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ORIGINAL PAPER

Synthesis and antibacterial evaluation of novel chalcone derivatives containing a benzothiazole scaffold

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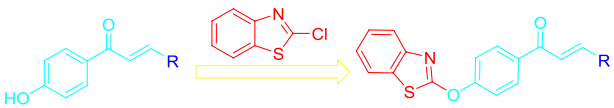
Abstract

A series of novel chalcone derivatives containing a benzothiazole scaffold were synthesized to develop novel antibacterial agents for solving the problem of drug resistance. Most compounds exhibited excellent antibacterial activity against *Xanthomonas oryzae pv. Oryzae* (Xoo), *Xanthomonas axonopodis pv. Citri* (Xac), and *Ralstonia solanacearum* (Rs) compared to a reference drug, bismerthiazol. The results indicated that chalcone derivatives containing a benzothiazole scaffold merit more research as promising antibacterial agents.

Graphical abstract



Keywords Synthesis · Chalcone · Benzothiazole · Antibacterial activity

Introduction

Plant pathogens *Xanthomonas oryzae pv. Oryzae* (Xoo), *Xanthomonas axonopodis pv. Citri* (Xac), and *Ralstonia solanacearum* (Rs) contribute to significant agriculture losses every year [1]. Although conventional agrochemicals have been effective, resistance develops quickly. Therefore, developing novel agrochemicals remains a huge challenge for pesticide scientists [2, 3].

Chalcone, a natural product, is widely used in a variety of drugs. Previous research studies have used chalcone derivatives for their bioactivities, antibacterial [4–6], antiviral [7–9], insecticidal [10], anticancer [11], antifungal [12], antiproliferative [13], and antioxidant [14] properties. Two series of chalcone derivatives based on sulfone and bisulfate have been demonstrated to have excellent antifungal activity against *C. albicans* and great antibacterial activity against *S. typhimurium* [15]. Some fluorinated chalcone–triazole hybrids have also noteworthy effects against bacteria and fungi [16].

Benzothiazole, a fused heterocycle, is a widely used pesticide due to its antibacterial [17, 18], antiviral [19], herbicidal [20], antifungal [21], antimicrobial [22–24], antitumour [25], antiproliferative [26], antitubercular [27], and anti-inflammatory [28] properties. Many pesticides are related to benzothiazole, such as the antiviral agent dufulin, benzthiazuron herbicide, tribunal, and mefenacet. In addition, one hybrid containing 2-aminobenzothiazole and substituted aryl azides showed twice the antibacterial activity

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against all Gram-positive bacteria and Gram-negative bacteria strains compared to ciprofloxacin [29]. The bioassay results exerted that the types of five member heterocyclic compounds bearing a benzothiazole moiety showed better antibacterial activity, compared to that containing an oxazole moiety [30].

Chalcone and benzothiazole possess many biological properties and are widely used in drug development. However, there are few reports on the combination of both chalcone and benzothiazole and its effects on antibacterial activity. Therefore, in this paper, we introduced a benzothiazole group into chalcone and investigated the antibacterial activity of the new hybrids against Xoo, Xac, and Rs to identify novel and high-efficiency antibacterial drugs.

Results and discussion

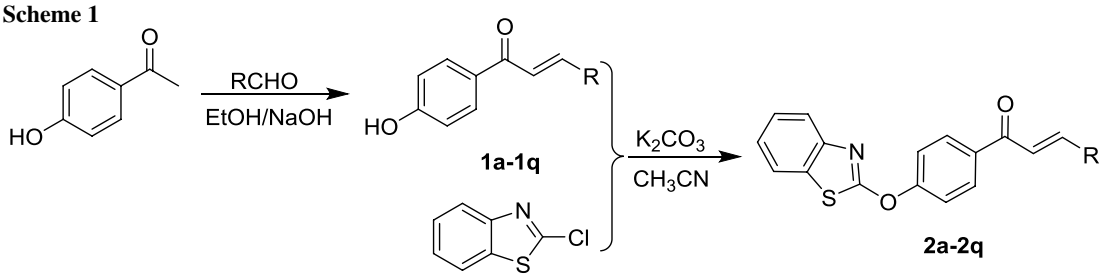
A description of the synthesis route is listed in Scheme 1. Based on previously published methods [31] for 4-hydroxyacetophenone, aldosterone condensation was performed in an ice bath under alkali conditions to prepare compound **1** with various substitutions of formaldehyde, 4-chlorobenzaldehyde, 4-methylbenzaldehyde, benzaldehyde, 4-nitrobenzaldehyde, and 2-pyridylaldehyde. Then, etherification between 2-chlorobenzothiazole and compound **1** to produce compound **2** was performed under acetonitrile as a solvent, potassium carbonate as a catalyser and refluxing at 85 °C. We attempted to use triethylamine and dimethyl formamide for compound **2**; however, the catalysts produced a low yield and long reaction time compared to potassium carbonate. As shown in Table 1, the title compounds were obtained with yields of 49–82%.

All structural characterizations of the new compounds were performed via ^{1}H NMR, ^{13}C NMR, and HRMS, and all the signals in the spectra are in agreement with the proposed structure. The ^{1}H NMR spectra of compounds **1c** and **1o** appear two distinct absorption peak at nearly 8.16 and 7.64 ppm, which contribute to the CO–CH and CO–C=CH, respectively. In the ^{13}C NMR spectra of compound **1c**, the strong absorption peak at 131.96 and 115.96 ppm are attributed to the Ar(4-OH), and the strong absorption peak at 150.78 and 122.93 ppm are attributed to the pyridine ring. For the compound **1o**, approximate characteristic peak values of 131.93 and 115.99 ppm are attributed to the Ar(4-OH), and the peak values range from 135.18 to 126.28 ppm contribute to the naphthalene nucleus. The ^{13}C NMR spectra of all title compounds exhibit two special lines at approximately 188.37 and 171.21 ppm, which are attributed to the S–C=N and C=O group, respectively. In the ^{1}H NMR spectra, all the title compounds possess two sets of peaks at approximately 7.48–7.44 ppm and 7.39–7.35 ppm, which

Table 1 Physical properties of compounds **2a**–**2q**

Compound	Formula	Appearance	Yield/%	M.p./°C
1c	$C_{14}H_{11}NO_2$	White solid	89	> 320
1o	$C_{19}H_{13}BrO_2$	White solid	79	244–245
2a	$C_{21}H_{14}N_2O_2S$	White solid	80	144–146
2b	$C_{22}H_{14}ClNO_2S$	White solid	68	147–149
2c	$C_{21}H_{14}N_2O_2S$	White solid	78	253–255
2d	$C_{23}H_{17}NO_2S$	White solid	68	99–101
2e	$C_{23}H_{17}NO_2S$	White solid	51	103–105
2f	$C_{25}H_{21}NO_2S$	White solid	58	93–95
2g	$C_{22}H_{15}NO_2S$	White solid	82	133–135
2h	$C_{24}H_{19}NO_4S$	Yellow solid	52	115–117
2i	$C_{20}H_{13}NO_2S_2$	White solid	75	143–145
2j	$C_{23}H_{17}NO_3S$	White solid	64	111–113
2k	$C_{23}H_{17}NO_3S$	White solid	51	124–126
2l	$C_{20}H_{13}NO_3S$	Yellow solid	68	112–114
2m	$C_{22}H_{14}N_2O_4S$	White solid	74	182–184
2n	$C_{22}H_{14}N_2O_4S$	Yellow solid	72	156–158
2o	$C_{26}H_{16}BrNO_2S$	White solid	58	> 320
2p	$C_{23}H_{14}F_3NO_2S$	White solid	49	142–144
2q	$C_{24}H_{19}NO_2S$	White solid	76	123–125

Scheme 1



2a: R = Pyridin-2-yl **2b**: R = 4-ClPh **2c**: R = Pyridin-4-yl **2d**: R = 4-MePh
2e: R = 3-MePh **2f**: R = 4-*i*PrPh **2g**: R = Ph **2h**: R = 2,4-di-OMePh
2i: R = Thiophen-2-yl **2j**: R = 2-OMePh **2k**: R = 4-OMePh **2l**: R = Furan-2-yl
2m: R = 3-NO_2Ph **2n**: R = 4-NO_2Ph **2o**: R = 1-Br-2-Naphthalene **2p**: R = 4-CF_3Ph
2q: R = 3,4-di-MePh

confirm the benzothiazole ring structure. The strong presence of $[M+H]^+$ ions indicates that the title compounds are in steady state.

Antibacterial activity

Xoo (strain PXO99A, Nangjing Agricultural University, China), Xac (strain 29-1, Shanghai Jiao Tong University, China), and Rs (strain MR111, Guizhou University, China) were used as test strains, commercial antibacterial agent bismerthiazol was used as positive control, and DMSO was used as a blank control. To determine the EC_{50} values of the compounds, the antibacterial activity against Xoo, Xac, and Rs was assessed by testing at five concentrations (100, 50, 25, 12.5, and 6.25 μg/cm^3).

As shown in Tables 2 and 3, more title compounds exhibited substantial antibacterial activity against Xoo, and the fewest compounds exhibited excellent antibacterial activity against Rs. The bioassay data analysis also indicated that the antibacterial activity of the title compounds was dependent on the R fragment.

For example, when R was substituted by an electron-withdrawing group (4-ClPh, 3-NO_2Ph, 4-NO_2Ph, and 4-CF_3Ph), corresponding compounds **2b**, **2m**, **2n**, and **2p** exhibited antibacterial activity against Xoo, with EC_{50} values of 48.66, 38.97, 46.64, and 65.65 μg/cm^3, respectively, which exceeded that of bismerthiazol (72.75 μg/cm^3). Compounds **2b** and **2p** also showed remarkable antibacterial activity against Xac, with EC_{50} values of 35.59 and 42.56 μg/cm^3, respectively, which exceeded that of bismerthiazol (54.58 μg/cm^3).

Table 3 EC_{50} values of target compounds against Xoo, Xac, and Rs

Compound	R	EC_{50}/μg/cm^3		
		Xoo	Xac	Rs
2a	Pyridin-2-yl	50.37	54.24	88.70
2b	4-ClPh	48.66	35.59	
2c	Pyridin-4-yl	52.40	50.97	36.49
2h	2,4-di-OMePh	65.48	–	64.64
2l	Furan-2-yl	–	13.42	
2m	3-NO_2Ph	38.97	–	
2n	4-NO_2Ph	46.64	–	
2p	4-CF_3Ph	65.65	42.56	
2q	3,4-di-MePh	66.74	–	
Bismerthiazol	–	72.75	54.58	97.18

Meanwhile, compounds **2h** and **2q** containing two electron groups (2,4-di-OMePh, 3,4-di-MePh) presented superior antibacterial activity. Compound **2h** exhibited antibacterial activity against Xoo and Rs, with EC_{50} values of 65.48 and 64.64 μg/cm^3, respectively, which were better than that of bismerthiazol (72.75 and 97.18 μg/cm^3, respectively). Compound **2q** against Xoo with an EC_{50} value of 66.74 μg/cm^3, which outperformed that of bismerthiazol (72.75 μg/cm^3).

Pyridine fragments resulting in compounds **2a** and **2c** with a broad spectrum and efficient antibacterial activity against Xoo with EC_{50} values of 50.37 and 52.40 μg/

Table 2 Inhibition (%) effect of the compounds against Xoo, Xac, and Rs

Compound	Xoo		Xac		Rs	
	100 μg/cm^3	50 μg/cm^3	100 μg/cm^3	50 μg/cm^3	100 μg/cm^3	50 μg/cm^3
2a	70	46	71	56	56	45
2b	74	44	84	57	14	11
2c	76	41	72	48	72	28
2d	45	29	56	45	11	9
2e	48	21	36	20	18	4
2f	54	29	59	31	21	0
2g	37	20	31	14	4	2
2h	62	40	29	24	77	29
2i	31	24	27	11	7	4
2j	53	43	34	21	27	11
2k	52	43	35	16	36	30
2l	49	33	100	72	18	6
2m	76	51	29	13	48	41
2n	71	54	60	34	34	15
2o	54	36	34	21	37	33
2p	68	33	74	50	24	20
2q	67	34	56	45	25	23
Bismerthiazol	59	39	70	49	52	28

cm^3, respectively, which exceeded that of bismerthiazol (72.75 μg/cm^3). The activity against Xac resulted in EC_{50} values of 54.24 and 50.97 μg/cm^3, respectively, which outperformed that of bismerthiazol (54.58 μg/cm^3). The activity against Rs resulted in EC_{50} values of 88.70 and 36.49 μg/cm^3, respectively, which were better than that of bismerthiazol (97.18 μg/cm^3).

Furthermore, compound **2l** contained a furan moiety and exhibited antibacterial activity against Xac, with an EC_{50} value of 13.42 μg/cm^3, which far surpassed that of bismerthiazol (54.58 μg/cm^3).

Interestingly, among all the title compounds, **2m**, **2l**, and **2c** exhibited the strongest antibacterial activity against Xoo, Xac, and Rs, respectively, with EC_{50} values of 38.97, 13.42, and 36.49 μg/cm^3, respectively, which far surpassed that of bismerthiazol (72.75, 54.58, and 97.18 μg/cm^3, respectively).

Conclusion

We report a series of novel chalcone derivatives that were structurally confirmed via ^{1}H NMR, ^{13}C NMR, and HRMS and evaluated for antibacterial activity against Xoo, Xac, and Rs in vitro. Antibacterial screening results demonstrated that compounds **2c**, **2l**, and **2m** exhibited the strongest antibacterial activity among all the title compounds. Based on this study, chalcone derivatives containing benzothiazolyl deserve further research as a potential agricultural fungicide.

Experimental

A Bruker ASCEND 400 NMR spectrometer (BRUKER OPTICS, Switzerland) was used to record the nuclear magnetic resonance (NMR) spectra with tetramethylsilane (TMS) as the internal standard and dimethylsulfoxide (DMSO-d_6, 2.50 ppm (^{1}H), 39.50 ppm (^{13}C)) or deuterochloroform ($CDCl_3$, 7.26 ppm (^{1}H), 76.00 ppm (^{13}C)) as the solvent. The melting point tests were conducted in an XT-4 binocular microscope (Beijing Tech Instrument Co). The measurements of all experiments were completed on a Sartorius electronic balance (Sartorius Group, Germany). A QY-20 three UV analyser (Shanghai Anting Electronic Instrument Factory) was used for thin layer chromatography (TLC). 2-Chlorobenzothiazole, 4-hydroxyacetophenone, all solvents and reagents were purchased from Shanghai Titan Scientific Co., Ltd and were analytical grade or chemically pure.

General procedure for the synthesis of compounds 1a–1q

According to the synthetic method of the literature [31], based on 4-hydroxyacetophenone and various substituted formaldehyde, compounds **1** were prepared by the method of aldosterone condensation, under the conditions both alkaline and ice bath. All of them are known compounds except **1c** and **1o**, their physical and chemical properties are listed in the electronic supplementary material.

(*E*)-1-(4-Hydroxyphenyl)-3-(pyridin-4-yl)prop-2-en-1-one (1c, $C_{14}H_{11}NO_2$) White solid; yield: 89%; m.p.: > 320 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.67 (d, J = 4.9 Hz, 2H, pyridine-3, 5-H), 8.16 (d, J = 16.1 Hz, 1H, CO–C = CH), 8.12 (d, J = 8.0 Hz, 2H, Ar(4-OH)-2, 6-H), 7.84 (d, J = 5.1 Hz, 2H, pyridine-2, 6-H), 7.64 (d, J = 15.6 Hz, 1H, CO–CH), 6.93 (d, J = 8.0 Hz, 2H, Ar(4-OH)-3, 5-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): δ = 187.37, 163.08, 150.78, 142.49, 140.30, 131.96, 131.19, 129.10, 126.88, 122.93, 115.96, 115.60 ppm; HRMS (ESI): *m/z* (calcd.) $[M+H]^+$ 226.0862, found 226.0858.

(*E*)-3-(1-Bromonaphthalen-2-yl)-1-(4-hydroxyphenyl)-prop-2-en-1-one (1o, $C_{19}H_{13}BrO_2$) White solid; yield: 79%; m.p.: 244–245 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.34–8.26 (m, 3H, CO–C=CH, naphthalene-3, 5-H), 8.16 (d, J = 8.8 Hz, 2H, Ar(4-OH)-2, 6-H), 8.11–8.03 (m, 3H, naphthalene-6, 7, 8-H), 7.76–7.66 (m, 2H, CO–CH, naphthalene-4-H), 6.95 (d, J = 8.7 Hz, 2H, Ar(4-OH)-3, 5-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): δ = 187.31, 163.09, 141.50, 135.18, 132.84, 132.29, 131.93, 129.29, 128.96, 128.73, 128.52, 127.95, 126.83, 126.28, 125.29, 115.99 ppm; HRMS (ESI): *m/z* (calcd.) $[M+H]^+$ 353.0171, found 353.0159.

General procedure for the synthesis of title compounds 2a–2q

Intermediates **1a** (1.0 mmol) and 2-chlorobenzothiazole (1.1 mmol) were added to a round-bottomed flask with K_2CO_3 (1.5 mmol) and 20 cm^3 acetonitrile as a solvent and stirred at 85 °C for 3–5 h. After the reaction was complete, the system was filtered with alcohol as a flushing fluid. A sufficient amount of water was then added to the filtrate, and liquid was separated under constantly stirring, suction filtration, drying, and recrystallization from petroleum ether to obtain the title compound **2a**. Compounds **2b**–**2q** were prepared in the same way.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(pyridin-2-yl)-prop-2-en-1-one (2a, $C_{21}H_{14}N_2O_2S$) White solid; yield: 80%; m.p.: 144–146 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.72

(t, $J = 5.2$ Hz, 1H, pyridine-3-H), 8.31–8.24 (m, 2H, Ph-2, 6-H), 8.20 (dd, $J = 15.5$, 6.2 Hz, 1H, benzothiazole-4-H), 8.01 (dd, $J = 13.8$, 6.4 Hz, 1H, benzothiazole-7-H), 7.93 (dd, $J = 15.5$, 6.9 Hz, 2H, CO–CH, CO–C=CH), 7.81–7.65 (m, 4H, benzothiazole-5, 6-H, pyridine-5, 6-H), 7.51–7.43 (m, 2H, Ph-3, 5-H), 7.39 (dd, $J = 14.1$, 7.3 Hz, 1H, pyridine-4-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.78$, 171.12, 158.16, 153.20, 150.55, 148.77, 143.80, 137.72, 135.64, 132.47, 131.40, 127.11, 125.51, 125.41, 125.06, 122.87, 121.90, 121.47 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 359.0848, found 359.0841.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(4-chlorophenyl)-prop-2-en-1-one (2b, $C_{22}H_{14}ClNO_2S$) White solid; yield: 47%; m.p.: 143–145 °C; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 8.33$ (d, $J = 8.6$ Hz, 2H, Ph-2, 6-H), 8.06–7.94 (m, 4H, Ar(4-Cl)-2, 6-H, benzothiazole-4-H, CO–C=CH), 7.76 (dd, $J = 19.1$, 11.8 Hz, 2H, benzothiazole-7-H, CO–CH), 7.67 (d, $J = 8.6$ Hz, 2H, Ar(4-Cl)-3, 5-H), 7.55 (d, $J = 8.3$ Hz, 2H, Ph-3,5-H), 7.48–7.44 (m, 1H, benzothiazole-5-H), 7.39–7.35 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.31$, 171.17, 158.09, 148.79, 143.28, 135.81, 135.68, 134.11, 132.45, 131.43, 131.17, 129.46, 127.11, 125.03, 123.11, 122.86, 121.88, 121.40 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 392.0506, found 392.0497.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(pyridin-4-yl)-prop-2-en-1-one (2c, $C_{21}H_{14}N_2O_2S$) White solid; yield: 78%; m.p.: 253–255 °C; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 8.70$ (d, $J = 6.0$ Hz, 2H, pyridine-3, 5-H), 8.38–8.33 (m, 2H, Ph-2, 6-H), 8.23 (d, $J = 15.7$ Hz, 1H, CO–C=CH), 8.01 (dd, $J = 8.0$, 0.8 Hz, 1H, benzothiazole-4-H), 7.88 (dd, $J = 4.6$, 1.4 Hz, 2H, pyridine-2, 6-H), 7.88 (dd, $J = 4.6$, 1.4 Hz, 2H, CO–CH, benzothiazole-7-H), 7.76–7.71 (m, 2H, Ph-3, 5-H), 7.49–7.44 (m, 1H, benzothiazole-5-H), 7.40–7.36 (m, 1.2 Hz, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.37$, 171.14, 158.28, 150.87, 148.76, 142.21, 141.78, 135.41, 132.46, 131.62, 127.12, 126.66, 125.05, 123.06, 122.88, 121.89, 121.50 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 359.0848, found 359.0841.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(*p*-tolyl)-prop-2-en-1-one (2d, $C_{23}H_{17}NO_2S$) White solid; yield: 68%; m.p.: 99–101 °C; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 8.32$ (d, $J = 8.8$ Hz, 2H, Ph-2, 6-H), 8.00 (dd, $J = 8.0$, 0.7 Hz, 1H, benzothiazole-4-H), 7.95 (d, $J = 15.6$ Hz, 1H, CO–C=CH), 7.82 (d, $J = 8.1$ Hz, 2H, Ar(4-CH_3)-2, 6-H), 7.79–7.72 (m, 2H, CO–CH, benzothiazole-7-H), 7.67 (d, $J = 8.8$ Hz, 2H, Ar(4-CH_3)-3, 5-H), 7.49–7.42 (m, 1H, benzothiazole-5-H), 7.40–7.35 (m, 1H, benzothiazole-6-H), 7.30 (d, $J = 8.0$ Hz, 2H, Ph-3, 5-H), 2.37 (s, 3H, CH_3) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.37$, 171.21, 157.96, 148.80, 144.86, 141.33, 136.04, 132.45, 132.41, 131.32, 130.05, 129.51, 127.10, 125.02, 122.85, 121.87, 121.37, 121.30, 21.59 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 372.1052, found 372.1047.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(*m*-tolyl)-prop-2-en-1-one (2e, $C_{23}H_{17}NO_2S$) White solid; yield: 51%; m.p.: 103–105 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.16$–8.12 (m, 2H, Ph-2, 6-H), 7.82 (d, $J = 15.7$ Hz, 1H, CO–C=CH), 7.77 (dd, $J = 8.1$, 0.5 Hz, 1H, benzothiazole-4-H), 7.72 (dd, $J = 8.0$, 0.7 Hz, 1H, benzothiazole-7-H), 7.55–7.50 (m, 3H, Ph-3, 5-H, CO–CH), 7.46 (d, $J = 7.1$ Hz, 2H, Ar(3-CH_3)-4, 6-H), 7.44–7.40 (m, 1H, benzothiazole-5-H), 7.35–7.29 (m, 3H, benzothiazole-6-H, Ar(3-CH_3)-2, 5-H), 2.41 (s, 3H, CH_3) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 189.09$, 170.58, 157.85, 148.83, 145.42, 138.71, 135.82, 134.75, 132.41, 131.59, 130.65, 129.12, 128.92, 126.46, 125.81, 124.51, 121.99, 121.49, 121.43, 120.36, 21.38 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 372.1052, found 372.1046.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(4-isopropylphenyl)prop-2-en-1-one (2f, $C_{25}H_{21}NO_2S$) White solid; yield: 58%; m.p.: 93–95 °C; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 8.34$–8.29 (m, 2H, Ph-2, 6-H), 8.00 (dd, $J = 8.0$, 0.8 Hz, 1H, benzothiazole-4-H), 7.95 (d, $J = 15.6$ Hz, 1H, CO–C=CH), 7.84 (d, $J = 8.2$ Hz, 2H, Ar(4-*i*Pr)-2, 6-H), 7.80–7.71 (m, 2H, benzothiazole-7-H, CO–CH), 7.69–7.64 (m, 2H, Ar(4-*i*Pr)-3, 5-H), 7.50–7.42 (m, 1H, benzothiazole-5-H), 7.42–7.32 (m, 3H, Ph-3, 5-H, benzothiazole-6-H), 3.00–2.89 (m, 1H, CH), 1.24 (s, 3H, CH_3), 1.22 (s, 3H, CH_3) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.40$, 171.21, 157.96, 152.07, 148.80, 144.86, 136.05, 132.83, 132.45, 131.31, 129.63, 127.41, 127.10, 125.02, 122.85, 121.87, 121.42, 121.37, 33.93, 24.09 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 400.1365, found 400.1357.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-phenylprop-2-en-1-one (2g, $C_{22}H_{15}NO_2S$) White solid; yield: 82%; m.p.: 133–135 °C; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 8.35$–8.31 (m, 2H, Ph-2, 6-H), 8.05 (d, $J = 15.6$ Hz, 1H, CO–C=CH), 8.00 (dd, $J = 8.0$, 0.7 Hz, 1H, benzothiazole-4-H), 7.89 (d, $J = 8.5$ Hz, 2H, Ar-2, 6-H), 7.75 (t, $J = 11.6$ Hz, 2H, CO–CH, benzothiazole-7-H), 7.70–7.66 (m, 4H, Ph-3, 5-H, Ar-3, 5-H), 7.48–7.44 (m, 1H, benzothiazole-5-H), 7.40–7.35 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 188.32$, 171.17, 158.09, 148.79, 143.37, 135.80, 134.43, 132.45, 132.38, 131.43, 131.36, 127.10, 125.03, 124.59, 123.15, 122.86, 121.88, 121.40 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 380.0715, found 380.0700.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(2,4-dimethoxyphenyl)prop-2-en-1-one (2h, $C_{24}H_{19}NO_4S$) Yellow solid; yield: 52%; m.p.: 115–117 °C; ^{1}H NMR (400 MHz,

DMSO-d_6): $\delta = 8.52–8.48$ (m, 2H, Ph-2, 6-H), 8.37 (d, $J = 15.6$ Hz, 1H, CO–C=CH), 8.18–8.14 (m, 1H, benzothiazole-4-H), 8.08 (d, $J = 8.6$ Hz, 1H, Ar(2, 4-di-OCH_3)-6-H), 8.04 (d, $J = 15.7$ Hz, 1H, CO–CH), 7.95–7.92 (m, 1H, benzothiazole-7-H), 7.88–7.84 (m, 2H, Ph-3, 5-H), 7.72–7.67 (m, 1H, benzothiazole-5-H), 7.64–7.56 (m, 1H, benzothiazole-6-H), 6.90 (d, $J = 2.3$ Hz, 1H, Ar(2, 4-di-OCH_3)-3-H), 6.86 (dd, $J = 8.7$, 2.3 Hz, 1H, Ar(2, 4-di-OCH_3)-5-H), 4.20 (s, 3H, OCH_3), 4.12 (s, 3H, OCH_3) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): $\delta = 198.56$, 181.29, 174.17, 171.10, 168.24, 159.55, 150.13, 147.08, 142.98, 141.29, 140.96, 140.91, 137.06, 135.07, 132.44, 132.12, 131.13, 129.59, 127.08, 125.76, 116.72, 108.67, 65.85, 65.61 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 418.1107, found 418.1101.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(thiophen-2-yl)-prop-2-en-1-one (2i, $C_{20}H_{13}NO_2S_2$) White solid; yield: 75%; m.p.: 143–145 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.15–8.10$ (m, 2H, Ph-2, 6-H), 7.98 (d, $J = 15.3$ Hz, 1H, CO–C=CH), 7.77 (d, $J = 7.7$ Hz, 1H, thiophene-3-H), 7.72 (dd, $J = 8.0$, 0.6 Hz, 1H, thiophene-5-H), 7.55–7.51 (m, 2H, Ph-3, 5-H), 7.45–7.29 (m, 5H, CO–CH, benzothiazole-4, 5, 6, 7-H), 7.11 (dd, $J = 5.0$, 3.7 Hz, 1H, thiophene-4-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 188.50$, 170.66, 157.94, 148.90, 140.40, 137.65, 135.76, 132.48, 132.43, 130.63, 129.12, 128.53, 126.54, 124.59, 122.07, 121.51, 120.45 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 364.0452, found 364.0460.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(2-methoxyphenyl)prop-2-en-1-one (2j, $C_{23}H_{17}NO_3S$) White solid; yield: 64%; m.p.: 111–113 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.17–8.11$ (m, 3H, Ph-2, 6-H, CO–C=CH), 7.76 (d, $J = 8.1$ Hz, 1H, benzothiazole-4-H), 7.73–7.70 (m, 1H, benzothiazole-7-H), 7.66–7.61 (m, 2H, CO–CH, Ar(2-OCH_3)-6-H), 7.54–7.49 (m, 2H, Ph-3, 5-H), 7.44–7.36 (m, 2H, benzothiazole-5-H, Ar(2-OCH_3)-4-H), 7.33–7.28 (m, 1H, benzothiazole-6-H), 7.00 (t, $J = 7.5$ Hz, 1H, Ar(2-OCH_3)-5-H), 6.95 (d, $J = 8.3$ Hz, 1H, Ar(2-OCH_3)-3-H), 3.92 (s, 3H, OCH_3) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 189.67$, 170.67, 158.91, 157.71, 148.85, 140.81, 136.12, 132.41, 131.94, 130.67, 129.45, 126.45, 124.49, 123.82, 122.53, 121.97, 121.44, 120.81, 120.29, 111.28, 55.59 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 388.1001, found 388.0990.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(4-methoxyphenyl)prop-2-en-1-one (2k, $C_{23}H_{17}NO_3S$) White solid; yield: 51%; m.p.: 124–126 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.16–8.10$ (m, 2H, Ph-2, 6-H), 7.82 (d, $J = 15.6$ Hz, 1H, CO–C=CH), 7.79–7.75 (m, 1H, benzothiazole-4-H), 7.72 (dd, $J = 8.0$, 0.6 Hz, 1H, benzothiazole-7-H), 7.62 (d, $J = 8.7$ Hz, 2H, Ar(4-OCH_3)-2, 6-H), 7.55–7.50 (m, 2H, Ar(4-OCH_3)-3, 5-H), 7.45–7.40 (m, 2H, CO–CH, benzothiazole-5-H), 7.35–7.29 (m, 1H, benzothiazole-6-H), 6.95 (d, $J = 8.8$ Hz, 2H, Ph-3, 5-H), 3.87 (s, 3H, OCH_3) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 189.10$, 170.65, 161.82, 157.71, 148.84, 145.08, 136.09, 132.41, 130.55, 130.34, 127.53, 126.45, 124.49, 121.98, 121.43, 120.32, 119.36, 114.49, 55.46 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 388.1001, found 388.0994.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(furan-2-yl)-prop-2-en-1-one (2l, $C_{20}H_{13}NO_3S$) Yellow solid; yield: 68%; m.p.: 112–114 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.17–8.12$ (m, 2H, Ph-2, 6-H), 7.76 (d, $J = 8.1$ Hz, 1H, benzothiazole-4-H), 7.72 (dd, $J = 8.0$, 0.6 Hz, 1H, benzothiazole-7-H), 7.63 (d, $J = 15.3$ Hz, 1H, CO–C=CH), 7.56–7.48 (m, 4H, Ph-3, 5-H, CO–CH, furan-3-H), 7.44–7.39 (m, 1H, benzothiazole-5-H), 7.33–7.29 (m, 1H, benzothiazole-6-H), 6.75 (d, $J = 3.4$ Hz, 1H, furan-5-H), 6.56–6.50 (m, 1H, furan-4-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 188.35$, 170.59, 157.86, 151.62, 148.83, 145.09, 135.73, 132.41, 130.97, 130.57, 126.45, 124.50, 121.99, 121.43, 120.35, 118.89, 116.58, 112.78 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 364.0460, found 364.0452.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(3-nitrophenyl)-prop-2-en-1-one (2m, $C_{22}H_{14}N_2O_4S$) White solid; yield: 74%; m.p.: 182–184 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.53$ (s, 1H, Ar(3-NO_2)-2-H), 8.28 (dd, $J = 8.2$, 1.2 Hz, 1H, benzothiazole-4-H), 8.17 (d, $J = 8.8$ Hz, 2H, Ph-2, 6-H), 7.94 (d, $J = 7.6$ Hz, 1H, Ar(3-NO_2)-4-H), 7.87 (d, $J = 15.7$ Hz, 1H, CO–C=CH), 7.75 (dd, $J = 13.5$, 8.0 Hz, 2H, benzothiazole-7-H, Ar(3-NO_2)-6-H), 7.69–7.61 (m, 2H, CO–CH, Ar(3-NO_2)-5-H), 7.58 (d, $J = 8.8$ Hz, 2H, Ph-3, 5-H), 7.45–7.41 (m, 1H, benzothiazole-5-H), 7.35–7.31 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 188.18$, 170.38, 158.24, 148.77, 141.97, 136.57, 135.05, 134.40, 132.41, 130.78, 130.12, 126.50, 124.80, 124.59, 124.25, 122.40, 122.02, 121.45, 120.50 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 403.0747, found 403.0738.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(4-nitrophenyl)-prop-2-en-1-one (2n, $C_{22}H_{14}N_2O_4S$) Yellow solid; yield: 72%; m.p.: 156–158 °C; ^{1}H NMR (400 MHz, $CDCl_3$): $\delta = 8.29$ (d, $J = 8.8$ Hz, 2H, Ar(4-NO_2)-3, 5-H), 8.15 (d, $J = 8.8$ Hz, 2H, Ar(4-NO_2)-2, 6-H), 7.85 (d, $J = 15.7$ Hz, 1H, CO–C=CH), 7.80 (d, $J = 8.7$ Hz, 2H, Ph-2, 6-H), 7.78–7.72 (m, 2H, benzothiazole-4, 7-H), 7.64 (d, $J = 15.7$ Hz, 1H, CO–CH), 7.57 (d, $J = 8.8$ Hz, 2H, Ph-3, 5-H), 7.45–7.41 (m, 1H, benzothiazole-5-H), 7.35–7.31 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): $\delta = 188.20$, 170.39, 158.26, 148.71, 148.64, 141.86, 140.93, 134.98, 132.39, 130.78, 129.02, 126.53, 125.34, 124.64, 124.28, 121.99, 121.48, 120.51 ppm; HRMS (ESI): *m/z* (calcd.) $[M + H]^+$ 403.0747, found 403.0737.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(1-bromonaphthalen-2-yl)prop-2-en-1-one (2o, $C_{26}H_{16}BrNO_2S$) White solid; yield: 58%; m.p.: > 320 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.52 (d, J = 15.7 Hz, 1H, CO–C=CH), 8.41 (d, J = 8.3 Hz, 1H, naphthalene-3-H), 8.21–8.16 (m, 2H, Ph-2, 6-H), 7.88–7.72 (m, 5H, benzothiazole-4, 7-H, naphthalene-6, 7, 8-H), 7.66–7.60 (m, 2H, naphthalene-4, 5-H), 7.59–7.52 (m, 3H, CO–CH, Ph-3, 5-H), 7.45–7.40 (m, 1H, benzothiazole-5-H), 7.35–7.30 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): δ = 188.97, 170.52, 158.00, 148.81, 144.57, 135.53, 135.08, 132.71, 132.55, 132.42, 130.82, 128.39, 128.23, 128.16, 128.12, 127.94, 127.82, 126.48, 125.06, 124.54, 123.94, 122.01, 121.44, 120.40 ppm; HRMS (ESI): *m/z* (calcd.) $[M+H]^+$ 486.0157, found 486.0149.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-[4-(trifluoromethyl)-phenyl]prop-2-en-1-one (2p, $C_{23}H_{14}F_3NO_2S$) White solid; yield: 49%; m.p.: 142–144 °C; ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.18–8.12 (m, 2H, Ph-2, 6-H), 7.85 (d, J = 15.7 Hz, 1H, CO–C=CH), 7.79–7.72 (m, 4H, Ar(4-CF_3)-2, 6-H, benzothiazole-4, 7-H), 7.69 (d, J = 8.4 Hz, 2H, Ar(4-CF_3)-3, 5-H), 7.63–7.58 (m, 1H, CO–CH), 7.57–7.54 (m, 2H, Ph-3, 5-H), 7.45–7.41 (m, 1H, benzothiazole-5-H), 7.35–7.32 (m, 1H, benzothiazole-6-H) ppm; ^{13}C NMR (101 MHz, $CDCl_3$): δ = 188.57, 170.45, 158.11, 148.77, 143.08, 138.17, 135.26, 132.40, 130.73, 128.58, 126.50, 126.00, 125.96, 124.58, 123.86, 122.00, 121.46, 120.46 ppm; ^{19}F NMR (376 MHz, $CDCl_3$): δ = − 62.84 ppm; HRMS (ESI): *m/z* (calcd.) $[M+H]^+$ 426.0770, found 426.0759.

(*E*)-1-[4-(Benzo[*d*]thiazol-2-yloxy)phenyl]-3-(3,4-dimethylphenyl)prop-2-en-1-one (2q, $C_{24}H_{19}NO_2S$) White solid; yield: 76%; m.p.: 123–125 °C; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 8.31 (d, J = 8.7 Hz, 2H, Ph-2, 6-H), 8.00 (d, J = 7.9 Hz, 1H, benzothiazole-4-H), 7.93 (d, J = 15.6 Hz, 1H, CO–C=CH), 7.73 (dd, J = 12.0, 3.2 Hz, 3H, CO–CH, Ar(3, 4-di-CH_3)-5, 6-H), 7.67 (d, J = 8.7 Hz, 2H, Ph-3, 5-H), 7.62 (d, J = 7.7 Hz, 1H, benzothiazole-7-H), 7.48–7.44 (m, 1H, benzothiazole-5-H), 7.39–7.35 (m, 1H, benzothiazole-6-H), 7.24 (d, J = 7.8 Hz, 1H, Ar(3, 4-di-CH_3)-2-H), 2.29 (s, 3H, CH_3), 2.28 (s, 3H, CH_3) ppm; ^{13}C NMR (101 MHz, DMSO-d_6): δ = 188.33, 171.22, 157.93, 148.80, 145.08, 140.22, 137.39, 136.08, 132.71, 132.44, 131.29, 130.52, 130.26, 127.33, 127.10, 125.01, 122.85, 121.86, 121.36, 121.05, 19.97, 19.76 ppm; HRMS (ESI): *m/z* (calcd.) $[M+H]^+$ 386.1209, found 386.1203.

Evaluation of the antibacterial activity

The turbidimetric method [32–34] was used to evaluate antibacterial activity of the title compounds at concentrations of 100 and 50 μg/cm^3. Bismerthiazol served as the positive control. The test compounds and bismerthiazol were dissolved in DMSO and diluted with sterile distilled water. DMSO was diluted with sterile distilled water for a blank control. 1 cm^3 of the liquid sample containing tested compounds or bismerthiazol was added to 4 cm^3 of the nutrient broth medium (NB: 6 g beef extract, 10 g peptone, 2 g yeast powder, 20 g glucose, and 2 dm^3 distilled water, pH 7.0–7.2). Then, 40 mm^3 of NB containing Xoo, Xac, or Rs was added to 5 cm^3 of the NB containing test compounds or bismerthiazol. The inoculated test tubes were incubated at 28 ± 1 °C under continuous shaking at 180 rpm for 24–48 h. The culture growth was monitored on a spectrophotometer at 600 nm (OD_{600}) and expressed as corrected turbidity. The formula, $I(\%) = (C_{tur} - T_{tur})/C_{tur} \times 100$, was used for calculating the relative inhibitory rate (I, %). C_{tur} represents the corrected turbidity value of bacterial growth on the blank control, and T_{tur} denotes the corrected turbidity value of bacterial growth on treated NB.

Some compounds were tested against Xoo, Xac, and Rs under different concentrations (100, 50, 25, 12.5, and 6.25 μg/cm^3) to obtain the EC_{50} values. EC_{50} values were determined using SPSS17.0, and the experiments were repeated three times.

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