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An one-pot approach to the synthesis of triazolobenzothiadiazepine 1,1-dioxide derivatives by basic alumina-supported azide–alkyne [3+2] cycloaddition

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ABSTRACT

An efficient one-pot strategy for the synthesis of triazolobenzothiadiazepine 1,1-dioxide derivatives has been developed by the reaction of 2-azido-*N*-substituted benzenesulfonamides and propargyl bromide in basic alumina under microwave condition via [3+2] azide–alkyne cycloaddition reaction. This protocol has synthetic advantages in terms of low environmental impact and short reaction time.

1. Introduction

Over the last decade, design of the sultam scaffold has attracted the attention of synthetic organic chemists due to their biological and pharmacological potency.[1] The ability to serve as amide surrogates, with unique physical properties, have made them ideal functionalities for the development of novel peptidomimetics.[2] A number of benzo-fused thiadiazepines have recently appeared that display potent activities including selective tumour necrosis factor inhibitor[3] and antidepressant agent.[4] In particular, benzothiadiazepine 1,1-oxides are a pivotal structural entity and possess a remarkable range of biological properties such as antiblastic, antitumour, apoptotic and antihypertensive activities.[5] On the other hand, the 1,2,3-triazole moiety and fused triazole have their own importance in synthetic, medicinal as well as materials chemistry.[6,7] Especially, fused triazolobenzothiadiazepines have gained much attention due to their important characteristics as antimicrobial and antibacterial agents.[8]

A large number of reports have appeared in the literature on the synthesis of 1,2,3-triazoles[6,9] and fused triazoles.[10] Although various approaches to the preparation of benzothiadiazepine 1,1-oxide derivatives have been reported,[5] most of them are associated with a number of drawbacks like the use of hazardous organic solvents, stepwise synthesis, use of catalyst or expensive reagents, requirement of inert atmosphere and stringent reaction condition. Development of mild and simple method to synthesize benzothiadiazepine 1,1-oxide derivatives is still desirable because of their biological significance. Huisgen's 1,3-dipolar cycloaddition reaction[11] of an azide with an alkyne may be useful for the above purpose. We employed the possibility of using a solid-supported reaction using alumina[12] or silica because organic compounds can get adsorbed on the surface of the inorganic oxide that itself does not absorb or restrict the transmission of microwave irradiation.[13] In continuation of our efforts on sustainable synthesis development,[14] we synthesized benzothiadiazepine 1,1-oxide derivatives via a microwave-assisted, one-pot reaction of 2-azido-*N*-substituted benzenesulfonamide and propargyl bromide in basic alumina. Herein we report the results of our investigation.

2. Results and discussion

At first we prepared 2-azido-*N*-phenylbenzenesulfonamides (**1a**–**i**) required for our study, using the standard protocol.[15] Initially, we treated 2-azido-*N*-phenylbenzenesulfonamide (**1a**) and propargyl bromide (**2a**) in DMF at room temperature using K_2CO_3 as base and obtained a solid product in 56% yield (Scheme 1). The spectral and analytical data of the compound revealed the formation of the desired product triazolobenzothiadiazepine 1,1-dioxide **3a**.

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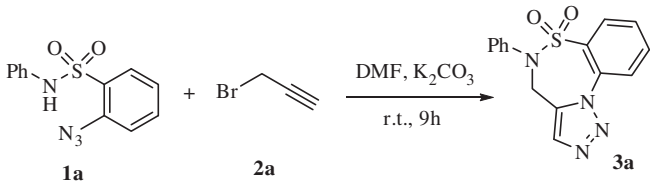
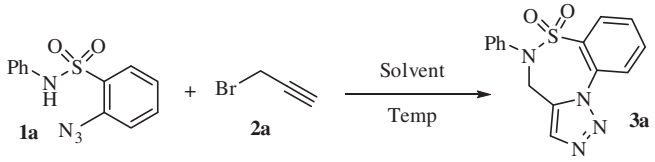


Scheme 1. Synthesis of triazolothiadiazepine 1,1-dioxide **3a** in DMF.

Application of heat increased the yield to 65% (entry 2, Table 1). Change in bases by Cs_2CO_3 and Et_3N did not give any better yield (entries 3 and 4, Table 1). Next we have performed a series of reactions with the replacement of the solvent by other solvents like CH_3CN, acetone, EMK, toluene, MeOH, EtOH and ^tBuOH, but none of them afforded any better result (entries 3–11, Table 1). The observations are summarized in Table 1. We became interested to explore the reaction using solid supports like silica and alumina because of the fact that organic groups can robustly anchor to their surface.[16] Minimization of energy consumption is an important factor for developing technologies. In this respect the microwave irradiation technique is well known for achieving energy efficiency and enhancing the rate of reaction.[17]

Table 1
Summary of intramolecular azide–alkyne [3+2] cycloaddition using different solvents[a]



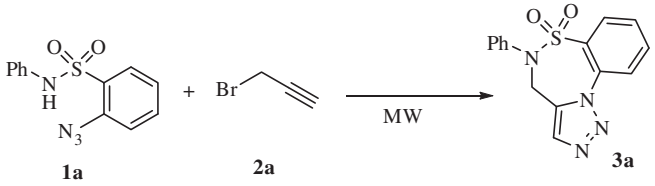
Entry	Solvent	Base	Temp (°C)	Time (h)	Yield[b] (%)
1	DMF	K_2CO_3	rt	9	56
2	DMF	K_2CO_3	80 °C	6	65
3	DMF	Cs_2CO_3	80 °C	6	59
4	DMF	Et_3N	80 °C	8	52
5	CH_3CN	K_2CO_3	rt	12	46
6	CH_3CN	K_2CO_3	Reflux	8	58
7	Acetone	K_2CO_3	Reflux	12	<10
8	Acetone	K_2CO_3	Reflux	10	36
9	EMK	K_2CO_3	80 °C	8	43
10	Toluene	K_2CO_3	80 °C	8	<20
11	MeOH	K_2CO_3	Reflux	10	30
12	EtOH	K_2CO_3	Reflux	7	26
13	*t*-BuOH	K_2CO_3	80 °C	8	37

[a] All the reactions were carried out using 1 equiv **1a** and 1.5 equiv **2a** and 2 equiv base used.
[b] Isolated yields.

Initially, reaction of 2-azido-*N*-phenylbenzenesulfonamide (**1a**) with 1.5 equiv propargyl bromide (**2a**) was investigated with neutral alumina using 2 equiv Et_3N as a base under microwave irradiation. After 15 min of stirring at 80 °C the desired product **3a** was obtained in 68% yield (entry 1, Table 2). We then tried to modify the reaction condition varying the bases by K_2CO_3, Na_2CO_3 and KF but they did not give any better result (entries 2–4, Table 2). The reaction in neutral alumina did not occur in absence of any bases (entry 5, Table 2). To further tune the reaction condition we performed the reaction replacing the solid support by basic alumina with a view to increase the yield of the product **3a**.

Pleasingly, the reaction in basic alumina without any external base improved the yield of the reaction to 92% at 80 °C under microwave heating for only 10 min (entry 6, Table 2). But the reaction in basic alumina at room temperature gave only 32% product after stirring for 14 h (entry 7, Table 2). However, the yield of the product

Table 2
Optimization of solid-supported intramolecular azide–alkyne [3+2] cycloaddition reaction under microwave irradiation[a]



Entry	Solid support–base	Temp (°C)	Time	Yield[d] (%)
1	Neutral alumina–Et_3N[b]	80	15 min	68
2	Neutral alumina–K_2CO_3[b]	90	20 min	56
3	Neutral alumina–Na_2CO_3[b]	90	20 min	52
4	Neutral alumina–KF[b]	90	20 min	54
5	Neutral alumina	90	20 min	0
6	Basic alumina	80	10 min	92
7	Basic alumina[c]	rt	14 h	32
8	Basic alumina	100	10 min	85
9	Basic alumina–K_2CO_3[b]	80	15 min	89
10	Silica gel–Et_3N[b]	80	20 min	45
11	Silica gel–K_2CO_3[b]	90	20 min	42
12	Silica gel–Na_2CO_3[b]	90	20 min	37
13	Silica gel–KF[b]	90	20 min	40

[a] All the reactions were carried out using 1 equiv **1a** and 1.5 equiv **2a** under microwave irradiation techniques.
[b] Bases (2 equiv) were added.
[c] Reaction was stirred at room temperature without MW.
[d] Isolated yields.

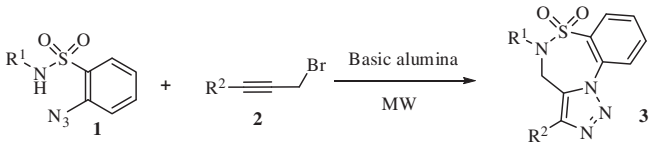
was decreased when the reaction was carried out at 100 °C. The decomposition of azide at higher temperature[18] may be responsible for this lower yield of the product. However, lower yields were obtained when the reactions were performed by changing the reaction condition by using the combination of bases and silica gel as solid supports (entries 10–13, Table 2). It is notable that basic alumina appears to be the most effective solid support for this reaction.

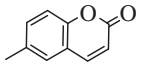
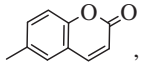
From the above set of experiments the optimized condition developed so far is treatment of 1 equiv 2-azido-*N*-substituted benzenesulfonamide (**1a**) with 1.5 equiv propargyl bromide (**2a**) in basic alumina under microwave irradiation at 80 °C for 10 min. We then carried out the reactions of a variety of substrates under the optimized condition and the results are summarized in Table 3.

The formation of products **3** may be rationalized by considering two alternative pathways (path-a or path-b). Path-a may involve N-alkylation of $-SO_2NH_2$ by the reaction of nucleophilic sulfonamide **1** with propargyl bromides **2** in the presence of basic alumina to form the intermediates **IA** that may then undergo 1,3-dipolar azide–alkyne cycloaddition leading to simultaneous formation of seven-membered sultam fused with five-membered triazole rings. An alternative pathway (path-b) may occur via the early formation of bromomethyl-1,2,3-triazole intermediates **IB** from the intermolecular azide–alkyne cycloaddition reaction of propargyl bromides with the azido group of nucleophilic sulfonamide **1**, followed by intramolecular coupling of bromomethyl group of triazole with $-SO_2NH$. It is well known that here the reaction is exclusively 1,4-directing and momentaneous. It should therefore follow the path-a rather than path-b. If the reaction would have followed path-b then that should have afforded some di- and oligomerized product along with the desired product followed by the formation of 1,5-regioisomer (**IB′**) directing the arrangement of the tethering group (Scheme 2).

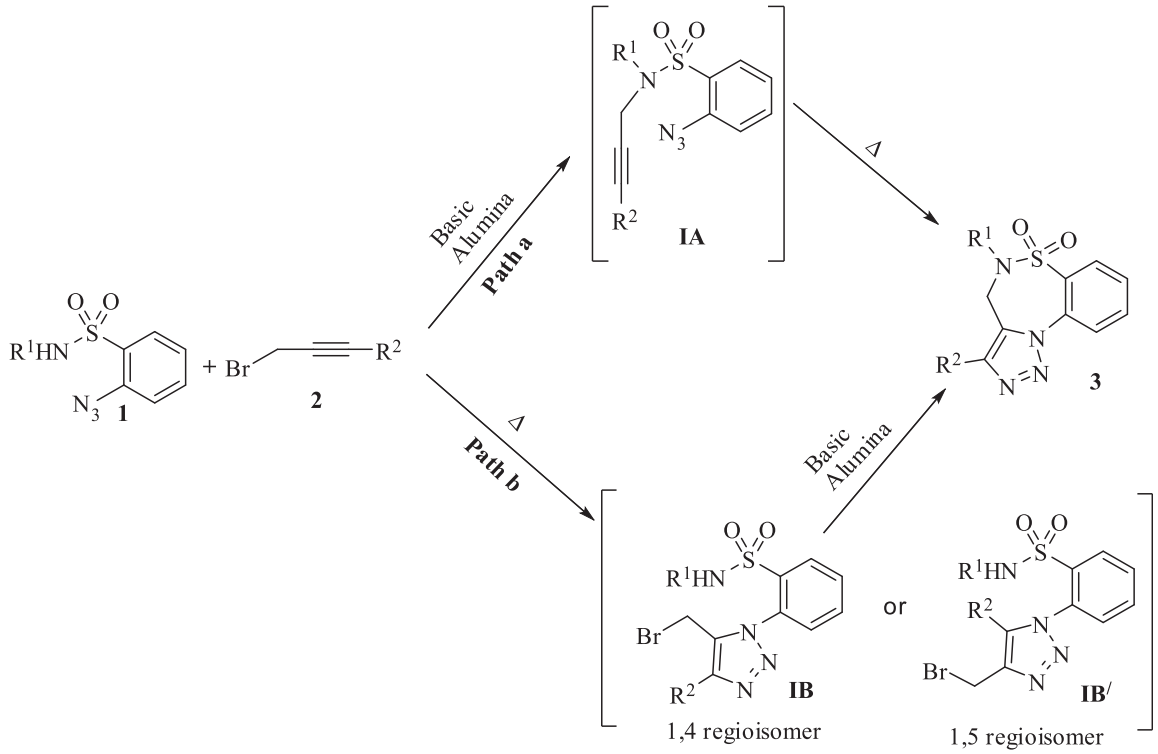
It is now evident that the microwave-assisted cyclization protocol is quite useful for the synthesis of triazolobenzothiadiazepine 1,1-dioxide derivatives as this procedure can effectively avoid the use of environmentally hazardous solvents as well as externally

Table 3
Intramolecular azide–alkyne cycloaddition with variety of substrates under microwave irradiation[a]



Entry	Azidosulphonamide	Alkyne	Time (min)	Product	Yield[b] (%)
1	**1a**, R^1=C_6H_5	**2a**, R^2=H	10	**3a**, R^1=C_6H_5, R^2=H	92
2	**1b**, R^1=4-Cl–C_6H_4	**2a**, R^2=H	10	**3b**, R^1=4-Cl–C_6H_4, R^2=H	94
3	**1b**, R^1=4-Cl–C_6H_4	**2b**, R^2=Me	10	**3c**, R^1=4-Cl–C_6H_4, R^2=Me	90
4	**1c**, R^1=4-Me–C_6H_4	**2a**, R^2=H	10	**3d**, R^1=4-Me–C_6H_4, R^2=H	89
5	**1c**, R^1=4-Me–C_6H_4	**2b**, R^2=Me	10	**3e**, R^1=4-Me–C_6H_4, R^2=Me	86
6	**1d**, R^1=4-OMe–C_6H_4	**2a**, R^2=H	10	**3f** , R^1=4-OMe–C_6H_4, R^2=H	91
7	**1e**, R^1=–$CH_2C_6H_5$	**2a**, R^2=H	10	**3g**, R^1=–$CH_2C_6H_5$, R^2=H	87
8	**1f**, R^1=Me	**2a**, R^2=H	10	**3h**, R^1=Me, R^2=H	95
9	**1g**, R^1=H	**2a**, R^2=H	10	**3i**, R^1=H_2C–C≡CH, R^2=H	88
10	**1h**, R^1 = 	**2a**, R^2=H	10	**3j**, R^1 =  , R^2 = H	83
11	**1i**, R^1 = 	**2a**, R^2=H	10	**3k**, R^1 =  , R^2 = H	85

[a] Reaction conditions: **1** (1 mmol), **2** (1.5 mmol) were microwave irradiated at 80 °C in basic alumina.
[b] Isolated yields.



Scheme 2. Rationalization for the formation of compound **3**.

added bases. Moreover, the reaction condition developed can also avoid the separation problems and use of expensive reagents. Recently Barange and his co-workers[19] have reported the synthesis of triazole-fused sultams via [3+2] cycloaddition followed by C–N bond formation using Cu catalyst and DMF solvent, which provides relatively low yields of the products. It should also be noted that we have achieved the triazolobenzothiadiazepine derivatives in a much simpler manner whereas Hemming et al.[20] have reported the synthesis of triazolobenzodiazepines and pyrrolobenzodiazepines using intramolecular 1,3-dipolar cycloaddition reaction in a multistep process.

3. Conclusions

In conclusion, we have developed a simple and efficient one-pot method for the synthesis of potentially bioactive triazol-

obenzothiadiazepine 1,1-dioxide derivatives via [3+2] azide–alkyne cycloaddition reaction avoiding the use of hazardous solvent, additional bases as well as costly reagents etc. The process affords considerable synthetic advantages in terms of shorter reaction time, product diversity, simplicity of the reaction procedure and good to excellent yields. This may be suitable for generation of library of relevant compounds.

4. Experimental section

4.1. General information

Melting points were determined in open capillaries and are uncorrected. Silica gel [(60–120 mesh) was used for chromatographic separation. Silica gel G was used for TLC. Petroleum ether (Pet) refers to the fraction boiling between 60 °C and 80 °C. IR spectra were recorded on a Perkin–Elmer L 120-000A spectrometer (ν_{max} in cm^{-1}) on KBr disks. ^1H NMR and ^{13}C spectra were recorded in $CDCl_3$ and DMSO (chemical shift in δ) with TMS as internal standard. MS were recorded on a Q-TOF micro™ instrument at the Indian Institute of Chemical Biology (Kolkata). CHN analyses were recorded on a Perkin–Elmer 2400 series II CHN analyser from University of Kalyani. HRMS were recorded on a Q-TOF Micro YA263 instrument at Indian Association for the Cultivation of Science (Kolkata). The MW reactor used for the reaction was a CEM, Discover, USA.

4.2. General procedure for the preparation of 2-azidobenzenesulfonic acid[15a]

2-Amino-benzenesulfonic acid (11.57 g, 66.8 mmol) was taken in water (45 mL) and concd H_2SO_4 (15 mL) mixture and cooled to 0 °C. A solution of $NaNO_2$ (6 g, 86.8 mmol) in water (30 mL) was added dropwise to it and the reaction mixture was stirred at 0 °C for 30 min. A cooled solution of NaN_3 (8.68 g, 133.6 mmol) in water (30 mL) was added and the reaction mixture was stirred over a period of 12 h. The resulting precipitate was collected, washed with cold water (10 mL) and dried under vacuo to give 2-azidobenzenesulfonic acid (6.64 g, 50%).

*4.2.1. 2-Azido-N-phenylbenzenesulfonamide (**1a**).* To a suspension of 2-azidobenzenesulfonic acid (500 mg, 2.510 mmol) in dichloromethane (30 mL) were added 2 M oxalyl chloride in CH_2Cl_2 (6.280 mmol) and four drops of DMF. The resulting mixture was heated under reflux for 3 h and then evaporated under reduced pressure. Then this 2-azidobenzenesulfonyl chloride was added in small portions to a vigorously stirred mixture of the aniline (350 mg, 3.760 mmol) and NaOAc (312 mg, 3.760 mmol) in 50% aq MeOH (15 mL) over a period of 30 min. The mixture was then stirred at 60 °C for 1 h, cooled to room temperature, diluted with water (30 mL) and acidified to pH 2 with concd HCl. The precipitate was filtered off, washed thoroughly with water and recrystallized from EtOAc to afford 571 mg of grey solid product **1a** (83%) [Found: C, 52.71; H, 3.86; N, 20.18%. $C_{12}H_{10}N_4O_2S$ requires C, 52.54; H, 3.67; N, 20.43%]. R_f (15% EtOAc/Pet) 0.48; mp 255–257 °C; IR (KBr): 1155, 1472, 2130, 3254 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=7.05–7.12 (m, 4H, ArH), 7.16–7.24 (m, 3H, ArH), 7.28 (s, 1H, NH), 7.53 (t, J=8.0 Hz, 1H, ArH), 7.90 (d, J=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=119.4, 121.2, 124.8, 125.5, 128.9, 129.3, 131.3, 134.4, 136.1, 137.7; MS (ESI): m/z found 275 (MH^+), $C_{12}H_{10}N_4O_2S$ requires 274.

*4.2.2. 2-Azido-N-(4-chlorophenyl)benzenesulfonamide (**1b**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 450 mg of *p*-chloroaniline, 627 mg of compound **1b** was isolated as gummy liquid (81%). R_f (15% EtOAc/Pet) 0.51; IR (neat): 1160, 1331, 2136, 3251 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=7.04–7.07 (m, 3H, ArH), 7.17–7.19 (m, 2H, ArH), 7.22 (s, 1H, NH), 7.27 (d, J=8.0 Hz, 1H, ArH), 7.56 (t, J=7.6 Hz, 1H, ArH), 7.88 (d, J=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=118.5, 121.8, 124.8, 125.4, 129.0, 129.1, 129.5, 132.5, 133.0, 136.5. 136.9; HRMS-ESI m/z found 331.0042 (MNa^+), $C_{12}H_9ClN_4O_2SNa$ requires 331.0032.

*4.2.3. 2-Azido-N-p-tolylbenzenesulfonamide (**1c**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 403 mg of *p*-methylaniline, 637 mg of compound **1c** was isolated as white solid (88%); [Found: C, 53.91; H, 4.12; N, 19.61%. $C_{13}H_{12}N_4O_2S$ requires C, 54.15; H, 4.20; N, 19.43%]. R_f (15% EtOAc/Pet) 0.47; mp 138–140 °C; IR (KBr): 1159, 1331, 2134, 3263 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=2.23 (s, 3H, CH_3), 6.98–7.02 (m, 5H, NH and ArH overlapped), 7.14 (dt, J=8.4, 0.8 Hz, 1H, ArH), 7.26 (d, J=8.8 Hz, 1H, ArH), 7.52 (dt, J=8.4, 1.6 Hz, 1H, ArH), 7.86 (dd, J=8.0, 1.2 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=20.8, 119.4, 121.9, 124.8, 129.0, 129.9, 131.3, 133.4, 134.3, 135.6, 137.6.

*4.2.4. 2-Azido-N-(4-methoxyphenyl)benzenesulfonamide (**1d**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 463 mg of *p*-methoxyaniline, 649 mg of compound **1d** was isolated as grey solid (85%); [Found: C, 51.03; H, 4.10; N, 18.57%. $C_{13}H_{12}N_4O_3S$ requires C, 51.31; H, 3.97; N, 18.41%]. R_f (15% EtOAc/Pet) 0.48; mp 116–118 °C; IR (KBr): 1163, 1511, 2139, 3277 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=3.72 (s, 3H, OCH_3), 6.72 (dd, J=6.8, 2.0 Hz, 2H, ArH), 6.87 (s, 1H, NH), 7.01 (dd, J=7.6, 2.0 Hz, 2H, ArH), 7.14 (t, J=7.6 Hz, 1H, ArH), 7.29 (d, J=8.0 Hz, 1H, ArH), 7.54 (dt, J=8.0, 1.6 Hz, 1H, ArH), 7.80 (dd, J=8.0, 1.6 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=55.4, 114.5, 119.3, 124.7, 124.8, 124.9, 128.6, 129.0, 131.3, 134.2, 137.6.

*4.2.5. 2-Azido-N-benzylbenzenesulfonamide (**1e**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 403 mg of benzylamine, 579 mg of compound **1e** was isolated as light tan solid (80%); [Found: C, 53.81; H, 4.06; N, 19.65%. $C_{13}H_{12}N_4O_2S$ requires C, 54.15; H, 4.20; N, 19.43%]. R_f (15% EtOAc/Pet) 0.50; mp 80–82 °C; IR (KBr): 1165, 1327, 2142, 3292 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=4.10 (d, J=6.0 Hz, 2H, NCH_2), 5.28 (br s, 1H, NH), 7.16–7.23 (m, 7H, ArH), 7.55 (t, J=7.2 Hz, 1H, ArH), 7.98 (d, J=7.6 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=47.6, 119.3, 124.8, 127.9, 128.0, 128.4, 128.5, 130.0, 130.6, 134.0, 136.0, 137.5.

*4.2.6. 2-Azido-N-methylbenzenesulfonamide (**1f**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 10 mL methylamine solution, 442 mg of compound **1f** was isolated as colourless solid (83%); [Found: C, 39.86; H, 4.05; N, 26.17%. $C_7H_8N_4O_2S$ requires C, 39.62; H, 3.80; N, 26.40%]. R_f (15% EtOAc/Pet) 0.53; mp 136–138 °C; IR (KBr): 1161, 1326, 2142, 3305 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=2.61 (d, J=5.2 Hz, 3H, CH_3), 4.94 (d, J=4.8 Hz, 1H, NH), 7.27 (d, J=8.0 Hz, 1H, ArH), 7.31 (d, J=8.4 Hz, 1H, ArH), 7.60 (dt, J=8.8, 0.8 Hz, 1H, ArH), 7.99 (d, J=7.6 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=29.4, 119.4, 124.9, 128.4, 131.1, 134.1, 137.6.

*4.2.7. 2-Azidobenzenesulfonamide (**1g**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 10 mL liquor ammonia, 398 mg of compound **1g** was isolated as grey solid (80%); [Found: C, 36.58; H, 3.37; N, 28.10%. $C_6H_6N_4O_2S$ requires C, 36.36; H, 3.05; N, 28.27%]. R_f (20% EtOAc/Pet) 0.48; mp 176 °C decomposed; IR (KBr): 1163, 1385, 2133, 3360, 3364 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=5.12 (s, 2H, NH), 7.28–7.33 (m, 2H, ArH), 7.60 (dt, J=8.8, 1.2 Hz, 1H, ArH), 7.99 (dd, J=9.2, 1.2 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=118.5, 124.1, 125.2, 132.6, 133.0, 137.6.

*4.2.8. 2-Azido-N-(naphthalen-1-yl)benzenesulfonamide (**1h**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 538 mg of 1-napthylamine,

643 mg of compound **1h** was isolated as grey solid (79%); [Found: C, 59.41; H, 3.55; N, 16.99%. $C_{16}H_{12}N_4O_2S$ requires C, 59.25; H, 3.73; N, 17.27%]. R_f (15% EtOAc/Pet) 0.60; mp 128–130 °C; IR (KBr): 1163, 1319, 2132, 3340 cm^{-1}; ^1H NMR (400 MHz, DMSO): δ_H=7.13 (t, *J*=7.6 Hz, 1H, ArH), 7.21–7.30 (m, 3H, ArH), 7.33 (s, 1H, NH), 7.48–7.58 (m, 3H, ArH), 7.66 (d, *J*=7.6 Hz, 1H, ArH), 7.80 (d, *J*=8.0 Hz, 1H, ArH), 7.85 (d, *J*=8.0 Hz, 1H, ArH), 8.15 (d, *J*=8.4 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=119.5, 120.8, 121.7, 124.8, 125.3, 126.5, 126.7, 127.0, 128.4, 128.8, 129.5, 131.1, 131.4, 134.2, 134.3, 137.9.

*4.2.9. 2-Azido-N-(2-oxo-2H-chromen-6-yl)benzenesulfonamide (**1i**).* Using the general procedure (as **1a**) starting from 500 mg of 2-azidobenzenesulfonic acid and 605 mg of 6-aminocoumarine, 773 mg of compound **1i** was isolated as brown solid (90%); [Found: C, 52.78; H, 3.15; N, 16.11%. $C_{15}H_{10}N_4O_4S$ requires C, 52.63; H, 2.94; N, 16.37%]. R_f (30% EtOAc/Pet) 0.50; mp 194–196 °C decomposed; IR (KBr):1160, 1310, 2128, 1711, 3282 cm^{-1}; ^1H NMR (400 MHz, DMSO): δ_H=6.43 (d, *J*=9.6 Hz, 1H, =CH of coumarin), 7.26–7.35 (m, 3H, ArH), 7.43 (d, *J*=2.4 Hz, 1H, ArH), 7.52 (d, *J*=8.0 Hz, 1H, ArH), 7.64 (t, *J*=7.6 Hz, 1H, ArH), 7.84 (d, *J*=8.0 Hz, 1H, ArH), 8.00 (d, *J*=9.6 Hz, 1H, =CH of coumarin), 10.48 (s, 1H, NH); ^{13}C NMR (100 MHz, $CDCl_3$+DMSO): δ_C=116.9, 117.3, 118.7, 119.0, 119.8, 124.4, 124.6, 129.0, 131.0, 133.8, 134.3, 137.8, 143.2, 150.6, 160.4.

4.3. General procedure for the preparation of compound 3a–k

The substrate, 2-azidosulfonamide 1 (1 equiv) was dissolved in a minimum amount of chloroform, added to a sealed tube and basic alumina (500 mg) was added to it. The organic solvent was evaporated to dryness under reduced pressure. To the residue, propargyl bromide **2** (1.5 equiv) was added. The solid mixture was then stirred at room temperature for an additional 10–15 min to ensure efficient mixing. The sealed tube was then fitted with a stopper, and the mixture was subjected to irradiation in a microwave reactor (CEM, Discover, USA) at 80 °C (external sensor type, 150 W) for 10 min (as monitored by TLC). After cooling, ethyl acetate was added and the slurry stirred at room temperature for 10 min. The mixture was then vacuum filtered through a sintered glass funnel. The filtrate was evaporated to dryness under reduced pressure and the residue was passed through a column to obtain the solid product **3**.

*4.3.1. Compound **3a**.* Using the general procedure starting from 150 mg (0.547 mmol) of **1a** and 98 mg (0.820 mmol) of **2a**, 157 mg of title compound **3a** was isolated as colourless solid (92%). R_f (50% EtOAc/Pet) 0.48; mp 222–224 °C; IR (KBr): 1172, 1347, 1488 cm^{-1}; ^1H NMR (400 MHz, DMSO): δ_H=5.06 (s, 2H, NCH_2), 7.23 (d, *J*=7.2 Hz, 2H, ArH), 7.36–7.41 (m, 3H, ArH), 7.75 (t, *J*=7.6 Hz, 1H, ArH), 7.85 (dd, *J*=8.0, 0.8 Hz, 1H, ArH), 7.98–8.02 (m, 2H, ArH), 8.20 (d, *J*=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$+DMSO): δ_C=44.2, 125.5, 127.0, 127.6, 128.3, 129.5, 129.9, 131.2, 133.0, 133.2, 134.1, 135.2, 140.6; HRMS-ESI *m*/*z* found 313.0753 (MH^+), $C_{15}H_{12}N_4O_2S$ requires 313.0751 (MH^+).

*4.3.2. Compound **3b**.* Using the general procedure starting from 150 mg (0.486 mmol) of **1b** and 87 mg (0.729 mmol) of **2a**, 158 mg of title compound **3b** was isolated as colourless solid (94%). R_f (50% EtOAc/Pet) 0.52; mp 154–156 °C; IR (KBr): 1177, 1350, 1487 cm^{-1}; ^1H NMR (200 MHz, $CDCl_3$): δ_H=4.98 (s, 2H, NCH_2), 7.01 (dd, *J*=6.6, 2.0 Hz, 2H, ArH), 7.21–7.27 (m, 1H, ArH), 7.43 (dt, *J*=7.6, 1.2 Hz, 1H, ArH), 7.74–7.78 (m, 2H, ArH), 7.82 (dd, *J*=7.8, 1.6 Hz, 2H, ArH), 8.27 (dd, *J*=8.2, 1.2 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=46.3, 125.4, 127.9, 128.6, 129.2, 129.9, 131.2, 132.9, 133.3, 133.4, 134.4, 134.8, 138.8; HRMS-ESI *m*/*z* found 347.0376 (MH^+), 349.0340 (MH^++2), $C_{15}H_{11}ClN_4O_2S$ requires 347.0361 (MH^+), 349.0361 (MH^++2).

*4.3.3. Compound **3c**.* Using the general procedure starting from 150 mg (0.486 mmol) of **1b** and 97 mg (0.729 mmol) of **2b**, 157 mg of title compound **3c** was isolated as colourless solid (90%). R_f (50% EtOAc/Pet) 0.57; mp 160–162 °C; IR (KBr): 1175, 1350, 1483 cm^{-1}; ^1H NMR (200 MHz, $CDCl_3$): δ_H=2.40 (s, 3H, CH_3), 4.91 (s, 2H, NCH_2), 7.04 (d, *J*=8.6 Hz, 2H, ArH), 7.24 (dd, *J*=8.8, 3.0 Hz, 2H, ArH), 7.46 (t, *J*=7.6 Hz, 1H, ArH), 7.74–7.87 (m, 2H, ArH), 8.30 (d, *J*=8.0 Hz, 1H, ArH); ^{13}C NMR (50 MHz, $CDCl_3$): δ_C=10.4, 46.4, 125.1, 127.8, 128.5, 128.8, 129.0, 129.8, 130.9, 133.6, 134.2, 134.8, 138.9, 141.8; HRMS-ESI *m*/*z* found 361.0532 (MH^+), 363.0532 (MH^++2), $C_{16}H_{13}ClN_4O_2S$ requires 361.0518 (MH^+), 363.0518 (MH^++2).

*4.3.4. Compound **3d**.* Using the general procedure starting from 150 mg (0.520 mmol) of **1c** and 93 mg (0.780 mmol) of **2a**, 151 mg of title compound **3d** was isolated as grey solid (89%). R_f (50% EtOAc/Pet) 0.54; mp 196–198 °C; IR (KBr): 1169, 1346, 1487 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=2.30 (s, 3H, CH_3), 4.98 (s, 2H, NCH_2), 6.98 (d, *J*=8.0 Hz, 2H, ArH), 7.09 (d, *J*=8.0 Hz, 2H, ArH), 7.53 (dt, *J*=7.6, 0.8 Hz, 1H, ArH), 7.72–7.77 (s, 1H, ArH), 7.80 (dd, *J*=8.0, 1.6 Hz, 1H, ArH), 7.87 (dd, *J*=8.0, 1.6 Hz, 1H, ArH), 8.28 (dd, *J*=8.0, 0.4 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=21.1, 46.5, 125.2, 126.5, 128.6, 129.0, 130.3, 131.7, 133.3, 133.4, 134.5, 137.7, 138.8; HRMS-ESI *m*/*z* found 327.0926 (MH^+), $C_{16}H_{14}N_4O_2S$ requires 327.0916 (MH^+).

*4.3.5. Compound **3e**.* Using the general procedure starting from 150 mg (0.520 mmol) of **1c** and 104 mg (0.780 mmol) of **2b**, 152 mg of title compound **3e** was isolated as colourless solid (86%). R_f (50% EtOAc/Pet) 0.58; mp 108–110 °C; IR (KBr): 1171, 1350, 1486 cm^{-1}; ^1H NMR (200 MHz, $CDCl_3$): δ_H=2.30 (s, 3H, CH_3), 2.39 (s, 3H, CH_3), 4.90 (s, 2H, NCH_2), 6.97 (d, *J*=6.4 Hz, 2H, ArH), 7.07 (d, *J*=8.2 Hz, 2H, ArH), 7.45 (dt, *J*=7.6, 1.0 Hz, 1H, ArH), 7.72 (dt, *J*=8.4, 1.6 Hz, 1H, ArH), 7.84 (dd, *J*=7.8, 1.4 Hz, 1H, ArH), 8.31 (d, *J*=8.2 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=10.4, 21.0, 46.7, 125.0, 126.4, 128.6, 128.7, 129.4, 130.2, 131.3, 133.7, 134.4, 137.8, 138.7, 141.6; HRMS-ESI *m*/*z* found 341.1074 (MH^+), $C_{17}H_{16}N_4O_2S$ requires 341.1072 (MH^+).

*4.3.6. Compound **3f**.* Using the general procedure starting from 150 mg (0.493 mmol) of **1d** and 88 mg (0.739 mmol) of **2a**, 153 mg of title compound **3f** was isolated as colourless solid (91%). R_f (50% EtOAc/Pet) 0.56; mp 152–154 °C; IR (KBr): 1168, 1352, 1487 cm^{-1}; ^1H NMR (200 MHz, $CDCl_3$): δ_H=3.77 (s, 3H, OCH_3), 4.97 (s, 2H, NCH_2), 6.78 (dd, *J*=6.6, 2.2 Hz, 2H, ArH), 7.00 (dd, *J*=6.8, 2.2 Hz, 2H, ArH), 7.50 (dt, *J*=7.6, 1.0 Hz, 1H, ArH), 7.77 (s, 1H, ArH), 7.80 (dd, *J*=7.8, 1.6 Hz, 1H, ArH),7.87 (dd, *J*=7.8, 1.4 Hz, 1H, ArH), 8.29 (dd, *J*=8.2, 1.0 Hz, 1H, ArH); ^{13}C NMR (50 MHz, $CDCl_3$): δ_C=46.8, 55.7, 115.0, 125.5, 128.4, 128.7, 129.3, 132.0, 133.2, 133.4, 133.6, 134.7, 159.8; HRMS-ESI *m*/*z* found 343.0858 (MH^+), $C_{16}H_{14}N_4O_3S$ requires 343.0865 (MH^+).

*4.3.7. Compound **3g**.* Using the general procedure starting from 150 mg (0.520 mmol) of **1e** and 93 mg (0.780 mmol) of **2a**, 148 mg of title compound **3g** was isolated as colourless solid (87%). R_f (50% EtOAc/Pet) 0.60; mp 126–128 °C; IR (KBr): 1163, 1334, 1474 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=4.33 (s, 2H, NCH_2), 4.34 (s, 2H, NCH_2), 7.28–7.30 (m, 2H, ArH), 7.32–7.35 (m, 3H, ArH), 7.60–7.64 (m, 2H, ArH), 7.79 (dt, *J*=8.4, 0.8 Hz, 1H, ArH), 8.09 (dd, *J*=8.0, 0.8 Hz, 1H, ArH), 8.22 (dd, *J*=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=41.2, 54.2, 125.0, 128.3, 128.6, 128.6, 129.0, 129.2, 131.6, 132.8, 133.0, 133.9, 134.0, 134.4; HRMS-ESI *m*/*z* found 327.0924 (MH^+), $C_{16}H_{14}N_4O_2S$ requires 327.0916 (MH^+).

*4.3.8. Compound **3h**.* Using the general procedure starting from 150 mg (0.707 mmol) of **1f** and 126 mg (1.060 mmol) of **2a**, 168 mg of title compound **3h** was isolated as colourless solid (95%). R_f (50%

EtOAc/Pet) 0.55; mp 110–112oC; IR (KBr): 1166, 1342, 1488 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=2.89 (s, 3H, NCH_3), 4.43 (s, 2H, NCH_2), 7.62 (dt, *J*=7.6, 0.8 Hz, 1H, ArH), 7.77 (s, 1H, ArH), 7.80 (dt, *J*=8.0, 1.2 Hz, 1H, ArH), 8.06 (dd, *J*=8.0, 1.2 Hz, 1H, ArH), 8.23 (d, *J*=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, $CDCl_3$): δ_C=38.0, 44.0, 125.0, 128.8, 129.4, 130.1, 132.6, 133.2, 134.0, 134.6; HRMS-ESI *m/z* found 251.0609 (MH^+), $C_{10}H_{10}N_4O_2S$ requires 251.0603 (MH^+).

4.3.9. Compound ***3i***. Using the general procedure starting from 150 mg (0.757 mmol) of **1g** and 136 mg (1.135 mmol) of **2a**, 182 mg of title compound **3i** was isolated as colourless solid (88%). R_f (50% EtOAc/Pet) 0.45; mp 180–182 °C; IR (KBr): 1166, 1343, 1488 cm^{-1}; ^1H NMR (400 MHz, DMSO): δ_H=3.19 (s, 1H, acetylenic CH), 4.22 (d, *J*=2.0 Hz, 2H, NCH_aH_b), 4.53 (s, 2H, NCH_2), 7.78 (t, *J*=7.6 Hz, 1H, ArH), 7.97 (s, 1H, ArH),7.99 (d, *J*=7.6 Hz, 1H, ArH), 8.05 (t, *J*=7.6 Hz, 2H, ArH); ^{13}C NMR (100 MHz, DMSO): δ_C=30.2, 40.2, 75.8, 77.4, 125.0, 128.0, 129.9, 130.9, 132.6, 132.8, 134.6, 135.2; HRMS-ESI *m/z* found 275.0612 (MH^+), $C_{12}H_{10}N_4O_2S$ requires 275.0603 (MH^+).

4.3.10. Compound ***3j***. Using the general procedure starting from 150 mg (0.462 mmol) of **1h** and 83 mg (0.693 mmol) of **2a**, 139 mg of title compound **3j** was isolated as colourless solid (83%). R_f (50% EtOAc/Pet) 0.50; mp 204–206 °C; IR (KBr): 1180, 1360, 1484 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ_H=4.69 (d, *J*=14.8 Hz, 1H, NCH_aH_b), 5.20 (d, *J*=14.8 Hz, 1H, $NCH_aH_\mathbf{b}$), 7.03 (d, *J*=7.6 Hz, 1H, ArH), 7.34 (t, *J*=7.6 Hz, 1H, ArH), 7.55–7.65 (m, 3H, ArH), 7.80 (s, 1H, ArH), 7.87 (dt, *J*=7.6, 0.8 Hz, 3H, ArH), 7.97 (dd, *J*=7.6, 1.2 Hz, 1H, ArH), 8.20 (d, *J*=8.0 Hz, 1H, ArH), 8.31 (dd, *J*=8.0, 0.8 Hz, 1H, ArH); ^{13}C NMR (100 MHz, DMSO): δ_C=44.2, 122.8, 125.0, 125.6, 125.9, 126.8, 127.3, 127.4, 128.2, 129.2, 130.2, 131.6, 131.7, 132.7, 133.0, 134.1, 134.5, 135.3, 137.3; HRMS-ESI *m/z* found 363.0921 (MH^+), $C_{19}H_{14}N_4O_2S$ requires 363.0916 (MH^+).

4.3.11. Compound ***3k***. Using the general procedure starting from 150 mg (0.438 mmol) of **1i** and 78 mg (0.657 mmol) of **2a**, 141 mg of title compound **3k** was isolated as colourless solid (85%). R_f (60% EtOAc/Pet) 0.48; mp 272–274 °C; IR (KBr): 1174, 1354, 1485, 1724 cm^{-1}; ^1H NMR (400 MHz, DMSO): δ_H=5.09 (s, 2H, NCH_2), 6.54 (d, *J*=9.6 Hz, 1H, =CH of coumarin), 7.45–7.47 (m, 2H, ArH), 7.72–7.78 (m, 2H, ArH), 7.86 (d, *J*=8.0 Hz, 1H, ArH), 8.01–8.06 (m, 3H, ArH and =CH of coumarin overlapped), 8.21 (d, *J*=8.0 Hz, 1H, ArH); ^{13}C NMR (100 MHz, DMSO): δ_C=43.9, 117.2, 117.5, 119.2, 125.6, 127.2, 127.7, 130.1, 130.6, 130.9, 132.8, 133.1, 134.2, 135.4, 136.4, 143.4, 152.8, 159.5; HRMS-ESI *m/z* found 381.0655 (MH^+), $C_{18}H_{12}N_4O_4S$ requires 381.0658 (MH^+).

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