For example, a mixture of DMF: toluene (1:1) led to the product in short reaction time (5 min) and high

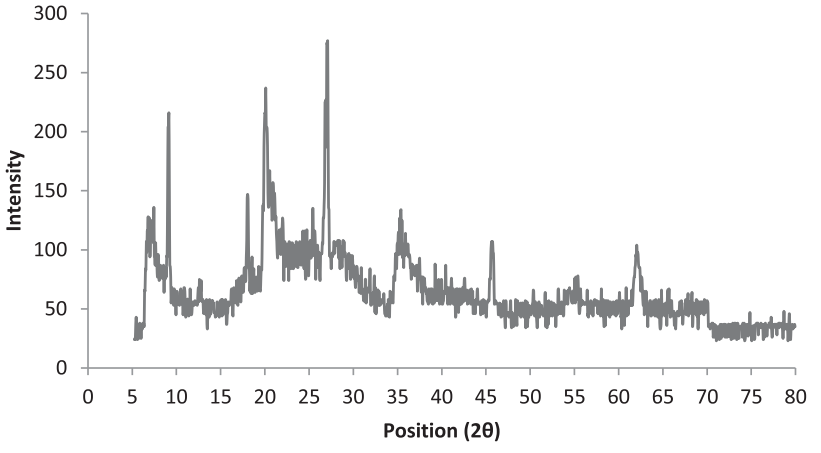


Figure 2. XRD spectra of KSF@Ni.

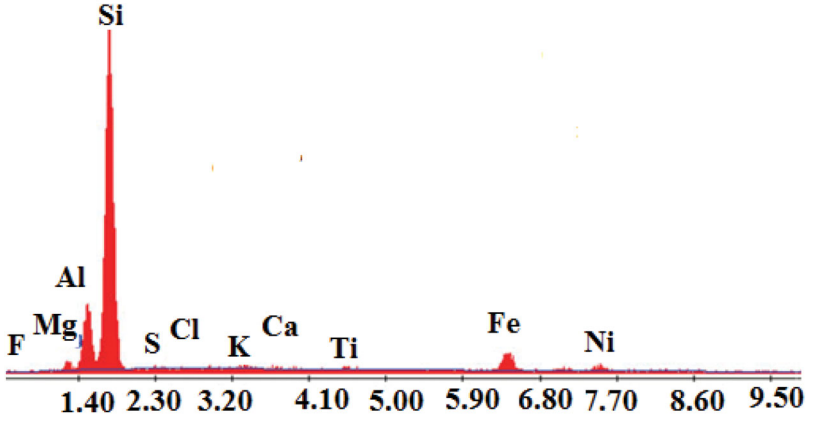


Figure 3. The EDX spectroscopy of the catalyst.

yield (60%) (entry 6). In another effort, the effect of the amount of the catalyst was investigated (entries 9–12) the results showed that 0.03 g of catalyst gave the product in reasonable yield and short reaction time (entry 6). Ultimately, the optimized reaction condition was obtained as shown in entry 6, Table 1.

In addition, the recyclability and reusability of KSF@Ni as catalyst for preparation of compound **4i** was investigated (Table 2). The catalytic activity of KSF@Ni was restored within the limit of catalyst amount for four runs. Furthermore, the amount of Ni in fresh and reused catalyst was obtained as 14.917 and 2.315 mg/L, respectively according to ICP-OES analysis.

Consequently, to investigate the generality of KSF@Ni as catalyst, different aldehydes **1a**–**j** in the synthesis of **4a**–**j** under optimized reaction condition were used (Table 3). As it can be seen from Table 2, all aldehydes bearing electron withdrawing and electron releasing groups reacted efficiently in short reaction times (3–5 min) and in high to good yields (60–97%).

Table 1. Optimization of the synthesis of **4i**.

Entry	Amount of catalyst (g)	Solvent	Condition	Time (min)	Yield (%)
1	0.03	EtOH	Reflux	420	40
2	0.03	DMF	Reflux	300	48
3	0.03	MeOH	Reflux	540	38
4	0.03	DMF	MW	12	50
5	0.03	Toluene	MW	15	47
6	**0.03**	**DMF: Toluene (1:1)**	**MW (70°C, 50 psi)**	**5**	**60**
7	0.03	DMF: Toluene (1:2)	MW (70°C, 50 psi)	6	52
8	0.03	DMF: Toluene (2:1)	MW (70°C, 50 psi)	5	56
9	0.01	DMF: Toluene (1:1)	MW (70°C, 50 psi)	10	48
10	0.02	DMF: Toluene (1:1)	MW (70°C, 50 psi)	8	50
11	0.04	DMF: Toluene (1:1)	MW (70°C, 50 psi)	8	48
12	–	DMF: Toluene (1:1)	MW (70°C, 50 psi)	50	35

Table 2. Recyclability of KSF@Ni.

Entry	Number of cycles	Time (min)	Yield (%)
1	1	7	56
2	2	10	51
3	3	10	46
4	4	14	40

Table 3. Synthesis of thiazolidinones **4a**–**j** under optimized reaction condition.

Product	R	Time (min)	Yield (%)	m.p. (°C)	m.p. (°C)
4a	2,4-diCl	3	74	304–306	This work
4b	2,6-diCl	4	80	259–260	This work
4c	4-Cl	3	74	273–275	273–274 [39]
4d	3-NO_2	3	95	279–281	This work
4e	2-NO_2	3	69	247–248	This work
4f	4-NO_2	4	97	268–271	270–271 [39]
4g	2-OH	5	90	289–290	This work
4h	4-OH	4	86	258–259	This work
4i	H	5	60	241–244	242–243 [39]
4j	4-OCH_3	3	70	261–263	262–263 [39]

This work is similar to reported studies [14,39] that used p-TSOH as the catalyst in the amounts of 0.2 and 0.01 mmol; however, in this one-pot 3MCRs recyclable KSF@Ni was used as the heterogeneous catalyst (0.03 g). In addition, in the previous studies the reaction time was reported as 5–12 and 10 min for the two steps with 33–82% and 33–76% yields, respectively. In the present study, the reaction times were reduced to 3–5 min with (60–97%) yields. Thus, KSF@Ni is an efficient catalyst for this procedure.

The structures of **4c**, **4f**, and **4i**–**j** were confirmed using FT-IR spectra and by comparison with literature melting points. The structures of the products **4a**–**b**, **4d**–**e**, and **4g**–**h** were characterized using FT-IR, ^{1}H NMR, and ^{13}C NMR spectra. The ^{1}H NMR spectral data were consistent with the expected structures as confirmed on the basis of their chemical shifts, multiplicities, and coupling constants. The N–H and CO_2H protons appeared together as a broad signal at 12.71–11.88 ppm. Proton of the imine group CH−N appeared as a sharp singlet at 8.67–8.28 ppm and also aromatic protons were observed in the expected chemical shift range of 8.59–6.83 ppm. Aliphatic protons of the CH–CH_2 moiety appeared with diastereomeric multiplicities as expected due to the chiral carbon,

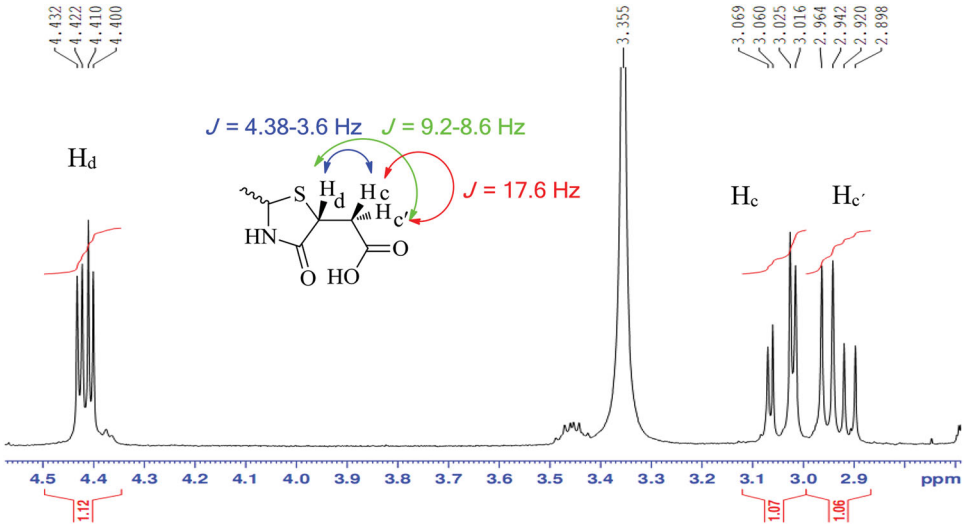


Figure 4. Diastereomeric coupling of CH–CH_2 moiety in products **4a**–**j**.

the proton of chiral carbon appeared as the doublet of doublet at 4.48–4.33 ppm and also diastereomeric protons appeared as the doublet of doublet at 3.09–2.85 ppm (Figure 4). The ^{13}C NMR spectra of **4a**–**b**, **4d**–**e**, and **4g**–**h** were in agreement with their structures. Carbons of amide and carboxylic acid appeared at the low field at 176.0–175.6 and 172.2 ppm, respectively. Carbon of CH−N and C−N groups appeared at 167.2–160.4 and 158.8–151.3 ppm, respectively. Also, aromatic carbons were observed at 162.9–116.1 ppm and aliphatic carbons were observed at 44.5–43.9 and 37.2–369 ppm.

2.2. Biology

In vitro antibacterial activity of compounds **4a**–**j** was screened against Gram-positive and Gram-negative bacterial strains including *Staphylococcus aureus* (*S. aureus*), *Micrococcus luteus* (*M. luteus*), *Escherichia coli* (*E. coli*), and *Pseudomonas aeruginosa* (*Ps. aeruginosa*) using the well-diffusion method. Tetracyline and Chloramphenicol were used as positive controls. DMSO was used as a negative control and showed no activity against mentioned bacterial strains. Also, the antibacterial activity of **4a**–**j** was screened at the concentration of 1000 µg/mL in DMSO. According to the results, none of the compounds inhibited *E. coli* and *Ps. aeruginosa*. However, *M. luteus* and *S. aureus* were sensitive to the compounds **4g**–**h** and **4b**, respectively.

3. Conclusion

In conclusion, an efficient, convenient, high-yield procedure with a simple set up and work up was reported for the one-pot three-component synthesis of thiazolidinones in the presence of KSF@Ni as catalyst under microwave irradiation. The prepared catalyst was confirmed using the FT-IR, XRD, ICP-OES and EDX analysis. Recyclability and reusability of the catalyst, simple procedure, the use of low volume of solvents and short reaction time are the prominent merits of this methodology.

4. Experimental

4.1. Material and instruments

Starting materials were obtained from Fluka and Merck. FT-IR spectra were measured with a Shimadzu IR-470 spectrophotometer. The ^{1}H NMR and ^{13}C NMR spectra were obtained on a Bruker Avance 400 and 100 MHz spectrometer, respectively. DMSO-d_6 was used as solvent. Melting points are uncorrected and were determined using MettlerFp5 apparatus. XRD was recorded on Brucker AXS-D8 Advance instrument. MW apparatus model 2016 latest microwave synthesis manufactured by Zhengzhou Keda Machinery and Instrument Equipment was used. KSF@Ni was prepared according to the literature using $NiCl_2$ instead of $ZnCl_2$ [40]. Metal content of catalyst was analyzed using the SPECTRO ARCOS ICP-OES analyzer. The chemical composition of catalyst was evaluated using Environmental Scanning electron microscope equipped with an EDX instrument (Philips XL30).

4.2. General procedure for synthesis of 4a–j

Aldehyde **1** (1 mmol), thiosemicarbazide **2** (1.1 mmol), maleic anhydride **3** (5 mmol), and KSF@Ni (0.03 g) were added to a mixture of DMF: Toluene 1:1 and was heated under microwave irradiation (70°C, 50 psi) for the appropriate time (Table 3). The progress of the reaction was monitored by TLC (*n*-hexane: EtOAc 4:3). After completion of the reaction water was added to the mixture and the precipitated solid was filtered off and dried at room temperature. The crude product was recrystallized from EtOH and the catalyst was separated at this stage.

Spectral data are given below.

4.2.1. {2-[(2,4-Dichlorobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl} acetic acid (4a)

White solid, Yield: 74%, m.p.: 304–306°C, FT-IR (ν, cm^{-1}): 3444 (stretch, OH), 3100, 1733, 1687, 1619, 1582, 1329, 1227, 1253, 1051, 878, 785. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.68 (br. s, 1H, H_a or H_f), 12.18 (br. s, 1H, H_f or H_a), 8.59 (s, 1H, H_h), 7.99 (d, J = 8.8 Hz, 1H, H_j), 7.77 (d, J = 2.0 Hz, 1H, H_m), 7.55 (dd, J = 10.2, 2.1 Hz, 1H, H_k), 4.41 (dd, J = 8.8, 4.0 Hz, 1H, H_d), 3.04 (dd, J_{syn} = 17.6, 3.6 Hz, 1H, H_c), 2.91 (dd, J_{anti} = 17.6, 8.8 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 175.9 (C_e), 172.2 (C_b), 167.0 (C_g), 151.3 (C_h), 136.2 (C_i), 134.7 (C_n), 130.7, 130.2, 129.2, 128.5 (C_k, C_l, C_j, C_m), 44.1 (C_d), 37.0 (C_c) ppm. HRMS ((+)-ESI): m/z = 344.9749 (calcd. 344.9742 for $C_{12}H_9Cl_2N_3O_3S$).

4.2.2. {2-[(2,6-Dichlorobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl} acetic acid (4b)

White solid, Yield: 80%, m.p.: 259–260°C, FT-IR (ν, cm^{-1}): 3443 (stretch, OH), 3018, 1722, 1636, 1584, 1332, 1220, 1258, 1034, 939, 848. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.58 (br. s, 1H, H_a or H_f), 12.27 (br. s, 1H, H_f or H_a), 8.56 (s, 1H, H_h), 7.59 (d, J = 8.0 Hz, 2H, H_k), 7.47 (t, J = 8.8 Hz, 1H, H_l), 4.39 (dd, J = 8.6, 3.8 Hz, 1H, H_d), 3.02 (dd, J_{syn} = 17.6, 4.0 Hz, 1H, H_c), 2.92 (dd, J_{anti} = 17.8, 8.6 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 176.0 (C_e), 172.2 (C_b), 167.2 (C_g), 152.1 (C_h), 134.5 (C_j), 131.9 (C_i), 130.4 (C_l), 129.7 (C_k), 44.1 (C_d), 36.9 (C_c) ppm. HRMS ((+)-ESI): m/z = 344.9738 (calcd. 344.9742 for $C_{12}H_9Cl_2N_3O_3S$).

4.2.3. {2-[(4-Chlorobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4c) [39]

White solid, Yield: 74%, m.p.: 273–275°C (obs. 273–274°C), FT-IR (ν, cm^{-1}): 3444, 3093, 1734, 1686, 1627, 1588, 1329, 1222, 1255, 1090, 824. HRMS ((+)-ESI): m/z = 311.0126 (calcd. 311.0131 for $C_{12}H_{10}ClN_3O_3S$).

4.2.4. {2-[(3-Nitrobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4d)

White solid, Yield: 95%, m.p.: 279–281°C, FT-IR (ν, cm^{-1}): 3435, 3033, 1721, 1638, 1592, 1529, 1346, 1226, 1261, 736, 674. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.71 (br. s, 1H, H_a or H_f), 12.22 (br. s, 1H, H_f or H_a), 8.59–8.58 (m, 2H, H_h, H_n), 8.31 (ddd, J = 8.2, 2.2 Hz, 1H, H_j), 7.21 (td, J = 6.8, 1.2 Hz, 1H, H_l), 7.77 (t, J = 7.8 Hz, 1H, H_k), 4.41 (dd, J = 8.6, 3.8 Hz, 1H, H_d), 3.05 (dd, J_{syn} = 17.6, 4.0 Hz, 1H, H_c), 2.93 (dd, J_{anti} = 17.8, 8.6 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 175.9 (C_e), 172.2 (C_b), 166.5 (C_g), 154.8 (C_h), 148.6 (C_m), 136.4 (C_j), 133.9 (C_i), 130.9 (C_k), 125.3 (C_l), 122.4 (C_n), 44.0 (C_d), 36.9 (C_c) ppm. HRMS ((+)-ESI): m/z = 322.0379 (calcd. 322.0372 for $C_{12}H_{10}N_4O_5S$).

4.2.5. {2-[(2-Nitrobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4e)

White solid, Yield: 69%, m.p.: 247–248°C, FT-IR (ν, cm^{-1}): 3445, 3101, 1734, 1686, 1621, 1581, 1529, 1347, 1248, 784, 747. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.74 (br. s, 1H, H_a or H_f), 12.16 (br. s, 1H, H_f or H_a), 8.67 (s, 1H, H_h), 8.08 (dd, J = 8.0, 1.2 Hz, 1H, H_n), 8.03 (dd, J = 7.6, 1.2 Hz, 1H, H_k), 7.83 (t, J = 7.4 Hz, 1H, H_m), 7.72 (dt, J = 7.7, 1.4 Hz, 1H, H_l), 4.41 (dd, J = 8.8, 4.0 Hz, 1H, H_d), 3.04 (dd, J_{syn} = 17.6, 4.0 Hz, 1H, H_c), 2.93 (dd, J_{anti} = 17.6, 8.8 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 175.9 (C_e), 172.2 (C_b), 167.0 (C_g), 152.7 (C_h), 148.7 (C_j), 134.0 (C_m), 131.6 (C_l), 129.4 (C_n), 128.8 (C_i), 125.1 (C_k), 44.1 (C_b), 36.9 (C_c) ppm. HRMS ((+)-ESI): m/z = 322.0367 (calcd. 322.0372 for $C_{12}H_{10}N_4O_5S$).

4.2.6. {2-[(4-Nitrobenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4f) [39]

White solid, Yield: 97%, m.p.: 268–271°C (obs. 270–271°C), FT-IR (ν, cm^{-1}): 3419, 3043, 1715, 1639, 1604, 1571, 1524, 1341, 1269, 847. HRMS ((+)-ESI): m/z = 322.0378 (calcd. 322.0372 for $C_{12}H_{10}N_4O_5S$).

4.2.7. {2-[(2-Hydroxybenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4g)

White solid, Yield: 90%, m.p.: 289–290°C, FT-IR (ν, cm^{-1}): 3443, 3051, 1731, 1636, 1336, 1215, 1268, 744. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.35 (br. s, 2H, H_a, H_f), 10.8 (s, 1H, H_k), 8.65 (s, 1H, H_h), 7.60 (dd, J = 8.0, 1.6 Hz, 1H, H_o), 7.35 (dt, J = 7.8, 1.7 Hz, 1H, H_m), 6.97–6.93 (m, 2H, H_l,H_n), 4.47 (dd, J = 8.6, 3.8 Hz, 1H, H_d), 3.06 (dd, J_{syn} = 17.6, 4.0 Hz, 1H, H_c), 2.95 (dd, J_{anti} = 17.6, 8.8 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 175.6 (C_e), 172.2 (C_b), 164.0 (C_g), 158.5 (C_h), 158.2 (C_j), 132.6 (C_m), 131.1 (C_o), 120.0 (C_n), 118.9 (C_i), 116.8 (C_l), 44.5 (C_d), 36.9 (C_c) ppm. HRMS ((+)-ESI): m/z = 279.0673 (calcd. 279.0678 for $C_{12}H_{13}N_3O_3S$).

4.2.8. {2-[(4-Hydroxybenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl}acetic acid (4h)

White solid, Yield: 86%, m.p.: 258–259°C, FT-IR (ν, cm^{-1}): 3211, 1691, 1636, 1513, 1336, 1166, 1242, 832. ^{1}H NMR (DMSO-d_6, 400 MHz) δ: 12.57 (br. s, 1H, H_a or H_f), 11.88 (br. s, 1H, H_f or H_a), 10.0 (s, 1H, H_m), 8.28 (s, 1H, H_h), 7.60 (d, J = 6.8 Hz, 1H, H_j), 6.84 (dd, J = 8.8 Hz, 1H, H_k), 4.35 (dd, J = 9.0, 3.8 Hz, 1H, H_d), 3.02 (dd, J_{syn} = 17.6, 4.4 Hz, 1H, H_c), 2.88 (dd, J_{anti} = 17.6, 9.2 Hz, 1H, $H_{c'}$) ppm. ^{13}C NMR (DMSO-d_6, 100 MHz) δ: 175.8 (C_e), 172.2 (C_b), 162.9 (C_l), 160.4 (C_g), 156.5 (C_h), 130.0 (C_j), 125.7 (C_i), 116.1 (C_k), 43.9 (C_d), 37.2 (C_c) ppm. HRMS ((+)-ESI): m/z = 293.0463 (calcd. 293.0470 for $C_{12}H_{11}N_3O_4S$).

4.2.9. [2-(Benzylidenehydrazinylidene)-4-oxo-1,3-thiazolidin-5-yl]acetic acid (4i) [39]

White solid, Yield: 60%, m.p.: 241–244°C (obs. 242–243°C), FT-IR (ν, cm^{-1}): 3422, 3033, 1712, 1640, 1335, 1255, 1261, 758, 689. m/z: 277.0521 Chemical Formula: $C_{12}H_{11}N_3O_3S$.

4.2.10. {2-[(4-Methoxybenzylidene)hydrazinylidene]-4-oxo-1,3-thiazolidin-5-yl} acetic acid (4j) [39]

White solid, Yield: 70%, m.p.: 261–263°C (obs. 262–263°C), FT-IR (ν, cm^{-1}): 3420, 3103, 2954, 2840, 1730, 1686, 1620, 1512, 1332, 1269, 1253, 829. HRMS ((+)-ESI): m/z = 293.0840 (calcd. 293.0834 for $C_{13}H_{15}N_3O_3S$).

4.3. Antibacterial assay

The antibacterial activity of products **4a**–**j** was investigated using a well-diffusion method. Nutrient agar and nutrient broth cultures were prepared according to manufactures instructions and were incubated at 37°C. The bacterial strains were prepared according to 0.5 McFarland standard. Thirty microliters of each bacterium was poured on each nutrient agar plate and then wells with 5 mm diameter were cuts into agar plates using sterilized glass tube. Each well received 30 µL of each compound (1000 compounds µg/ml in DMSO). Then, the plates were incubated at 37°C for 24 h and the zone of inhibition was measured and expressed in mm. Tetracycline and Chloramphenicol were used as positive controls and DMSO was used as negative control.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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