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Experimental and theoretical studies on methanesulfonic acid 1-methylhydrazide: Antimicrobial activities of its sulfonyl hydrazone derivatives

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ABSTRACT

Methanesulfonic acid 1-methylhydrazide (*msmh*) and its sulfonyl hydrazone derivatives, salicylaldehyde-*N*-methylmethanesulfonylhydrazone (*salmsmh*) and 2-hydroxy-1-naphthaldehyde-*N*-methylmethanesulfonylhydrazone (*nafmsmh*) were synthesized and characterized by using FT-IR, ^{1}H NMR, ^{13}C NMR, LC–MS and elemental analysis. Conformation analysis of *msmh* based on DFT/B3LYP/6-311G(d) method was performed. ^{1}H and ^{13}C shielding tensors of *msmh* for the most stable conformer were calculated with GIAO/DFT/B3LYP/6-311++G(2d, 2p) methods in vacuo and various solvents such as DMSO, THF, acetonitrile, methanol and aqueous solution. The harmonic vibrational wavenumbers for the most stable conformer were calculated using at B3LYP/6-311G(d) level. Antimicrobial activity of the compounds was also screened against Gram-positive bacteria (*Staphylococcus aureus* ATCC 25923, *Bacillus cereus* RSKK 863) and Gram-negative bacteria (*Escherichia coli* ATCC 11230, *Salmonella enteritidis* ATCC 40376, *Pseudomonos aeruginosa* ATCC 28753) by both disc diffusion and micro dilution methods.

1. Introduction

Sulfonamides and sulfonyl hydrazones have been shown to be active in several pharmacological tests, demonstrating antibacterial, antitumor, diuretic, antiviral, antinociceptive activity, specific enzyme inhibition such as carbonic anhydrase, γ-secretase HIV-protease, metalloproteinase, and hormone regulation among others [1,2].

Furthermore, sulfonyl-hydrazine carrying ion-exchange beads were used as an ion-exchange support for adsorption of human serum albumin (HSA) from aqueous solution [3]. Sulfonyl hydrazides grafted onto the nanoparticles are also used as polymerizable foaming agent [4].

Despite to its importance in medicinal and polymeric fields, only a small amount of papers on calculations of sulfonyl hydrazides and their derivatives have been reported so far [5–10]. In our previous study, experimental and theoretical study of methanesulfonic acid hydrazide was reported [11]. Recently, we have published some calculations and experimental data of some sulfonyl hydrazone [12,13] and their metal carbonyl complexes [14,15].

As part of our ongoing studies, the aims of the present work is to study the conformational behavior of *msmh*, as well as its NMR spectra from experimental and theoretical viewpoints as a contribution to the understanding of the rational drug design of sulfonyl hydrazide. Antibacterial activities of *msmh* and its sulfonyl hydrazones obtained by the condensation of salicylaldehyde and naphthaldehyde were also evaluated against Gram-positive bacteria (*Staphylococcus aureus* ATCC 25923, *Bacillus cereus* RSKK 863) and Gram-negative bacteria (*Escherichia coli* ATCC 11230, *Salmonella enteritidis* ATCC 40376, *Pseudomonos aeruginosa* ATCC 28753) by both disc diffusion and micro dilution methods.

2. Experimental

2.1. Physical measurements

The elemental analyses (C, H, N and S) were performed on a LECO-CHSNO-9320 type elemental analyzer. The ^{13}C NMR and ^{1}H NMR spectra were recorded on a Bruker WM-400 spectrometer at 400 MHz and at ambient temperature using d_6-DMSO solutions with TMS as internal reference. IR spectrum was recorded in the range of 4000–400 cm^{-1} as KBr disc with Mattson 1000 FT spectrometer. LC/MS-EI was recorded on AGILENT 1100. TLC was conducted on 0.25 mm silica gel plates (60F254, Merck). Chemicals were obtained from Aldrich and used without further purification.

2.2. Synthesis of msmh

Methanesulfonyl chloride (0.04 mol) in tetrahydrofuran (25 mL) was added methylhydrazine (0.08 mol) in dropwise while

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the temperature was maintained between 268 and 273 K. The mixture was stirred for 24 h (completion of the reaction was monitored by TLC), then solvent was evaporated. The colorless crude compound was purified in THF/*n*-hexane by column chromatography then the product was crystallized from THF/*n*-hexane mixture (3:1). Yield 65%; mp: 115–117 °C. EI-MS (70 eV) m/z: 248.2 ($2\,M^+$), 249.1 $(2\,M+1)^+$, Elemental analysis: Calcd for ($C_2H_8SO_2N_2$): C, 19.35; H, 6.45; N, 22.50; S, 22.80. Found: C, 19.45; H, 6.34; N, 22.45; S, 22.61.

2.3. Synthesis of salmsmh

Ethanolic solution of methanesulfonic acid 1-methylhydrazide (0.65 g, 5.24 mmol) was added dropwise to an ethanolic solution of salicylaldehyde (0.65 mL g, 6.25 mmol), maintaining the temperature at about 268 K. Then, the mixture was stirred for 5–8 h at room temperature. The precipitated product was crystallized from ethanol/*n*-hexane (3:1) mixture. The yellow crystalline solid was dried in vacuo and stored at ethanol/*n*-hexane vapour. Yield 60%, mp 110–111 °C. IR (KBr) cm^{-1}: 1622 υ(C=N), 1268 υ(C—O), 1314 $\upsilon_{as}(SO_2)$, 1152 $\upsilon_s(SO_2)$; ^1H NMR (d_6-DMSO) δ: 2.31 ppm(t, 3H, CH_3), 5.02 ppm (s, 3H, N—CH_3), 8,69 ppm (s, 1H, N=CH), 11.35 ppm (s, 1H, OH), 6.94–7.36 ppm (m, ArH); ^{13}C NMR(d_6-DMSO) δ: 40.22 ppm (CH_3), 40.01 ppm (N=CH_3), 164.90 ppm (N=CH); 117.25–132.55 ppm (ArC); EI-MS (70 eV) m/z: 228 (M^+, 11.4%, M + 1, 3.7%); Anal. Calcd. for $C_9H_{12}SO_3N_2$: C, 47.4; H, 5.30; N, 12.30; S, 14.03. Found: C, 47.6; H, 5.34; N, 12.37; S, 13.98.

2.4. Synthesis of nafmsmh

Ethanolic solution of methanesulfonic acid 1-methylhydrazide (0.32 g, 2.58 mmol) was added dropwise to an ethanolic solution of 2-hydroxy-1-naphthaldehyde (0.66 g, 3.8 mmol), maintaining the temperature at about 268 K. Then, the mixture was stirred for 5 h at room temperature. The precipitated product was crystallized from ethanol/*n*-hexane (4:1) mixture. The yellow crystalline solid was dried in vacuo and stored at ethanol/*n*-hexane vapour. Yield 65%; mp: 132–133 °C. IR (KBr) cm^{-1}: 1621 υ(C=N), 1240 υ(C—O), 1320 $\upsilon_{as}(SO_2)$, 1165 $\upsilon_s(SO_2)$; ^1H NMR (d_6-DMSO) δ: 3.13 ppm (t, 3H, CH_3), 3,45 ppm (s, 3H, N—CH_3), 8,80 ppm (s, 1H, N=CH), 11.85 ppm (s, 1H, OH), 7.23–8.58 ppm (m, ArH); ^{13}C NMR (d_6-DMSO) δ: 35.69 ppm (CH_3), 33.27 ppm (N=CH_3), 157,61 ppm (N=CH); 109.68–144.08 ppm (ArC); EI-MS (70 eV) m/z: 278 (M^+, 10.1%, M + 1, 5.7%); Anal. Calcd. for $C_{13}H_{14}SO_3N_2$: C, 56.12; H, 5.03; N, 10.07; S, 11.51. Found: C, 56.16; H, 5.06; N, 10.04; S, 11.59.

2.5. Theoretical calculations

An extensive search for low energy conformations on the potential energy surface (PES) of (*msmh*) was carried out by using a systematic search with DFT/B3LYP/6-311G(d) levels. Vibration frequencies calculated ascertain the structure was characterized to be the stable structure (no imaginary frequencies). For the NMR calculations, the optimized geometry at DFT/B3LYP/6-311++G(2d, 2p) level in gas phase was used. ^1H and ^{13}C NMR chemical shifts were calculated by GIAO method at DFT/B3LYP/6-311++G(2d, 2p) level. The ^1H and ^{13}C shieldings were converted into the predicted chemical shifts using TMS values, calculated at the same level of theory. After being optimized at DFT/B3LYP/6-31G(d) level, vibrational wavenumbers of the most stable conformer were calculated at the same level. All calculations were performed on a Pentium IV computer with the default convergence criteria and using Gaussian 03 software package [16].

2.6. Determination of MIC for the bacteria

E. coli ATCC 11230, *P. aeruginosa* ATCC 28753, *S. enteritidis* ATCC 40376, *S. aureus* ATCC 25923, *B. cereus* RSKK 863 cultures were obtained from Gazi University, Biology Department and Refik Saydam Hygiene Center Culture Collection. Bacterial strains were cultured overnight at 310 K in Nutrient Broth. These stock cultures were stored in the dark at 277 K during the survey.

The antibacterial activity of the compounds was examined by determination the minimum inhibitory concentration (MIC) in accordance with CLNS methodology (NCCLS) [17,18]. All tests were performed in Nutrient Broth supplemented with dimethyl sulfoxide at a final concentration of 10% (v/v) to enhance their solubility. Bacterial strains were cultured overnight at 310 K in Nutrient Broth. Briefly, test strains were suspended in Nutrient Broth by adjusting to 0.5 McFarland. The test compounds dissolved in dimethyl sulfoxide (DMSO) were first diluted to the highest concentration (1000 μg mL^{-1}) to be tested, and then serial twofold dilutions were made in a concentration range from 15.6 to 1000 μg mL^{-1} in 10 mL sterile test tubes containing nutrient broth. MIC values of compounds bacterial strains were determined based on a micro-well dilution method with some modifications. Then 96-well plates were prepared by dispensing into each well 95 μL of nutrient broth and 5 μL of the inoculums. 100 μL from test compounds initially prepared at the concentration of 1000 μg mL^{-1} was added into the first wells. Then, 100 μL from their serial dilutions was transferred into 13 consecutive wells. The test compounds in this study were screened two times against each micro organism. The MIC values were determined from visual examinations as beings the lowest concentration of the extracts in the wells with no bacterial growth [19].

2.7. Disc diffusion method

Bacterial susceptibility testing was performed by the disc diffusion method according to the guidelines of Clinical and Laboratory Standards Institute (CLSI) [20]. The sterilized (autoclaved at 393 K for 30 min), liquefied Mueller Hinton agar (313–323 K) was inoculated with the suspension of the micro organism (matched to 0.5 McFarland) and poured into a Petri dish to give a depth of 3–4 mm. The paper discs impregnated with the test compounds (60 μg) were placed on the solidified medium. Discs were placed on agar plates and the cultures were incubated at 310 K for 24 h for bacteria. Inhibition zones formed on the medium were evaluated in mm. Ampicillin was chosen as a standard in antibacterial activity measurements (positive control). DMSO poured disc was used as negative control.

3. Results and discussion

3.1. Structure analysis

The numbering scheme and molecular geometry of *msmh* are presented in Fig. 1. One-dimensional potential energy scans were performed for three torsion angles-τ_1 O2—S—N1—C1, τ_2 C2—S—N1—C1 and τ_3 C2—S—N1—N2 in the full range of 0–360°, starting with the eclipsed conformation and increasing of 30° at DFT/B3LYP/6-311G(d) (Figs. 2–4), to determine the all the possible conformations in vacuo. As seen in Figs. 2–4, the energy profiles of τ_1 shows local minima at about 150°, global minima at 210°, local maxima at about 180°, 300° and global maxima at 60°. Second scan for τ_2 shows two local minima at 90° and 270°, global minima at 330°, local maxima at 30°, 300° and global maxima at 180°. Final scan for τ_3 shows two local minima at about 120° and 270°, global minima at about 360°, two local maxima at 60° and 300°, global maxima at

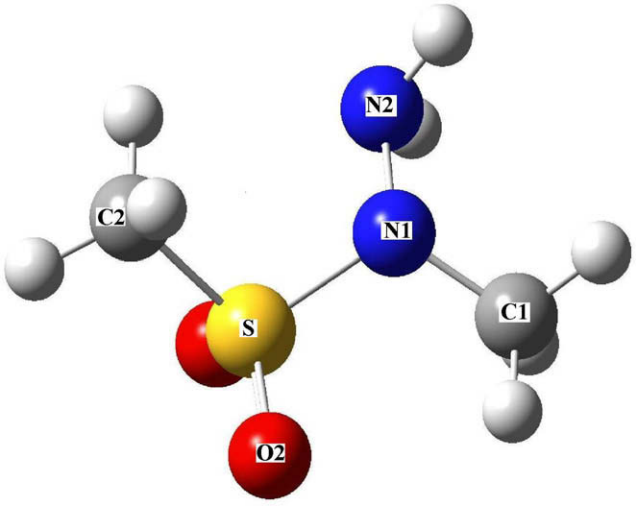

Fig. 1. Optimized geometry of *msmh*.

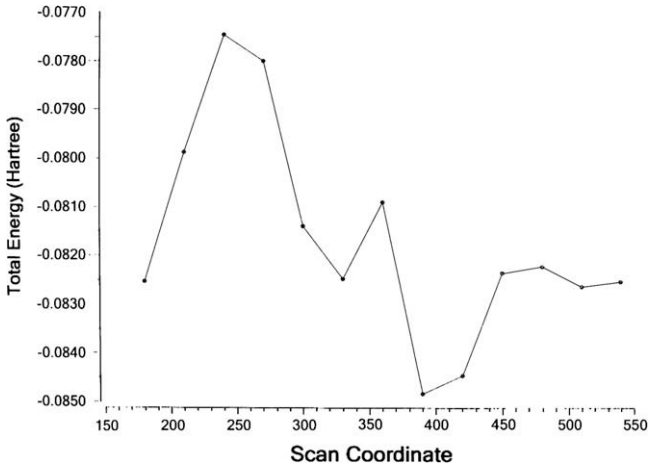

Fig. 2. Energy profile for the rotation about O2—S—N1—C1 dihedral angle.

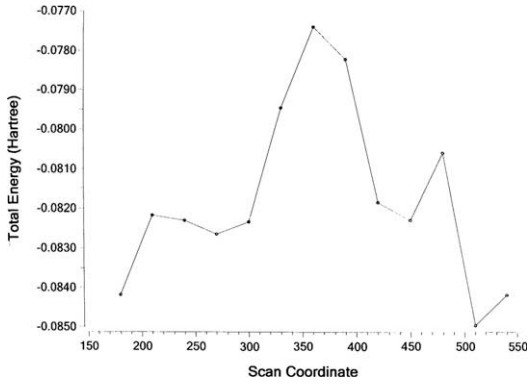

Fig. 3. Energy profile for the rotation about C2—S—N1—C1 dihedral angle.

about 210°. After calculation, four conformers were characterized for *msmh*. The structure of optimized geometries of the conformations is illustrated in Fig. 5. Total and relative energies of the conformers and the relative contribution of every conformation at 298.15 K according to the Maxwell–Boltzmann statistical average are listed in Table 1. The gauche conformation about the S—N bond, indicated by the torsional angle τ_3, is important for the energy minimum of the *msmh*

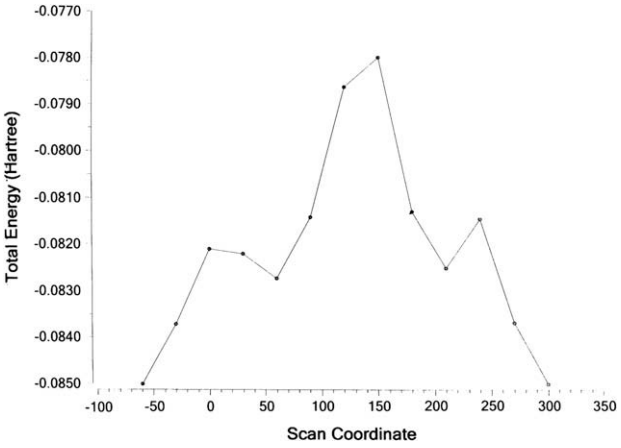

Fig. 4. Energy profile for the rotation about C2—S—N1—N2 dihedral angle.

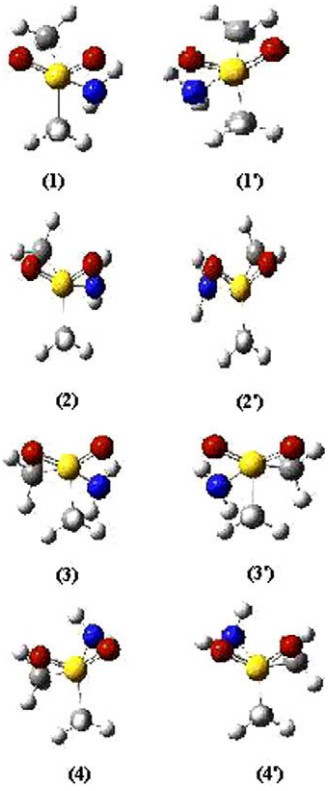

Fig. 5. Optimized (DFT/B3LYP/6-311G(d)) geometries of conformations.

conformers. The conformer **1** (or **1**′) is the most stable form. Energy difference between conformers **1** and **2** is 0.38 kcal/mol. The principal differences between the two conformers lie in the orientation of hydrogen atom of NH_2 moiety with respect to SO_2 group. The percent-

Table 1
Total energies calculated with DFT/B3LYP/6-311G(d) basis set of the conformers.

Conformers	τ_1	τ_2	τ_3	Energy (kcal/mol)	ΔE (kcal/mol)
(**1**,**1**′)	48.42	162.51	−67.32	−463842.08	(0.00)
(**2**,**2**′)	31.68	145.14	−86.25	−463841.70	(0.38)
(**3**,**3**′)	161.52	−84.04	54.56	−463839.23	(2.85)
(**4**,**4**′)	−27.99	85.27	−144.04	−463836.39	(5.69)

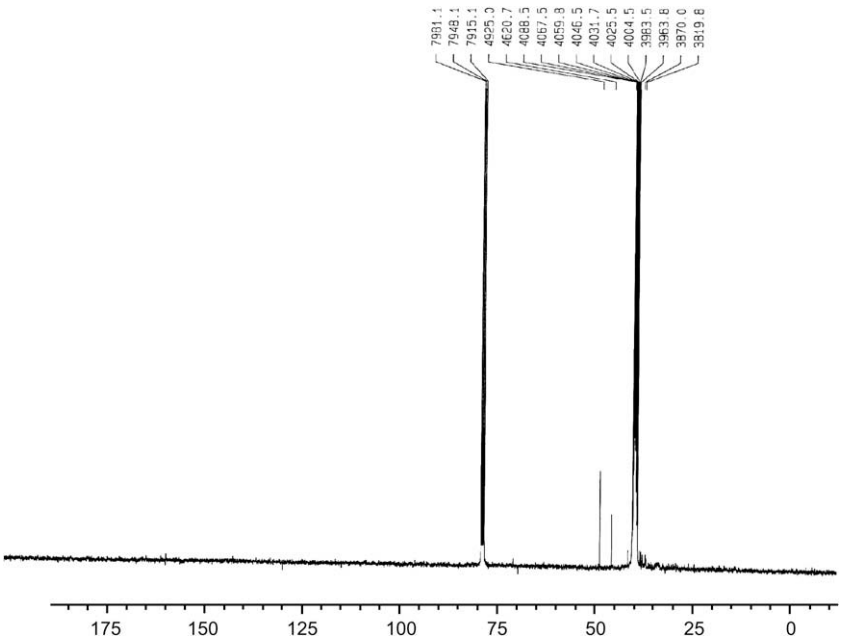

Fig. 6. ^{13}C NMR spectrum of *msmh*.

age distributions for these conformers are 65.16 and 34.31% for (**1**) and (**2**), respectively. The results allow us to predict the presence of first two conformers at the room temperature.

In order to facilitate the interpretation of the NMR spectra, quantum-chemical calculations were performed using B3LYP/6-311G++(2d, 2p) basis set for the most stable conformer in several phases [21]. ^{13}C NMR and ^{1}H spectra of *msmh* are given in Figs. 6 and 7. The experimental and calculated chemical shift values are shown in Table 2. Calculations of the carbon chemical shifts in gas phase gave the following results: 40.22 ppm for *C*—S and 39.80 ppm for *C*—N, which shows good agreement with experimental values (40.32 ppm and 38.49 ppm, respectively). However, the calculated proton shifts in gas phase conformed poorly to the measured proton shifts. The comparison of calculated values at different solvents revealed that good conformity is obtained at DMSO and water phases. This means that solvent effect on the carbon atoms is relatively weak, whereas, on hydrogen atoms are very strong, depending on solvent polarity. Experimental NH_2 proton shift (5.20 ppm) showed poor correlation with the calculated proton shifts (3.50 ppm in DMSO, 4.00 ppm in water).This suggests

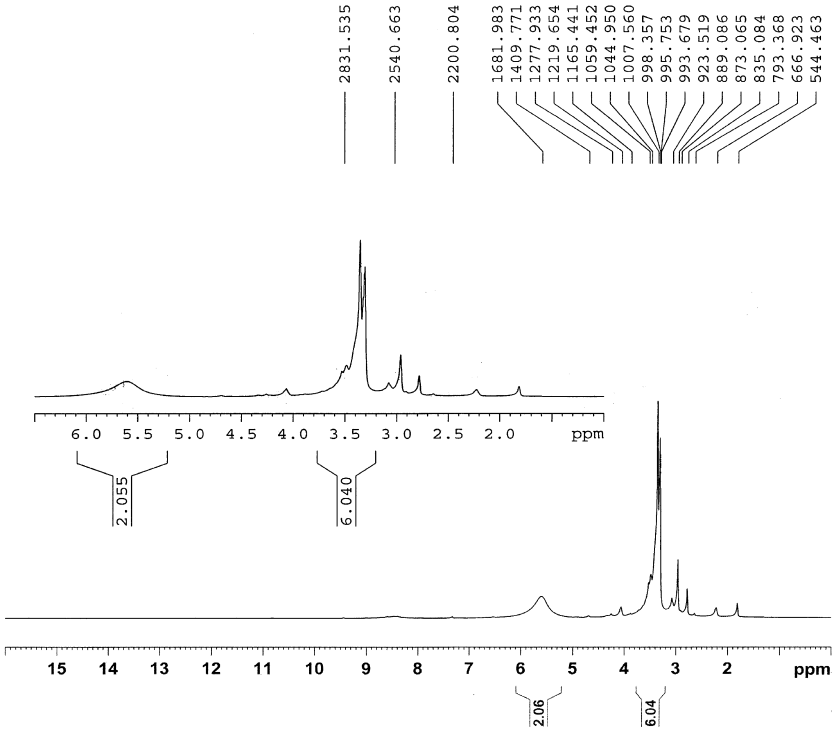

Fig. 7. ^{1}H NMR spectrum of *msmh*.

Table 2
Experimental and calculated ^{13}C and ^{1}H NMR chemical shifts (ppm).

	DFT/B3LYP/6-311++G(2d, 2p)						
	Gas	Aqueous	DMSO	THF	Methanol	Acetonitrile	Exp.
S—*C*	40.32	42.60	41.05	41.36	41.53	41.54	40.22
N—*C*	38.49	39.79	39.28	39.48	39.33	39.33	39.80
S—CH_3	2.73[a]	3.18[a]	2.93[a]	2.90[a]	2.93[a]	2.93[a]	3.20(s)
N—CH_3	2.76[a]	3.29[a]	3.18[a]	2.83[a]	2.84[a]	2.85[a]	3.40(s)
NH_2	2.98[a]	4.00[a]	3.50[a]	3.37[a]	3.51[a]	3.51[a]	5.20(s)

[a] Average values.

that NH_2 protons have both intra molecular and intermolecular hydrogen bonding.

The harmonic and scaled harmonic vibrational wavenumbers of the most stable conformer of *msmh* calculated with B3LYP/6-311G(d) levels and scaled by 0.9739 were given in Table 3, experimental data in solid phase and approximate assignment also given in Table 3. The assignment were made by taking care of the vibrational motions by using Gaussian View and by taking into consideration the data reported in the literature for compounds that contain the same functional fragments [11,22]. The infrared bands of solid substance are generally not coincident in wavenumbers due to intermolecular interactions in the crystal (correlation effects). As seen in Table 3, scaled harmonic frequencies agree with the experimental data, except NH_2 stretching vibrations. The difference between experimental data and calculated anharmonic wavenumbers of NH_2 stretching vibrations may be explained by molecular association with hydrogen bonding in solid state. This comment is in accordance with the data of Ienco et al. [11].

Table 3
The calculated and scaled vibrational wavenumbers (cm^{-1}) of the global conformer (**1**) of *msmh*.

Experimental IR (solid)	DFT/B3LYP/6-311G(d) Harmonic	Approximate assignment Scaled	
3353	3568	3475	$\nu_{as}(NH_2)$
3237	3438	3348	$\nu_{s}(NH_2)$
3125	3194	3110	$\nu_{as}(S—CH_3)$
3110	3175	3092	$\nu_{as}(S—CH_3)$
3080	3155	3072	$\nu_{as}(N—CH_3)$
3025	3095	3014	$\nu_{s}(N—CH_3)$
2980	3078	2997	$\nu_{s}(S—CH_3)$
2934	3002	2923	$\nu_{s}(N—CH_3)$
1697	1720	1675	$\delta(NH_2)$
1490	1517	1477	$\delta_{as}(N—CH_3)$
–	1506	1466	$\delta_{as}(N—CH_3)$
1451	1483	1444	$\delta_{as}(S—CH_3)$
1434	1464	1425	$\delta_{as}(S—CH_3$
1418	1454	1416	$\delta_{s}(N—CH_3)$
1324	1360	1324	$\nu_{as}(SO_2)$
1314	1342	1306	$\omega(NH_2) + \rho(N—CH_3)$
1293	1306	1272	$\nu(CN) + \rho(N—CH3)$
1233	1248	1215	$\rho(NH_2)$
1157	1157	1127	$\nu_{s}(SO_2$
1126	1135	1105	$\delta(NSC)$
–	1119	1089	$\nu(NN) + \delta(CNN)$
994	1017	990	$\delta(SNC) + \rho(N—CH_3)$
978	997	971	$\rho(CH_3) + \delta(HCS)$
–	989	963	$\delta(N—CH_3)$
875	844	822	$\nu(SN)$
753	757	737	$\nu(CS)$
622	618	602	$\rho(N—CH_3) + \delta(SNC)$
523	501	488	$\delta(SO_2)$
–	486	473	$\rho(SO_2)$
455	463	451	$\omega(SO_2)$
409	412	401	$\tau(NH_2)$

as = asymmetric, s = symmetric, δ = bending, ν = stretching, ρ = rocking, τ = twisting, ω = wagging.

3.2. Antibacterial activity

msmh, *salmsmh* and *nafmsmh* were screened for its antibacterial activity against Gram-positive bacteria (*S. aureus* ATCC 25923, *B. cereus* RSKK 863) and Gram-negative bacteria (*E. coli* ATCC 11230, *S. enteritidis* ATCC 40376, *P. aeruginosa* ATCC 28753) by both disc diffusion and micro dilution methods. The antibacterial activity results were presented in Tables 4 and 5. Microbiological results indicated that the synthesized compounds possessed a broad spectrum of activity against the tested microorganisms and showed relatively better activity against Gram-negative as compared to Gram-positive bacteria.

msmh showed the highest activity with lowest MIC values (25 μg/mL, 50 μg/mL) against Gram-negative bacteria *P. aeruginosa, E. coli* and *S. enteritidis*, respectively. However, the sulfonyl hydrazones showed a remarkable decrease in antibacterial activity (100–500 μg/mL) than the parent sulfonyl hydrazide (25–100 μg/mL). The presence of primer amine group in the sulfonyl hydrazide may be responsible for their higher activity in comparison to sulfonyl hydrazones [13,23,24]. Furthermore, antimicrobial activity of *msmh* is higher than aliphatic and aromatic disulfonamides published in the literature [23,24].

4. Conclusion

In this study, energetically favorable conformations of the *msmh* were investigated firstly at B3LYP/6-311G(d) theory level. The obtained results indicated that at room temperature the molecule in electronically ground state has two stable conformers. Harmonic vibrational wavenumber, ^{1}H and ^{13}C shielding tensors of *msmh*

Table 4
Antimicrobial activity of the compounds with micro dilution method.

Bacteria	MIC (μg/mL)		
	msmh	*salmsmh*	*nafmsmh*
E. coli ATCC 11230	50	125	100
P. aeruginosa ATCC 28753	25	125	62.5
S. enteritidis ATCC 40376	50	125	100
S. aureus ATCC 25923	100	250	125
B. cereus RSKK 863	100	500	125

Table 5
Result of the antimicrobial test of compounds (disc potency 60 μg).

Bacteria	Diameter of inhibition zone (mm)			
	msmh	*salmsmh*	*nafmsmh*	Ampicillin (10 μg/disc)
E. coli ATCC 11230	21	10	12	11
P. aeruginosa ATCC 28753	25	10	12	10
S. enteritidis ATCC 40376	20	11	13	10
S. aureus ATCC 25923	17	8	9	16
B. cereus RSKK 863	15	7	10	12

for the most stable conformer were calculated. *msmh* showed the highest antimicrobial activity against Gram-negative bacteria. The sulfonyl hydrazones showed a remarkable decrease in antibacterial activity than the parent sulfonyl hydrazide.

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