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Regioselective reaction of imidazole-2-thiols with *N*-sulfonylphenyldichloroacetaldimines: en route to novel sulfonylamino-substituted imidazo[2,1-*b*]thiazoles and thiazolo[3,2-*a*]benzimidazoles

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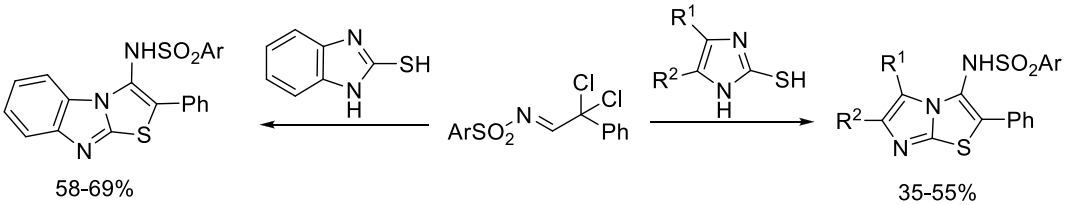
Abstract

The reaction of *N*-(2,2-dichloro-2-phenylethylidene)arenesulfonamides with 2-mercaptoimidazoles affords *N*-(2-phenylimidazo[2,1-*b*][1,3]thiazol-3-yl)arenesulfonamides or *N*-(2-phenyl[1,3]thiazolo[3,2-*a*]benzimidazol-3-yl)arenesulfonamides. Formation of the annulated heterocyclic derivatives is tentatively triggered by a nucleophilic addition of mercaptoimidazole to the activated azomethine group of halogen-containing imines followed by intramolecular heterocyclization and aromatization.



Keywords: Imidazo[2,1-*b*][1,3]thiazole, thiazolo[3,2-*a*]benzimidazole, imines, sulfonamides

Introduction

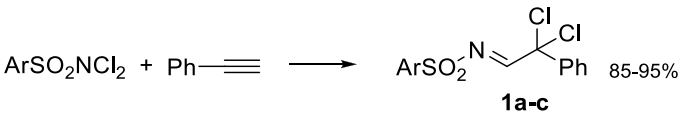
Derivatives of annulated heterocyclic compounds bearing an imidazo[2,1-*b*]thiazole structural motif are known to possess a wide spectrum of biological activity.[1] The prospects for application of biologically active imidazothiazoles stem from their ability of inhibiting or activating the various enzymes and receptors.[2-5] Importantly, these compounds show antitumor,[6-11] antimicrobial,[12,13] antidiabetic,[14] diuretic,[15] antihelmintic,[16] and fungicidal[17] properties. Besides, imidazo[2,1-*b*]thiazoles and thiazolo[3,2-*a*]benzimidazoles are useful reagents in heterocyclic chemistry and can be transformed into benzimidazolone derivatives.[18] Thus, the development of expedient methods for the preparation of substituted imidazo[2,1-*b*]thiazoles and synthesis of novel representatives of these heterocycles are directly connected with a possibility of drug design and, hence, represent an urgent challenge.

The most popular methods for the preparation of imidazo[2,1-*b*]thiazoles are based on the reaction of 2-aminothiazoles or 2-mercaptoimidazoles with α-halocarbonyl compounds.[19] 3-Alkyl-thiazolo[3,2-*a*]benzimidazole derivatives were also obtained from 1,2-diaminobenzene, CS_2, and haloketones via the intermediated 4-alkyl-*N*-3-(2-aminophenyl)-thiazoline-2-thiones.[20] Very promising are multi-component syntheses of aminosubstituted imidazo[2,1-*b*]thiazoles from 2-aminothiazoles, isocyanides and aldehydes or bromoacetophenones, aromatic aldehydes, thiourea and isocyanides.[19,21] Thiazolobenzimidazoles bearing the imidazo[2,1-*b*]thiazole fragment were obtained by the reaction of benzimidazole-2-thiol with bromomalononitrile.[11,22-24]

We are engaged in elaboration of approaches to the preparation of annulated imidazole derivatives containing synthetically important pharmacophoric sulfonylamino group.[25-28]

It is a common knowledge that the sulfonamide compounds are intensively employed for drug design.[29-32] First of all, these are medicinally classical antagonists of folic acid having antimicrobial and antibiotic properties.[29-34] Also, we should take note of sulfonamides exerting antitumor[35-37] and anti-inflammatory activity,[38,39] neuroleptics,[40] anticonvulsants, diuretics, analgetics and antimigraine remedies.[41] Among sulfonamides there are efficient inhibitors of proteases,[41,42] COX-2[43-47] and caspase inhibitors[48,49] associated with different diseases. Some sulfonamides also exhibit herbicidal activity.[50] Sulfonamides are widely used in organic syntheses.[51-55] A sulfonamide fragment can be regarded as a protected amino group.[56,57] Thus, importance of the sulfonamide compounds for modern medicine and organic synthesis can hardly be overestimated.

In continuation of our research,[25-28] here we have studied for the first time the reaction of imidazole-2-thiols with *N*-(2,2-dichloro-2-phenylethylidene)arenesulfonamides **1** to elaborate the approach for the synthesis of novel sulfonylamino-substituted derivatives of imidazo[2,1-*b*]thiazole. *N*-Sulfonylimines **1** represent activated electron-deficient halogen-containing imines, which are effective reagents for a wide range of sulfonamide derivatives.[58,59]



Ar = Ph (**a**), 4-MeC_6H_4 (**b**), 4-ClC_6H_4 (**c**).

Scheme 1. Synthesis of phenyldichloroacetaldimine **1a-c.**[58]

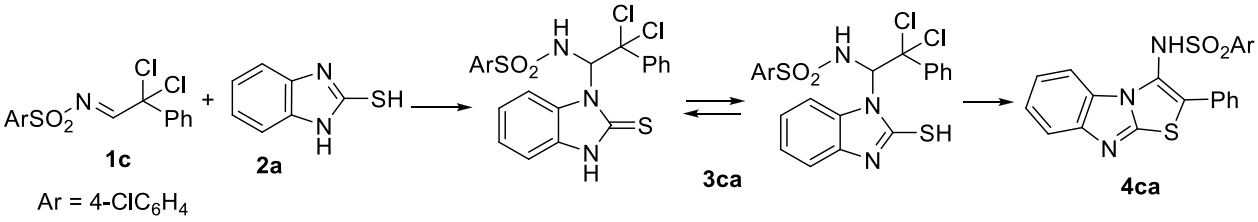
Imines **1a-c** became available owing to a convenient method for their preparation[60] via the radical reaction of phenylacetylene with *N*,*N*-dichloroarenesulfonamides (obtained in turn by chlorination of the corresponding sulfonamides[61]) (Scheme 1).

To reach the goal of this work, the synthesized imines **1a-c** were subjected to the reaction with 2-imidazolethiols **2a-e**.

Results and Discussion

First, screening of conditions for the reaction of imine **1c** with benzimidazolethiol **2a** has been performed. It has been found that the reaction proceeds via formation of the unstable intermediate adduct **3ca**. Further transformations of the latter lead either to resinification of the reaction mixture or formation of the target imidazothiazole **4ca** (Table 1).

Table 1. Conditions for reaction of imine **1c** with benzimidazolethiol **2a**



Entry	Solvent	Base	T, °C	Time, h	Yield of **3ca**, %[a]	Yield of **4ca**, %[a]
1	acetonitrile	TEA	room temp.	5	0	0
2	acetonitrile	K_2CO_3	room temp.	5	0	0
3	acetonitrile	DABCO	room temp.	5	0	0
4	acetonitrile	-	room temp.	1	58	0
5	acetonitrile	-	reflux	5	0	26
6	acetonitrile	-	reflux	15	0	39
7	acetonitrile	-	reflux	30	0	56
8	DMF	-	100°C	5	0	35
9	DMF	-	reflux	5	0	12
10	pyridine	-	reflux	5	0	44
11	1,4-dioxane	-	reflux	5	0	25
12	1,4-dioxane	-	reflux	15	0	57
13	toluene	-	reflux	5	0	60
14	toluene	-	reflux	15	0	66
15	o-xylene	-	reflux	5	0	66
16	o-xylene	TEA	reflux	5	0	0
17	-	-	150°C	5	0	55

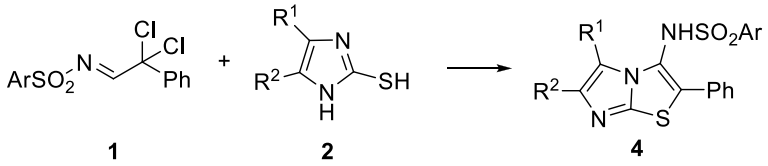
[a] Yields were calculated on starting imine **1c**.

Adduct **3ca** has been isolated in highest yield, when the reaction is carried out in acetonitrile (Table 1, Entry 4). Further heating of adduct **3ca** delivers imidazothiazole **4ca**. Upon refluxing the starting reagents **1c** and **2a** in ***o***-xylene or toluene, a one-pot synthesis of imidazothiazole **4ca** in a highest yield has been implemented (Table 1, Entries 14, 15). The reaction is carried out without isolation of the intermediate adduct **3ca**. But the reaction in toluene takes much more time (Table 1, Entries 14), presumably because of lower temperature of refluxing. Therefore, we used refluxe in ***o***-xylene for futher synthesis.

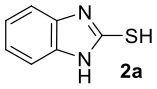
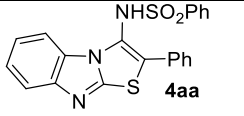
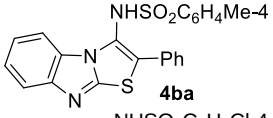
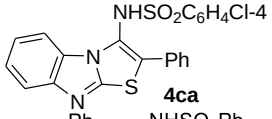
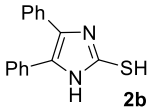
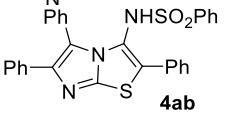
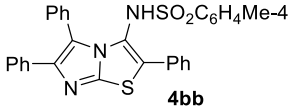
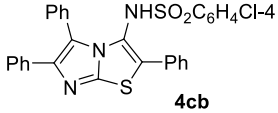
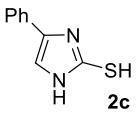
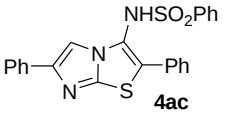
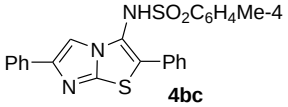
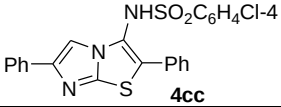
The reaction without a solvent in melt of reagents **1c** and **2a** leads to the formation of imidazothiazole **4ca** also (Table 1, Entries 17). But in this case strong resinification took place, a procedure for isolation of the target product in a pure form was more labour-consuming, and yield of **4ca** was less.

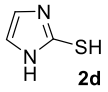
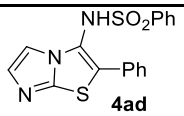
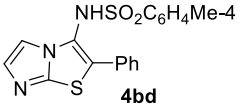
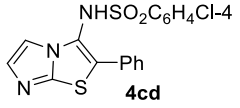
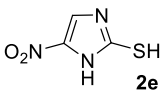
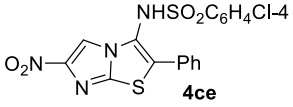
Next, we have tried to extend conditions, optimum for the synthesis of compound **4ac**, to the reactions of imines **1a-c** with benzimidazolethiol **2a** and other representatives of 2-imidazolethiols **2b-e** (Table 2).

Table 2. Synthesis of imidazothiazoles **4** from imines **1** and imidazole thioles **2**



1 **2** **4**

Entry	Imine **1** (Ar)	Imidazole thiol **2**	Imidazothiazole **4**	Yield, %
1	**1a** (Ph)	 **2a**	 **4aa**	58[a]
2	**1b** (4-MeC$_6$H$_4$)	**2a**	 **4ba**	69[a]
3	**1c** (4-ClC$_6$H$_4$)	**2a**	 **4ca**	66[a]
4	**1a** (Ph)	 **2b**	 **4ab**	42[a]
5	**1b** (4-MeC$_6$H$_4$)	**2b**	 **4bb**	55[a]
6	**1c (**4-ClC$_6$H$_4$)	**2b**	 **4cb**	50[a]
7	**1a** (Ph)	 **2c**	 **4ac**	35[a]
8	**1b** (4-MeC$_6$H$_4$)	**2c**	 **4bc**	40[a]
9	**1c** (4-ClC$_6$H$_4$)	**2c**	 **4cc**	37[a]

Entry	Imine **1** (Ar)	Imidazole thiol **2**	Imidazothiazole **4**	Yield, %
10	**1a** (Ph)	 **2d**	 **4ad**	35[b]
11	**1b** (4-MeC_6H_4)	**2d**	 **4bd**	50[b]
12	**1c** (4-ClC_6H_4)	**2d**	 **4cd**	37[b]
13	**1c** (4-ClC_6H_4)	 **2e**	 **4ce**	0[a,b]

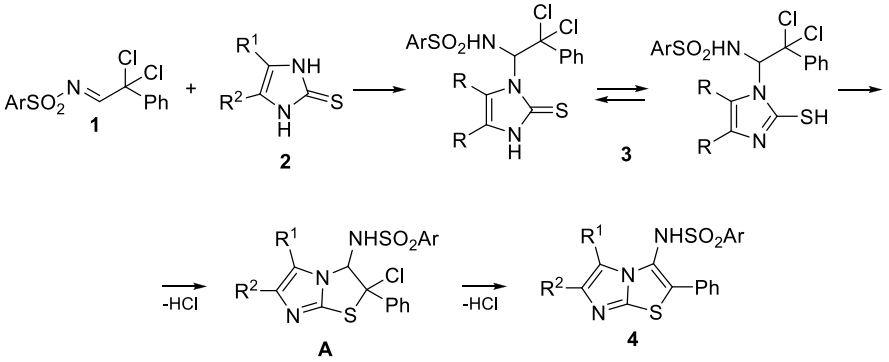
[a] Reflux in *o*-xylene for 5 h.

[b] Heating at 100ºC in DMF for 5 h.

It has been established that 4,5-diphenyl and 4-phenylsubstituted 2-mercaptoimidazoles **2b,c**, like benzimidazolethiol **2a**, smoothly form the corresponding sulfonylaminoimidazothiazoles, when carrying out the reaction in *o*-xylene. At the same time, 2-mercaptoimidazole **2d**, unlike its substituted derivatives **2a-c**, in *o*-xylene as a solvent, do not afford the target imidazothiazoles **4ad**, **4bd**, **4cd**. However, the latter compounds have been synthesized by heating the reagents to 100°C in DMF (Table 2, Entries 10-12).

Besides, despite varying the process conditions, the attempts to obtain imidazothiazoles from the nitrosubstituted mercaptoimidazole **2e** (Table 2, Entries 13) failed, presumably, due to a low nucleophilicity of this reagent.

The tentative mechanism of imidazothiazoles assembly (Scheme 2) is, apparently, triggered by the formation of *N*-adduct **3** via addition of the NH group of mercaptoimidazole **2**, existing as 1,3-dihydro-2*H*-imidazole-2-thione tautomer, to the activated azomethine fragment imine **1**. Further transformations probably include tautomerization of adduct **3** with the formation of the thiol group, intramolecular heterocyclization involving this thiol group and dichloromethylene moiety followed by aromatization owing to elimination of hydrogen chloride.



Scheme 2. The tentative reaction mechanism for the formation of imidazothiazole derivatives **4**.

The structures of compounds **3ca** and **4** were proved by NMR technique. The ^{1}H and ^{13}C NMR spectra of compounds **3ca** show signals of protons and carbon atoms, which relative integrated intensities, multiplicity and chemical shifts correspond to the proposed structure. In the ^{13}C NMR spectrum, the signal at 171 ppm is assigned to the C=S group that, first, confirms formation of adduct **3ca** due to addition of benzimidazolethiol via the NH function, and, second, demonstrates that **3ca** mainly exists as a thiocarbonyl tautomer. In the IR spectra of compound **3ca**, the absorption band at 1218 cm^{-1} also corresponds to the thiocarbonyl structure.

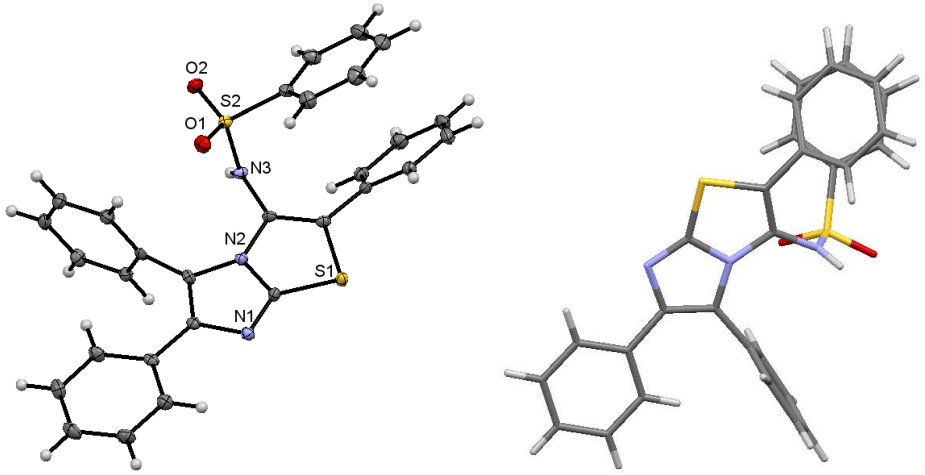


Figure 1. Molecular structures of **4ab** according to X-ray diffraction analysis

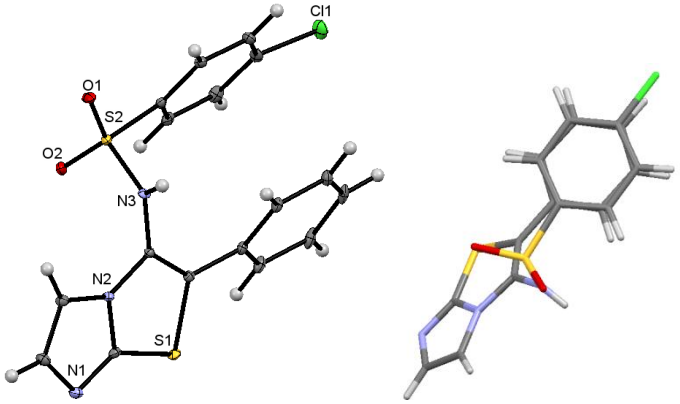


Figure 2. Molecular structures of **4cd** according to X-ray diffraction analysis.

The structure of imidazothiazole derivatives **4** is unambiguously confirmed by single crystal X-ray diffraction analysis used for **4ab** and **4cd** (Figures. 1, 2). The XRD analysis data show that compounds **4ab** and **4cd** are isostructural. Sulfur atom S2 adopts a conformation of the distorted tetrahedron. The valence angles within experimental errors are close to the corresponding values of sulfonylamino derivatives and are O1S2O2 121.5(1)°, C12S2O2 108.8(1)°, N3S2O2 104.5(1)° for **4ab** and O1S2O2 120.7(1)°, C12S2O2 109.5(1)°, N3S2O2 107.5(1)° for **4cd**. The imidazo[2,1-*b*]thiazole fragment has a planar structure, deviation of the N1, N2 and S1 heteroatoms from the plane is 0.026Å, 0.011Å, 0.070Å for **4ab** and 0.001Å, 0.033Å, 0.025Å for **4cd** correspondingly. The plane of the phenyl substituent in the position 2 is with an angle to the plane of

imidazo[2,1-*b*]thiazole fragment: 46.78° for **4ab** and 34.17° for **4cd**. The plane of the aromatic ring of arenesulfonylamino group is almost parallel to the plane of the phenyl substituent in the position 2, an angle between the planes being 6.84° for **4ab** and 6.15° for **4cd**. Besides, atoms S2 of arylsulfonyl groups deviate from the plane of the aryl fragment: 0.014Å for **4ab** and 0.229Å for **4cd**. For the compound **4cd** chlorine atom Cl1 deviates from the plane on 0.150Å. Distances between centroids of the aromatic rings in the position 2 and in arylsulfonyl groups are 3.681Å for **4ab** and 3.653Å for **4cd** that evidences the presence of intramolecular π-stacking. For **4ab**, the almost orthogonal planes (88.12°) of the phenyl groups in the positions 5 and 6 of imidazo[2,1-*b*]thiazole core and distance between their centroids (3.644Å) testify in favor of intramolecular *t*-stacking.

A peculiarity of **4ab** and **4cd** structures is the conformational difference in the arrangement of the phenyl fragments (Figures 1, 2). Compound **4ab** is characterized by the staggered conformation, while compound **4cd** is of the eclipsed conformation. Alteration of the conformation is associated with the change of the torsion angles $C3N3S2C_{Ar}$ [(65.9(2)° for **4ab**, -90.7(2)° for **4cd**] and arrangement of phenyl group in the position 2 relative to imidazothiazole core - the torsion angles $N3C3C2C_{Ph}$ are -10.7(4)° for **4ab** and 3.8(4)° for **4cd**.

To establish the structure of imidazothiazole **4bc** HMBC $^{1}H–^{13}C$ technique was used (Figure 3), which was optimized for $^{13}C–^{1}H$ spin–spin coupling constants of 10 Hz, which are typical for carbon and proton atoms separated by three bonds. The $^{13}C–^{1}H$ correlation through three bonds with participation of C-3 carbon atom permits to draw a conclusion about absence of a substituent in the position 5 of the imidazothiazol cycle, and, therefore, on presence of the phenyl group in the position 6.

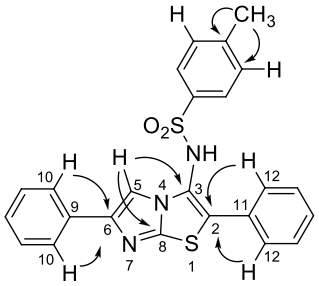


Figure 3. Main ^{1}H-^{13}C HMBC correlations for imidazothiazole **4bc**.

^{1}H and ^{13}C NMR spectra are similar in the series **4ac**, **4bc**, **4cc**. In the ^{13}C NMR spectra there are similar signals corresponding to HC-5 carbon atoms in the region of 109 ppm. So, based on this, we believe that these compounds are 6-Ph substituted derivatives.

Conclusions

The reaction of 2-mercaptoimidazoles with *N*-(sulfonyl)phenyldichloroacetaldimines leads to the formation of *N*-(2-phenylimidazo[2,1-*b*][1,3]thiazol-3-yl)arenesulfonamides or *N*-(2-phenyl[1,3]thiazolo[3,2-*a*]benzimidazol-3-yl)arenesulfonamides. The advantages of the method for the preparation of the imidazothiazole and thiazolobenzimidazole derivatives are available starting reagents, catalyst-free one-step procedure, and high selectivity. The known methods[19,21-24] have never been used for the preparation of sulfonylaminosubstituted derivatives containing synthetically useful and pharmacophoric sulfonamide groups. It can be argued that the method proposed herein complements the known literature protocols and expands the scope of

functionalized imidazothiazoles derivatives which are now available for further investigation of biological activity and other properties.

Experimental Section

General. Imines **1a-c** were synthesized according to the known procedure[60] from corresponding N,N-dichloroarensulfonamides.[61] All other used reagents were reagent grade. The solvents were dried by standard procedures and distilled prior to use. NMR spectra were recorded on a Bruker DPX 400 spectrometer (^{1}H, 400.13 MHz; ^{13}C, 100.61 MHz) at 25 °C with HMDS as an internal standard. Chemical shifts are reported in ppm values (δ) and coupling constants (*J*) in Hz. IR spectra were recorded on a Bruker IFS-25 spectrophotometer in KBr. All melting points were measured on a Kofler micro hot stage apparatus. Elemental analyses for C, H, N and S were obtained using a Thermo Finnigan Flash series1112 EA analyzer. Column chromatography was carried out in a glass column with a diameter 2 sm and length of silica gel 30 sm (230–400 mesh). Sorbent : product ratio was 100 : 1.

X-Ray crystallographic data were collected on a BRUKER D8 VENTURE PHOTON 100 CMOS diffractometer with MoK$_\alpha$ radiation (λ 0.71073 Å) using the ϕ and ω scans technique. The structures were solved and refined by direct methods using the SHELX[62]. Data were corrected for absorption effects using the multi-scan method (SADABS). All non-hydrogen atoms were refined anisotropically using SHELX.[62] The coordinates of the hydrogen atoms were calculated from geometrical positions. Crystallographic data for the structures **4ab** and **4cd** in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication numbers CCDC 1564758 for **4ab** and 1564759 for **4cd**. Copies of the data can be obtained, free of charge, on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK [fax: +44(0)-1223-336033 or e-mail: deposit@ccdc.cam.ac.uk].

4-Chloro-*N*-[2,2-dichloro-2-phenyl-1-(2-thioxo-2,3-dihydro-1*H*-benzimidazol-1-yl)ethyl]benzenesulfonamide (3ca). A mixture of imine **1c** (0.500 g, 1.4 mmol) and benzimidazolethiol **2a** (0.210 g, 1.4 mmol) in acetonitrile (10 mL) was stirred for 1 h. The residue precipitated was filtered off, and the solid product was washed with acetonitrile and dried in vacuum over P_2O_5 to give **3ca** as a white solid. Yield 58%, 420 mg, mp 145–146 °C. IR (KBr, ν, cm^{-1}): 1585, 1472, 1431 (C=C, C=N), 1358, 1166 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 6.93-6.99 (m, 1H), 6.99-7.15 (m, 4H), 7.32-7.47 (m, 5H), 7.52 (d, ^3J 10.2 Hz, 1H), 7.71-7.80 (m, 2H), 7.88-7.95 (m, 1H), 9.70 (d, *J* 10.2 Hz, 1H), 12.78 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 73.37, 92.86, 109.32, 113.16, 121.73, 122.31, 123.19, 127.15, 127.63, 128.18, 128.27, 129.21, 129.82, 130.65, 137.34, 138.70, 171.00. Anal. calcd for $C_{21}H_{16}Cl_3N_3O_2S_2$ (512.86): C, 49.18; H, 3.14; N, 8.19; S 12.50; Found: C, 49.33; H, 3.17; N, 8.31; S 12.56.

Procedure for synthesis of 4aa, 4ba, 4ca. A mixture of imine **1a** (0.460 g, 1.4 mmol) and benzimidazolethiol **2a** (0.255 g, 1.7 mmol) in o-xylene (10 mL) was refluxed for 5 h. The mixture was cooled, filtered, and the solid product was washed with diethyl ether (30-50 mL) and hot methanol (10 mL) and recrystallized from ethanol. Reactions of **2a** with imines **1b** (0.480 g, 1.4 mmol) or **1c** (0.500 g, 1.4 mmol) were carried out at the same manner.

***N*-(2-Phenyl[1,3]thiazolo[3,2-*a*]benzimidazol-3-yl)benzenesulfonamide (4aa).** White solid. Yield 58%, 329 mg, mp 270-271 °C. IR (KBr, ν, cm^{-1}): 1619, 1596, 1468, 1448 (C=C, C=N), 1348, 1171 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.06-7.44 (m, 10H), 7.45-7.61 (m, 2H), 7.73 (m, 1H), 8.08 (m, 1H), 11.35 (br. s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 112.15, 118.51, 119.59, 120.84, 123.42, 124.08, 126.28, 127.81, 128.65, 128.74, 128.85,

129.00, 129.79, 132.96, 140.22, 147.03, 150.34. Anal. calcd for $C_{21}H_{15}N_3O_2S_2$ (405.49): C, 63.85; H, 3.73; N, 10.36; S, 15.82; Found: C, 63.98; H, 3.62; N, 10.41; S, 15.97.

4-Methyl-*N*-(2-phenyl[1,3]thiazolo[3,2-*a*]benzimidazol-3-yl)benzenesulfonamide (4ba). White solid. Yield 69%, 405 mg, mp 269-271 °C. IR (KBr, ν, cm^{-1}): 1618, 1598, 1468, 1446 (C=C, C=N), 1349, 1168 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.20 (s, 3H), 6.86-7.01 (m, 2H), 7.08-7.46 (m, 9H), 7.73 (m, 1H), 8.10 (m, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 20.89, 112.29, 118.51, 119.67, 120.88, 123.46, 124.14, 126.36, 127.86, 128.36, 128.59, 128.96, 129.36, 129.84, 130.34, 136.87, 143.09, 143.44, 144.25, 147.01, 150.36. Anal. calcd for $C_{22}H_{17}N_3O_2S_2$ (419.52): C, 62.99; H, 4.08; N, 10.02; S, 15.29; Found: C, 62.87; H, 3.95; N, 10.23; S, 15.39.

4-Chloro-*N*-(2-phenyl[1,3]thiazolo[3,2-*a*]benzimidazol-3-yl)benzenesulfonamide (4ca). White solid. Yield 66%, 407 mg, mp 280-282 °C. IR (KBr, ν, cm^{-1}): 1616, 1584, 1464, 1451 (C=C, C=N), 1328, 1133 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.10-7.52 (m, 11H), 7.68-7.81 (m, 1H), 8.05-8.16 (m, 1H) ppm. ^{13}C NMR (101 MHz, DMSO-d_6): δ 112.16, 118.58, 119.46, 120.97, 123.51, 124.20, 127.95, 128.23, 128.49, 128.67, 128.88, 129.04, 129.81, 138.57, 138.57, 138.18, 147.00, 150.40. Anal. calcd for $C_{21}H_{14}ClN_3O_2S_2$ (439.94): C, 57.33; H, 3.21; N, 9.55; S, 14.58; Found: C, 57.20; H, 3.17; N, 9.74; S, 14.79.

Procedure for synthesis of 4ab, 4bb, 4cb. A mixture of imine **1a** (0.460 g, 1.4 mmol) and 4,5-diphenylimidazole-2-thiol **2b** (0.429 g, 1.7 mmol) in *o*-xylene (10 mL) was refluxed for 5 h. The mixture was cooled, filtered, and the solid product was washed with diethyl ether (30-50 mL). Purified by column chromatography on silica gel, acetonitrile : chloroform (1:9) was used as eluent, and recrystallized from ethanol. Reactions of **2b** with imines **1b** (0.480 g, 1.4 mmol) or **1c** (0.500 g, 1.4 mmol) were carried out at the same manner.

***N*-(2,5,6-Triphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4ab)**. White solid. Yield 42%, 302 mg, mp 236-238 °C. IR (KBr, ν, cm^{-1}): 1601, 1588, 1474, 1467 (C=C, C=N), 1368, 1170 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.01-7.33 (m, 13H), 7.41-7.44 (m, 2H), 7.46-7.50 (m, 5H), 10.4 (br.s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 119.3, 124.4, 126.0, 126.87, 126.96, 126.99, 127.01, 127.94, 128.05, 128.08, 128.44, 128.46, 128.64, 128.74, 129.16, 131.95, 132.27, 133.93, 140.70, 141.98, 142.72. Anal. calcd for $C_{29}H_{21}N_3O_2S_2$ (507.63): C, 68.62; H, 4.17; N, 8.28; S, 12.63; Found: C, C, 68.81; H, 4.25; N, 8.17; S, 12.78.

4-Methyl-*N*-(2,5,6-triphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4bb). Yellow solid. Yield 55%, 400 mg, mp 130-132 °C. IR (KBr, ν, cm^{-1}): 1600, 1504, 1477 (C=C, C=N), 1362, 1166 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.16 (s, 3H), 6.75-6.84 (m, 2H), 7.05-7.13 (m, 4H), 7.16-7.28 (m, 6H), 7.38-7.42 (m, 2 H), 7.45-7.52 (m, 5H), 10.24-10.34 (m, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 20.86, 79.18, 119.38, 124.43, 126.16, 126.89, 126.92, 127.01, 128.03, 128.15, 128.20, 128.21, 128.49, 129.04, 129.11, 129.32, 132.05, 134.30, 137.48, 142.38, 142.70, 142.88. Anal. calcd for $C_{30}H_{23}N_3O_2S_2$ (521.65): C, 69.07; H, 4.44; N, 8.06; S, 12.29; Found: C, 68.97; H, 4.51; N, 8.15; S, 12.36.

4-Chloro-*N*-(2,5,6-triphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4cb). Yellow solid. Yield 50%, 380 mg, mp 136-138 °C. IR (KBr, ν, cm^{-1}): 1603, 1477, 1433 (C=C, C=N), 1366, 1169 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.03-7.30 (m, 12H), 7.30-7.57 (m, 7H), 10.52 (br.s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 48.55, 119.02, 124.27, 126.78, 126.92, 127.96, 128.03, 128.07, 128.09, 128.29, 128.44, 128.50, 128.72, 128.96, 129.19, 131.98, 134.22, 137.52, 139.16, 142.41, 142.84. Anal. calcd for $C_{29}H_{20}ClN_3O_2S_2$ (542.07): C, 64.29; H, 3.72; N, 7.75; S, 11.83; Found: C, 64.39; H, 3.68; N, 7.88; S, 11.94.

Procedure for Synthesis of 4ac, 4bc, 4cc. A mixture of imine **1a** (0. 0.460 g, 1.4 mmol) and 4-phenylimidazole-2-thiol **2c** (0.300 g, 1.7 mmol) in *o*-xylene (10 mL) was refluxed for 5 h. The mixture was cooled, filtered, the solid product was washed with diethyl ether (30 mL), and recrystallized from ethanol. Reactions of **2c** with imines **1b** (0.480 g, 1.4 mmol) or **1c** (0.500 g, 1.4 mmol) were carried out at the same manner.

***N*-(2,5-Diphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4ac).** White solid. Yield 35%, 210 mg, mp 242–243 °C. IR (KBr, ν, cm^{-1}): 1597, 1514, 1493, 1450 (C=C, C=N), 1344, 1167 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.23-7.39 (m, 5H), 7.40-7.52 (m, 5H), 7.57 (m, 2H), 7.78-7.86 (m, 2H), 7.94 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 109.04, 118.97, 125.02, 126.64, 127.21, 128.04, 128.15, 128.74, 128.80, 128.83, 128.97, 129.15, 129.24, 133.30, 139.81, 142.71, 142.77 ppm. Anal. calcd for $C_{23}H_{17}N_3O_2S_2$ (431.53): C, 64.05; H, 3.97; N, 9.74; S, 14.86; Found: C, 64.24; H, 4.09; N, 9.88; S, 14.65.

4-Methyl-*N*-(2,5-diphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4bc). White solid. Yield 40%, 250 mg, mp 231–233 °C. IR (KBr, ν, cm^{-1}): 1597, 1493, 1470 (C=C, C=N), 1340, 1166 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.25 (s, 3H), 7.07-7.14 (m, 2H), 7.24-7.36 (m, 4H), 7.38-7.48 (m, 6H), 7.74-7.82 (m, 3H), 11.11 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 20.96, 108.74, 118.71, 124.91, 126.66, 126.75, 127.81, 127.99, 128.67, 128.71, 128.86, 128.92, 129.65, 132.39, 136.57, 142.91, 143.80, 143.84. Anal. calcd for $C_{24}H_{19}N_3O_2S_2$ (445.56): C, 64.07; H, 14.30; N, 9.43; S 14.39; Found: C, 64.26; H, 14.15; N, 9.35; S 14.47.

4-Chloro-*N*-(2,5-diphenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4cc). White solid. Yield 37%, 240 mg, mp 251–253 °C. IR (KBr, ν, cm^{-1}): 1585, 1472, 1431 (C=C, C=N), 1358, 1166 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.20-7.58 (m, 12H), 7.80-7.91 (m, 2H), 7.99 (s, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 108.85, 118.53, 124.97, 127.04, 127.86, 128.00, 128.41, 128.58, 128.64, 128.79, 128.85, 129.11, 131.95, 138.19, 138.26, 142.83, 143.36. Anal. calcd for $C_{23}H_{16}ClN_3O_2S_2$ (465.98): C, 59.28; H, 3.46; N, 9.02; S 13.76; Found: C, 59.45; H, 3.37; N, 8.84; S 13.62.

Procedure for Synthesis of 4ad, 4bd, 4cd. A mixture of imine **1a** (0.460 g, 1.4 mmol) and imidazole-2-thiol **2d** (0.170 g, 1.7 mmol) in DMF (10 mL) was heated at 100°C for 5 h. The mixture was cooled, poured into water (100 mL). In one day, the residue precipitated was filtered off and purified by column chromatography on silica gel, acetonitrile – chloroform (2:3) was used as eluent. Reactions of **2d** with imines **1b** (0.480 g, 1.4 mmol) or **1c** (0.500 g, 1.4 mmol) were carried out at the same manner.

***N*-(2-Phenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4ad)**. Yellow solid. Yield 35%, 175 mg, mp 114–117 °C. IR (KBr, ν, cm^{-1}): 1595, 1486, 1467 (C=C, C=N), 1347, 1170 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 7.21-7.42 (m, 9H), 7.46-7.52 (m, 1H) 7.54-7.59 (m, 2H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 112.51, 118.66, 125.25, 126.35, 127.81, 128.62, 128.70, 129.04, 129.45, 133.01, 133.05, 139.96, 142.36. Anal. calcd for $C_{17}H_{13}N_3O_2S_2$ (355.43): C, 57.45; H, 3.69; N, 11.82; S, 18.04; Found: C, 57.58; H, 3.77; N, 11.93; S, 18.16.

4-Methyl-*N*-(2-phenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4bd). White solid. Yield 50%, 260 mg, mp 246–248 °C. IR (KBr, ν, cm^{-1}): 1597, 1489, 1467 (C=C, C=N), 1335, 1165 (SO_2). ^{1}H NMR (400 MHz, DMSO-d_6): δ 2.27 (s, 3H), 7.08 (m, 2H), 7.17-7.46 (m, 9H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 20.98, 112.66, 118.59, 125.48, 126.45, 127.90, 128.55, 128.60, 129.40, 129.53, 133.22, 136.64, 142.42, 143.58. Anal. calcd for $C_{18}H_{15}N_3O_2S_2$ (369.46): C, 58.52; H, 4.09; N, 11.37; S, 17.36; Found: C, 58.64; H, 4.05; N, 11.48; S, 17.46.

4-Chloro-*N*-(2-phenylimidazo[2,1-*b*][1,3]thiazol-3-yl)benzenesulfonamide (4cd). White solid. Yield 37%, 200 mg, mp 243-245 °C. IR (KBr, ν, cm^{-1}): 1488, 1467 (C=C, C=N), 1347, 1172 (SO_2).^{1}H NMR (400 MHz, DMSO-d_6): δ 7.17-7.39 (m, 9H), 7.41-7.51 (m, 2H), 7.51-7.59 (m, 1H). ^{13}C NMR (101 MHz, DMSO-d_6): δ 112.67, 118.92, 124.97, 127.78, 128.17, 128.43, 128.52, 129.00, 129.28, 132.74, 137.89, 138.59, 142.33. Anal. calcd for $C_{17}H_{12}ClN_3O_2S_2$ (389.88): C, 52.37; H, 3.10; N, 10.78; S, 16.45; Found: C, 52.46; H, 3.21; N, 10.69; S, 16.68.

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Supplementary Material

Copies of ^{1}H, ^{13}C NMR spectra for compounds **3ac**, **4aa-4ac**, and **4cc**-**4cd**; X-Ray crystallographic data for compound **4ab**, **4cd**; Copies of ^{1}H, ^{13}C and 2D NMR spectra for compounds **4bc**.

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