Simin Vazirimehr, Abolghasem Davoodnia*, S. Ali Beyramabadi,
Mahboobeh Nakhaei-Moghaddam and Niloofar Tavakoli-Hoseini

Two new pyrrolo[2,3-*d*]pyrimidines (7-deazapurines): ultrasonic-assisted synthesis, experimental and theoretical characterizations as well as antibacterial evaluation

DOI 10.1515/znb-2017-0004
Received January 6, 2017; accepted March 31, 2017

Abstract: Two new pyrrolo[2,3-*d*]pyrimidines (7-deazapurines) were synthesized in high yields by the reaction of 2-amino-1-methyl-4,5-diphenyl-1*H*-pyrrole-3-carbonitrile with triethyl orthoformate followed by cyclocondensation with methyl or benzyl amine in refluxing glacial acetic acid or using ultrasonic irradiation containing a catalytic amount of glacial acetic acid at 60°C. For each product, the correct structural isomer was identified using the FT-IR, ^{1}H NMR, ^{13}C NMR, 2D nuclear Overhauser effect spectroscopy spectral and microanalytical data together with comparison of the experimental and calculated chemical shifts at the B3LYP/6-31+G(d,p) level of theory. Furthermore, the synthesized compounds were evaluated for their antibacterial activity against Gram-positive bacteria (*Staphylococcus aureus* and *Micrococcus luteus*) and Gram-negative bacteria (*Escherichia coli*) by the agar dilution method using 24-well microtiter plates.

Keywords: 7-deazapurines; antibacterial; DFT; pyrrolo[2,3-*d*]pyrimidines; ultrasonic irradiation.

1 Introduction

One of the important heterocyclic systems with high structural resemblance to purine (**I**) is pyrrolo[2,3-*d*]pyrimidine (**II**) in which the N-7 of purine (in purine numbering) has been replaced by a CH group and is named as 7-deazapurine (in this paper, the numbering of type **II** is used) (Fig. 1). These compounds have attracted organic chemists very much because of diverse and interesting biological activities such as antitumor [1], antifungal [2], antibacterial [3], antiangiogenic [4], antiviral [5], antiinflammatory [6], and anti-HCV [7] activities. Several pyrrolo[2,3-*d*]pyrimidines exhibit high inhibitory activities toward human tumor cell lines and enzymes such as KB [8], SW620 [8], A549 [8], cryptosporidium hominis thymidylate synthase [9], tumor necrosis factor-α [10], Janus kinase 2 [11], Bruton's tyrosine kinase [12], glycinamide ribonucleotide formyltransferase [13], 5-aminoimidazole-4-carboxamide ribonucleotide formyltransferase [8, 13], and protein kinase B [14]. Also, a number of these compounds have been shown to induce neurogenesis in murine embryonic stem cells [15]. Furthermore, various derivatives of these compounds have been substantially investigated as a part of the synthesis of new C-nucleosides with potential biomedical interest, since they have been found to exhibit pronounced growth inhibitory activity to several leukemic cell lines [16–18].

On the other hand, ultrasonic irradiation is used as a convenient method for accelerating organic reactions [19, 20]. Compared with traditional methods, the attractive features of this technique include short reaction times, high selectivity, high yields, reduced energy consumption, and clean reaction with an easy work-up [21–25].

Inspired by these facts and due to our interest in the synthesis of heterocyclic compounds with potential biological activities [26–35], and in continuation of our previous works in the synthesis of new pyrrolo[2,3-*d*]pyrimidines [36–39], herein we report a convenient ultrasonic-assisted synthesis of two new pyrrolo[2,3-*d*]pyrimidines in high yields (Scheme 1). These new compounds were also evaluated for their antibacterial activity against two strains of Gram-positive bacteria, *Staphylococcus aureus* (*S. aureus*, PTCC 1112) and *Micrococcus luteus* (*M. luteus*,

***Corresponding author: Abolghasem Davoodnia,** Department of Chemistry, Mashhad Branch, Islamic Azad University, Mashhad, I.R. Iran, e-mail: adavoodnia@yahoo.com, adavoodnia@mshdiau.ac.ir
Simin Vazirimehr and S. Ali Beyramabadi: Department of Chemistry, Mashhad Branch, Islamic Azad University, Mashhad, I.R. Iran
Mahboobeh Nakhaei-Moghaddam: Department of Biology, Mashhad Branch, Islamic Azad University, Mashhad, I.R. Iran
Niloofar Tavakoli-Hoseini: Department of Chemistry, Mashhad Branch, Islamic Azad University, Mashhad, I.R. Iran; and Department of Biochemistry and Clinical Laboratories, Tabriz University of Medical Sciences, Tabriz, I.R. Iran

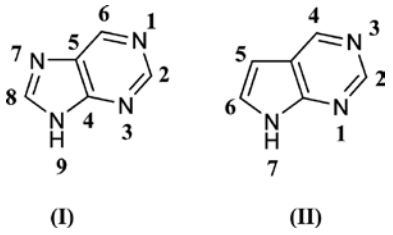


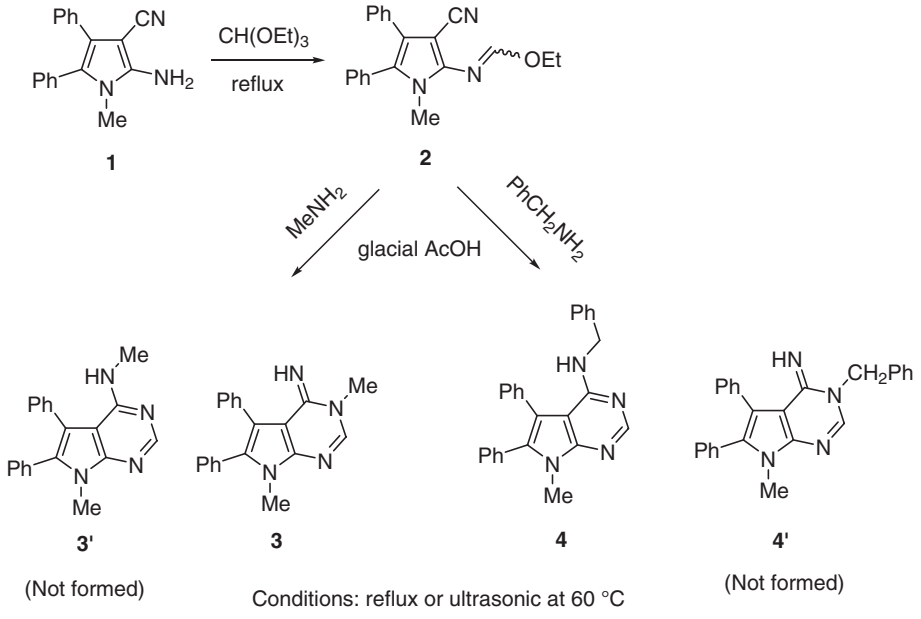
Fig. 1: Purine core (**I**) and its 7-deaza analogue (**II**).

PTCC 1110), and one strain of Gram-negative bacteria, *Escherichia coli* (*E. coli*, PTCC 1330), by the agar dilution method using 24-well microtiter plates (PTCC = Persian Type Culture Collection).

2 Results and discussion

Initially, the condensation reaction of 2-amino-1-methyl-4,5-diphenyl-1*H*-pyrrole-3-carbonitrile (**1**) [40] with excess triethyl orthoformate under reflux gave the condensed compound **2** in high yield [33]. This compound was then allowed to interact with methyl or benzyl amine in the presence of glacial acetic acid under ultrasonic irradiation at 60°C. Monitoring of the reactions with thin-layer chromatography (TLC) showed the formation of a product in each case which was isolated from the reaction mixture, as described in the Experimental section. The same products were also obtained under conventional heating in refluxing glacial acetic acid. The results, however, show that the ultrasonic irradiation approach is faster and the yields are higher than by the conventional heating method.

The structural elucidation of the isolated products was based upon spectral and microanalytical data. The ^{1}H NMR spectrum of the compound isolated from the reaction of methyl amine and compound **2** in $CDCl_3$ showed two singlets at $\delta = 3.52$ and 3.64 ppm for two methyl groups, a sharp singlet at $\delta = 7.70$ ppm belonging to the CH in the pyrimidine ring and the characteristic signals at $\delta = 7.18$–7.31 ppm for the aromatic protons and the NH group. The FT-IR spectrum was devoid of the CN absorption band of the precursor **2**, which shows the inclusion of the nitrile moiety in the cyclocondensation process. Furthermore, the ^{13}C NMR spectrum showed the characteristic signals at $\delta = 29.9$, 35.6, 103.2, 117.4, 126.8, 127.9, 128.3 (two carbons at 128.30 and 128.34), 130.5, 130.7, 130.8, 132.8, 134.1, 143.2, 145.6, and 156.2 ppm for two methyl groups as well as the aromatic carbons. Also this compound gave satisfactory elemental analysis data corresponding to the molecular formula $C_{20}H_{18}N_4$. In accord with these data, two structural isomers **3** and **3′** are possible for the isolated product (Scheme 1). However, based on the above-mentioned spectral and microanalytical data, the structure of the product cannot be assigned. For finding the correct structure of the isolated product, the 2D nuclear Overhauser effect spectroscopy (NOESY) spectrum was also prepared. As shown in the expanded view of the 2D NOESY spectrum in Fig. 2, the interaction between pyrimidine CH at $\delta = 7.70$ ppm and the methyl group at $\delta = 3.52$ ppm indicates that these groups are close together, and therefore the correct structure is **3**.



Scheme 1: Synthesis of new pyrrolo[2,3-*d*]pyrimidines.

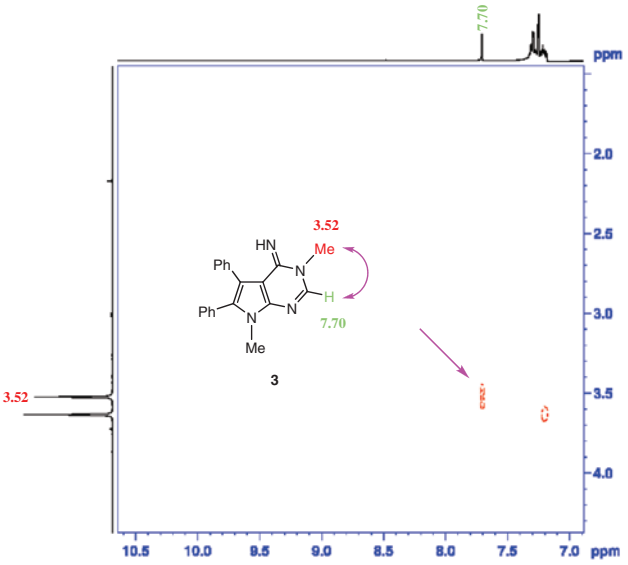


Fig. 2: The expanded view of the 2D NOESY spectrum of compound **3**.

Surprisingly, as shown in Fig. 3, the ^{1}H NMR spectrum of the isolated product from the reaction of benzyl amine and compound **2** in $CDCl_3$ showed a doublet for the CH_2 group at $\delta = 4.70$ ppm with a coupling constant (J value) of 5.7 Hz. Such splitting is in accord with structure **4**, and not **4′**, which in the CH_2 group can be split via vicinal coupling with the NH group in an A_2X splitting pattern. Such splitting is expected when the proton exchange in the NH group is slow. On the other hand, the pyrimidine CH signal is also seen as a doublet at $\delta = 8.54$ ppm with a coupling constant of 3.6 Hz. We believe that this is due to a long-range coupling across five bonds with the NH group. For the NH group, however, because of the nitrogen electric quadrupole moment effect, a relatively broad signal at $\delta = 5.38$ ppm is observed which was removed on deuteration. Other signals contain a singlet at $\delta = 3.37$ ppm for the methyl group and the characteristic signals at $\delta = 7.17$–7.42 ppm for the aromatic protons. In the 2D NOESY spectrum, there is no interaction between pyrimidine CH at $\delta = 8.54$ ppm and the methylene group at $\delta = 4.70$ ppm, which indicates that these groups are not close together and confirms structure **4**. Furthermore, the FT-IR, ^{13}C NMR, and elemental analysis data confirm the formation of compound **4** (Experimental section and Supplementary Information).

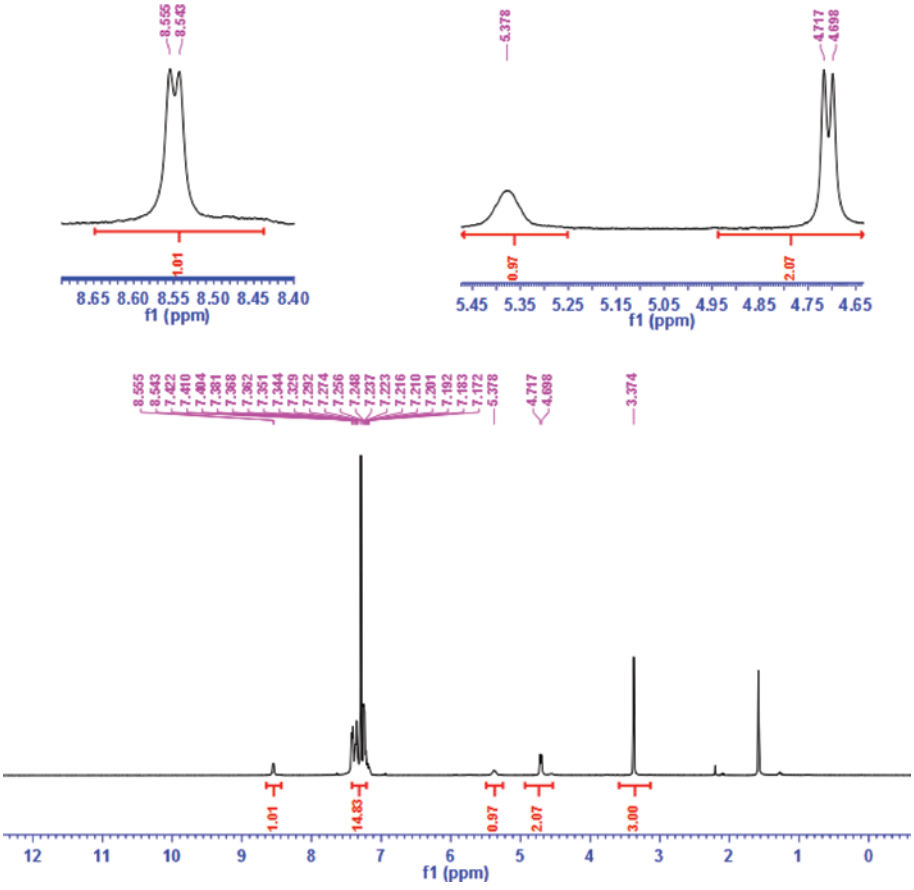


Fig. 3: The ^{1}H NMR spectrum of compound **4** in $CDCl_3$ and its expanded view.

Table 1: The comparison of selected calculated (Cal.) ^{1}H and ^{13}C NMR chemical shifts data (δ, ppm) with those obtained from the experimental (Exp.) spectroscopy.

Compound	Position of H	^{1}H NMR		Deviation $\lvert\delta_{exp}-\delta_{calcd}\rvert$	Position of C	^{13}C NMR		Deviation $\lvert\delta_{exp}-\delta_{calcd}\rvert$
		Calcd.	Exp.			Calcd.	Exp.	
3	N=CH	7.66	7.70	0.04	N–CH_3 (on pyrrole)	30.6	29.9	0.7
3′	N=CH	8.53	–	0.83	N–CH_3 (on pyrrole)	31.0	–	1.1
3	NH	6.72	7.18–7.31	0.46–0.59	N–CH_3 (on pyrimidine)	37.7	35.6	2.1
3′	NH	4.91	–	2.27–2.40	N–CH_3 (on pyrimidine)	28.9	–	6.7
4	N=CH	8.68	8.54	0.14	N–CH_3	30.3	29.9	0.4
4′	N=CH	7.80	–	0.74	N–CH_3	31.2	–	1.3
4	NH	5.05	5.38	0.33	CH_2	45.9	44.6	1.3
4′	NH	6.79	–	1.41	CH_2	50.5	–	5.9

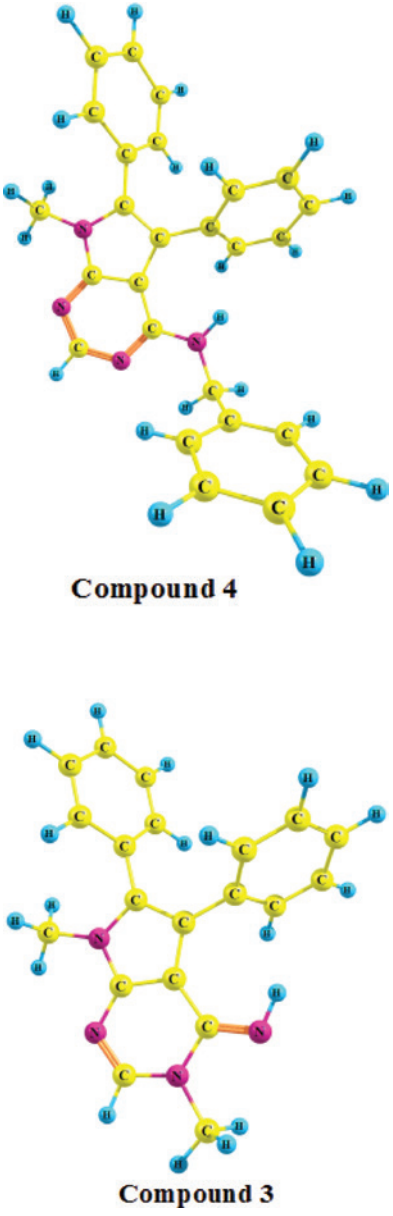


Fig. 4: The optimized geometries of compounds **3** and **4**.

The experimental ^{1}H NMR and ^{13}C NMR chemical shifts were also compared with the density functional theory (DFT)-calculated ones. The obtained results for selected atoms are given in Table 1. As can be seen, the observed chemical shifts are closer to the calculated values for compounds **3** and **4** than **3′** and **4′**. Based on the good consistency and less deviation between the experimental and DFT chemical shifts of compounds **3** and **4**, it seems likely that the isolated isomers are structurally similar to compounds **3** and **4** not to **3′** and **4′**. Optimized geometries of compounds **3** and **4** are shown in Fig. 4.

The synthesized compounds **3** and **4** were screened for the antibacterial activity against reference strains of *S. aureus*, *M. luteus*, and *E. coli* bacteria. The growth of tested bacteria was inhibited at the concentration of 6 mg mL^{-1} of compounds **3** and **4**. The minimum inhibitory concentration (MIC) of these compounds against *S. aureus* and *E. coli* was 5 and 6 mg mL^{-1}, respectively. The MIC values of compounds **3** and **4** against *M. luteus* were 3 and 4 mg mL^{-1}, respectively.

3 Conclusions

We have reported the synthesis of two new pyrrolo[2,3-*d*]-pyrimidines **3** and **4** by the reaction of 2-amino-1-methyl-4,5-diphenyl-1*H*-pyrrole-3-carbonitrile with triethyl orthoformate followed by cyclocondensation with methyl or benzyl amine in refluxing glacial acetic acid or using ultrasonic irradiation containing a catalytic amount of glacial acetic acid at 60°C. The results showed that the classical approach is a tedious method affording relatively lower yields with much longer reaction times. From the spectral, especially 2D NOESY and ^{1}H NMR, and also microanalytical data, it is confirmed that while the reaction with methyl amine gave the 3-methyl-4-imine derivative **3**, the 4-benzylamino derivative **4** was obtained when

benzyl amine was used as the respective amine. The theoretical ^{1}H NMR and ^{13}C NMR chemical shifts of compounds **3** and **4** are in good agreement with the experimental ones, confirming the structure of these compounds as isolated products of the reaction. The new synthesized compounds **3** and **4** have growth-inhibiting effects on *S. aureus*, *M. luteus*, and *E. coli* bacteria. While the MIC of two new compounds **3** and **4** against *S. aureus* and *E. coli* were 5 and 6 mg mL^{-1}, respectively, the MIC of the compounds against *M. luteus* were 3 and 4 mg mL^{-1}, respectively.

4 Experimental section

4.1 Chemicals and apparatus

All chemicals were purchased from Merck and Aldrich and used without additional purification. Ultrasonication was performed by Soltec sonicator (Italy, 2200ETH S3) at a frequency of 40 kHz and a nominal power of 260 W. IR spectra were obtained with KBr pellets using a Tensor 27 Bruker spectrophotometer. The ^{1}H and ^{13}C NMR spectra were recorded with a Bruker 300 FT spectrometer at 300 and 75 MHz frequencies for ^{1}H and ^{13}C, respectively, using tetramethylsilane (TMS) as an internal standard. Elemental analysis was performed on a Thermo Finnigan Flash EA microanalyzer. Melting points were recorded on a Stuart SMP3 melting point apparatus.

4.2 General experimental procedure for the synthesis of compounds 3 and 4

4.2.1 Method A (using ultrasonic irradiation)

A mixture of ethyl *N*-3-cyano-1-methyl-4,5-diphenyl-1*H*-pyrrol-2-ylformimidate (**2**) (1 mmol) and methyl or benzyl amine (1.1 mmol) in the presence of a few drops of glacial acetic acid was sonicated at 60°C for 50 min. The reaction was monitored by TLC. After the completion of the reaction, the mixture was cooled to room temperature, cold ethanol was added, and the precipitate was filtered off. The crude product was recrystallized from ethanol to give the pure compounds **3** and **4** in 90 and 85 yields, respectively.

4.2.2 Method B (using conventional heating)

A mixture of ethyl *N*-3-cyano-1-methyl-4,5-diphenyl-1*H*-pyrrol-2-ylformimidate (**2**) (1 mmol) and methyl or benzyl amine (1.1 mmol) in glacial acetic acid (5 mL) was heated under reflux for 4 h. The reaction was monitored by TLC. After the completion of the reaction, the solvent was evaporated *in vacuo* and the residue crude product was recrystallized from ethanol to give the pure compounds **3** and **4** in 85 and 81 yields, respectively.

4.3 Spectral and microanalytical data

4.3.1 3,7-Dimethyl-5,6-diphenyl-3*H*-pyrrolo[2,3-*d*]-pyrimidin-4(7*H*)-imine (3)

M.p. 231–233°C. – IR (KBr disk): υ = 3318, 3026, 2949, 1632, 1565, 1502, 1325, 1181, 1077, 791, 758, 703 cm^{-1}. – ^{1}H NMR (300 MHz, $CDCl_3$, 25°C, TMS): δ = 3.52 (s, 3H, CH_3), 3.64 (s, 3H, CH_3), 7.18–7.31 (m, 11H, arom-H and NH), 7.70 (s, 1H, pyrimidine CH). – ^{13}C NMR (75 MHz, $CDCl_3$, 25°C, TMS): δ = 29.92, 35.56, 103.19, 117.43, 126.85, 127.86, 128.30, 128.34, 130.52, 130.66, 130.84, 132.79, 134.06, 143.22, 145.59, 156.25. – $C_{20}H_{18}N_4$ (314.4): calcd. C 76.41, H 5.77, N 17.82; found C 76.12, H 5.69, N 17.91. (See also Supplementary Information.)

4.3.2 4-Benzylamino-7-methyl-5,6-diphenyl-7*H*-pyrrolo[2,3-*d*]pyrimidine (4)

M.p. 173–175°C. IR (KBr disk): υ = 3429, 3025, 2910, 1660, 1584, 1454, 1412, 1334, 1265, 1180, 1134, 1071, 825, 795, 766 cm^{-1}. – ^{1}H NMR (300 MHz, $CDCl_3$, 25°C, TMS): δ = 3.37 (s, 3H, CH_3), 4.70 (d, 2H, J = 5.7 Hz, CH_2), 5.38 (br, 1H, NH), 7.17–7.42 (m, 15H, arom-H), 8.54 (d, 1H, J = 3.6 Hz, pyrimidine CH). – ^{13}C NMR (75 MHz, $CDCl_3$, 25°C, TMS): δ = 29.88, 44.61, 101.80, 113.30, 127.03, 127.15, 127.17, 128.14, 128.37, 128.53, 128.58, 130.57, 130.87, 134.47, 134.61, 138.82, 150.15, 151.97, 156.27. – $C_{26}H_{22}N_4$ (390.5): calcd. C 79.97, H 5.68, N 14.35; found C 80.27, H 5.57, N 14.21. (See also Supplementary Information.)

4.4 Computational details

In this work, all of the calculations have been performed using DFT methods as implemented in the Gaussian 03 program package [41]. The B3LYP functional [42] and the 6-31+G(d,p) basis sets were used. First, the geometry of the compounds was fully optimized, which was confirmed to have no imaginary frequency of the Hessian. Then, the optimized geometries were employed to compute the chemical shifts. The ^{1}H and ^{13}C NMR chemical shifts were

predicted with respect to TMS, where the gauge-independent atomic orbital method was used [43].

4.5 Biological

Bacterial strains including *S. aureus* (PTCC 1112) and *M. luteus* (PTCC 1110) as Gram-positive and *E. coli* (PTCC 1330) as Gram-negative bacteria were obtained from the Iranian Research Organization for Science and Technology in Iran. Antimicrobial assay was examined by the agar dilution method in 24-well microtiter plates. Every well was filled with 500 µL of molten Mueller Hinton agar (QUELAB, Canada) at a double concentration, 150 µL of every synthesized compound stock solution (0.04 g of compound in 1 mL distilled water), and 350 µL distilled water, so that the concentration in the well was 6 mg mL^{-1}. After the mixing and solidification of the media, 0.01 mL of every bacterial suspension, equivalent to McFarland tube No. 0.5 (10^8 CFU mL^{-1}) was inoculated on the agar of every well. The culture plates were then incubated at 37°C for 24 h. All tests were repeated three times with controls. For compounds **3** and **4** with antibacterial activity in 6 mg mL^{-1}, tests were conducted at lower concentrations (1–5 mg mL^{-1}) to determine the MIC.

5 Supplementary information

FT-IR, ^{1}H NMR, ^{13}C NMR, and 2D NOESY spectra are given as Supplementary Information available online (DOI: 10.1515/znb-2017-0004).

Acknowledgments: We gratefully acknowledge financial support from the Islamic Azad University, Mashhad Branch, Iran.

References

[1] A. Gangjee, O. A. Namjoshi, S. N. Keller, C. D. Smith, *Bioorg. Med. Chem.* **2011**, *19*, 4355.
[2] M. S. A. El-Gaby, A. M. Gaber, A. A. Atalla, K. A. Abd Al-Wahab, *Farmaco* **2002**, *57*, 613.
[3] K. M. Hassan Hilmy, M. M. A. Khalifa, M. A. Allah Hawata, R. M. Aboalzeen Keshk, A. A. El-Torgman, *Eur. J. Med. Chem.* **2010**, *45*, 5243.
[4] A. Gangjee, Y. Zhao, S. Raghavan, M. A. Ihnat, B. C. Disch, *Bioorg. Med. Chem.* **2010**, *18*, 5261.
[5] P. Januszczyk, J. Fogt, J. Boryski, K. Izawa, T. Onishi, J. Neyts, E. De Clercq, *Nucleosides Nucleotides Nucleic Acids* **2009**, *28*, 713.
[6] M. S. Mohamed, R. Kamel, R. H. Abd El-Hameed, *Med. Chem. Res.* **2013**, *22*, 2244.
[7] A. Thiyagarajan, M. Toyama, M. Baba, A. Sharon, C. Bal, *Nucleosides Nucleotides Nucleic Acids* **2016**, *35*, 305.
[8] Y. Liu, M. Li, H. Zhang, J. Yuan, C. Zhang, K. Zhang, H. Guo, L. Zhao, Y. Du, L. Wang, L. Ren, *Eur. J. Med. Chem.* **2016**, *115*, 245.
[9] V. P. Kumar, K. M. Frey, Y. Wang, H. K. Jain, A. Gangjee, K. S. Anderson, *Bioorg. Med. Chem. Lett.* **2013**, *23*, 5426.
[10] K. M. H. Hilmy, H. G. Abdul-Wahab, D. H. Soliman, M. M. A. Khalifa, A. M. Hegab, *Med. Chem. Res.* **2015**, *24*, 2097.
[11] T. Wang, X. Liu, M. Hao, J. Qiao, C. Ju, L. Xue, C. Zhang, *Bioorg. Med. Chem. Lett.* **2016**, *26*, 2936.
[12] X. Zhao, W. Huang, Y. Wang, M. Xin, Q. Jin, J. Cai, F. Tang, Y. Zhao, H. Xiang, *Bioorg. Med. Chem.* **2015**, *23*, 891.
[13] Y. Wang, S. Mitchell-Ryan, S. Raghavan, C. George, S. Orr, Z. Hou, L. H. Matherly, A. Gangjee, *J. Med. Chem.* **2015**, *58*, 1479.
[14] T. McHardy, J. J. Caldwell, K.-M. Cheung, L. J. Hunter, K. Taylor, M. Rowlands, R. Ruddle, A. Henley, A. H. De Brandon, M. Valenti, T. G. Davies, L. Fazal, L. Seavers, F. I. Raynaud, S. A. Eccles, G. Wynne Aherne, M. D. Garrett, I. Collins, *J. Med. Chem.* **2010**, *53*, 2239.
[15] S. Ding, T. Y. H. Wu, A. Brinker, E. C. Peters, W. Hur, N. S. Gray, P. G. Schultz, *Proc. Natl. Acad. Sci.* **2003**, *100*, 7632.
[16] M. I. Lim, R. S. Klein, J. J. Fox, *Tetrahedron Lett.* **1980**, *21*, 1013.
[17] M. I. Lim, R. S. Klein, *Tetrahedron Lett.* **1981**, *22*, 25.
[18] M. I. Lim, W. Y. Ren, B. A. Otter, R. S. Klein, *J. Org. Chem.* **1983**, *48*, 780.
[19] E. Ruiz, H. Rodríguez, J. Coro, E. Salfrán, M. Suárez, R. Martínez-Alvarez, N. Martín, *Ultrason. Sonochem.* **2011**, *18*, 32.
[20] S. M. Zhang, H. Li, X. C. Zheng, B. Q. Li, S. H. Wu, W. P. Huang, Z. G. Liu, Y. Feng, *Chin. J. Org. Chem.* **2002**, *22*, 603.
[21] R. Cella, H. Stefani, *Tetrahedron* **2009**, *65*, 2619.
[22] G. Cravotto, P. Cintas, *Chem. Soc. Rev.* **2006**, *35*, 180.
[23] T. J. Mason, D. Peters, *Practical Sonochemistry, Power Ultrasound Uses and Applications*, 2nd edition, Horwood Chemical Science Series, London, **2002**.
[24] J. T. Li, Y. J. Bian, H. J. Zang, T. S. Li, *Synth. Commun.* **2002**, *32*, 547.
[25] H. Zang, M. Wang, B. W. Cheng, J. Song, *Ultrason. Sonochem.* **2009**, *16*, 301.
[26] A. Davoodnia, M. Bakavoli, N. Pooryaghoobi, M. Roshani, *Heterocycl. Commun.* **2007**, *13*, 323.
[27] A. Davoodnia, M. Bakavoli, S. Mohseni, N. Tavakoli-Hoseini, *Monatsh. Chem.* **2008**, *139*, 963.
[28] A. Davoodnia, R. Zhiani, N. Tavakoli-Hoseini, *Monatsh. Chem.* **2008**, *139*, 1405.
[29] A. Davoodnia, M. Rahimizadeh, H. Atapour-Mashhad, N. Tavakoli-Hoseini, *Heteroat. Chem.* **2009**, *20*, 346.
[30] A. Davoodnia, M. Bakavoli, M. Soleimany, N. Tavakoli-Hoseini, *Monatsh. Chem.* **2009**, *140*, 355.
[31] A. Davoodnia, M. Bakavoli, R. Moloudi, N. Tavakoli-Hoseini, M. Khashi, *Monatsh. Chem.* **2010**, *141*, 867.
[32] S. Gholipour, A. Davoodnia, M. Nakhaei-Moghaddam, *Chem. Heterocycl. Compd.* **2015**, *51*, 808.
[33] M. Khashi, A. Davoodnia, V. S. Prasada Rao Lingam, *Res. Chem. Intermed.* **2015**, *41*, 5731.
[34] S. Vazirimehr, A. Davoodnia, M. Nakhaei-Moghaddam, *Heterocycl. Lett.* **2016**, *6*, 167.
[35] A. Davoodnia, A. Nakhaei, N. Tavakoli-Hoseini, *Z. Naturforsch.* **2016**, *71b*, 219.

[36] A. Davoodnia, M. Bakavoli, R. Moloudi, M. Khashi, N. Tavakoli-Hoseini, *Chin. Chem. Lett.* **2010**, *21*, 1.
[37] A. Davoodnia, M. Bakavoli, M. Khashi, R. Moloudi, N. Tavakoli-Hoseini, *Asian J. Chem.* **2010**, *22*, 4625.
[38] A. Davoodnia, M. Khashi, N. Tavakoli-Hoseini, R. Moloudi, H. A. Zamani, *Monatsh. Chem.* **2013**, *144*, 677.
[39] M. Khashi, A. Davoodnia, J. Chamani, *Phosphorus Sulfur Silicon Relat. Elem.* **2014**, *189*, 839.
[40] H. J. Roth, K. Eger, *Arch. Pharm.* **1975**, *308*, 179.
[41] M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, J. A. Montgomery, Jr., T. Vreven, K. N. Kudin, J. C. Burant, J. M. Millam, S. S. Iyengar, J. Tomasi, V. Barone, B. Mennucci, M. Cossi, G. Scalmani, N. Rega, G. A. Petersson, H. Nakatsuji, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, M. Klene, X. Li, J. E. Knox, H. P. Hratchian, J. B. Cross, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, P. Y. Ayala, K. Morokuma, G. A. Voth, P. Salvador, J. J. Dannenberg, V. G. Zakrzewski, S. Dapprich, A. D. Daniels, M. C. Strain, O. Farkas, D. K. Malick, A. D. Rabuck, K. Raghavachari, J. B. Foresman, J. V. Ortiz, Q. Cui, A. G. Baboul, S. Clifford, J. Cioslowski, B. B. Stefanov, G. Liu, A. Liashenko, P. Piskorz, I. Komaromi, R. L. Martin, D. J. Fox, T. Keith, M. A. Al-Laham, C. Y. Peng, A. Nanayakkara, M. Challacombe, P. M. W. Gill, B. Johnson, W. Chen, M. W. Wong, C. Gonzalez, J. A. Pople, Gaussian 03, revision B. 05, Gaussian, Inc., Pittsburgh, PA, USA, **2003**.
[42] C. Lee, W. Yang, R. G. Parr, *Phys. Rev. B* **1988**, *37*, 785.
[43] R. Ditchfield, *Mol. Phys.* **1974**, *27*, 789.

Supplemental Material: The online version of this article (DOI: 10.1515/znb-2017-0004) offers supplementary material, available to authorized users.