

Org. Chem. Res., Vol. 6, No. 2, 173-178, September 2020.

DOI: 10.22036/org.chem.2020.214359.1227

Synthesis of Functionalized Dihydropyrimidothiazinoquinoline Derivatives *via* Reaction of 3-(Chloromethyl)-2-chloroquinolines and 2-Thioxo-2,3-dihydropyrimidin-4(1*H*)-one

M. Shiri*, S. Fazelzadeh and V. Zadsirjan

Chemistry Department, Faculty of Physics and Chemistry, Alzahra University, Vanak, 1993893973 Tehran, Iran (Received 3 January 2020, Accepted 26 April 2020)

An efficient method for the synthesis of novel dihydropyrimidothiazinoquinoline derivatives through the reaction of 2-chloro-3-(chloromethyl)quinolines and 2-thioxo-2,3-dihydropyrimidin-4(1H)-one in the presence of K_2CO_3 is demonstrated.

Keywords: Quinoline, Cyclization, 3-(Chloromethyl)-2-chloroquinolines, Cascade reaction

INTRODUCTION

Quinoline is one of the most widespread *N*-heteroaromatic cores incorporated into the structures of various pharmaceuticals. Quinolines signify an important group of heterocyclic compounds, as they are critical moiety of various biologically potent naturally occurring compounds as well as diverse pharmacologically fascinating compounds [1-3]. The quinoline ring has also gained a remarkable interest because of its significant applications in many arenes, for instance, they can be used as corrosion inhibitors [4], precursors of oil-soluble food colorants, and as chemo sensors in luminescence chemistry [5].

Moreover, quinolines contain various pharmacological and biological properties including antifungal [6], antileishmanial [7], antitumor [8], antibacterial [9], and antiamebic [10] and also chloroquinolines are extensively applied for the treatment of malaria [11].

Because of the importance and various usages of quinolines, different classical methods, such as the Skraup [12], Doebner [13], Doebner-von Miller [14], Pfitzinger [15], Combes [16] and Friedländer protocols [17] were reported to from the quinoline unit.

2-Chloroquinoline-3-carbaldehydes [18] as significant

*Corresponding author. E-mail: mshiri@alzhara.ac.ir

synthons were used for the formation of a number of heterocyclic compounds such as pyrazolo[3,4-b]quinolines [19], pyrano-[4,3-b]quinolines [20], quinolino[3,2-f][1,2,4] triazolo[4,3-b]-[1,2,4]triazepines [21], isoxazolo[5,4-b]quinolines [22], and benzo[g]naphtho[b][1,8]naphthyridines [23]. Nawaz Khan et al. in (2010) reported the synthesis of 3-[(2-chloroquinolin-3-yl)methyl]pyrimidin-4-(3H)-ones using potassium hydroxide and Fe nano particles [24]. Various quinoline based 1,3,4-oxadiazoles were synthesized via a chloro-amine coupling reaction in the presence of iodobenzenediacetate. These compounds were examined for antifungal, antibacterial, anti-malarial, antituberculosis properties. Some of them demonstrated good antituberculosis property [25].

In continuation of our interest on quinolines chemistry [26-35], herein, we report the synthesis of novel dihydropyrimido-thiazinoquinolinone derivatives using substituted 2-chloro-3-chloromethyl quinoline as a starting material.

EXPERIMENTAL

Chemicals and Apparatus

Chemicals were purchased from Fluka, Merck and Aldrich chemical companies. Melting points are uncorrected. The FT-IR spectra were recorded on a FT-IR

Tensor 27 Spectrophotometer. Nuclear magnetic resonance (NMR) spectra were recorded on a Bruker Avance 400 and 500 MHz Spectrometer in DMSO-d₆ as the solvent. All products were characterized using IR, ¹H NMR and ¹³C NMR spectroscopies.

General procedure for the synthesis of dihydro pyrimido[2',1':2,3][1,3]thiazino[4,5-b]quinolin-1(6H)-one (5a-g). 3-(Chloromethyl)-2-chloroquinolines 3a-g (0.5 mmol) and 2-thioxo-2,3-dihydropyrimidin-4(1H)-one 4 (0.5 mmol) in DMF in the presence of K₂CO₃ (1 mmol) was heated under reflux conditions for 4-9 h. The progress of the reaction was monitored by TLC (ethyl acetate: petroleum ether (9:1)). After completion, the reaction was quenched with ice-water. The mixture was filtered and the precipitate washed with MeOH to afford the product. The crude solid was purified with 95:5 ethyl acetate: n-hexane using column chromatography to obtain the pure white product.

Spectra Data

11-Methyl-4,4a-dihydropyrimido[2',1':2,3][1,3] thiazino[4,5-b]quinolin-1(6H)-one: (C₁₅H₁₁N₃OS) (5a). White powder (81%), m.p.: 289-291 °C; FT-IR (KBr): $v_{max} = 1480$, 1500, 1748 cm⁻¹; ¹H NMR (400 MHz, DMSO-d₆): $\delta = 2.74$ (3H, s, CH₃), 4.56 (2H, s, CH₂), 6.32 (1H, d, ${}^3J_{HH} = 8.0$ Hz, CH-16), 7.59 (1H, t, ${}^3J_{HH} = 7.6$ Hz, CH-1), 7.72 (1H, d, ${}^3J_{HH} = 7.2$ Hz, CH-6), 7.91 (1H, d, ${}^3J_{HH} = 7.6$ Hz, CH-2), 8.50 (1H, s, CH-10), 8.97 (1H, d, ${}^3J_{HH} = 8.0$ Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): $\delta = 18.0$, 27.4, 111.9, 120.1, 127.3, 127.7, 131.3, 136.4, 137.2, 139.8, 144.4, 146.3, 161.6, 167.0 ppm.

9-Methyl-4,4a-dihydropyrimido[2',1':2,3][1,3] thiazino[4,5-b]quinolin-1(6H)-one: (C₁₈H₁₁N₃OS) (5b). White powder (85%), m.p.: 317-319 °C; FT-IR (KBr): v_{max} = 1481, 1671 cm⁻¹; ¹H NMR (400 MHz, DMSO-d₆): δ = 2.12 (3H, s, CH₃), 4.56 (2H, s, CH₂), 6.30 (1H, d, ³J_{HH} = 8.0 Hz, CH-16), 7.73 (1H, d, ³J_{HH} = 8.4 Hz, CH-2), 7.87 (1H, s, CH-6), 7.96 (1H, d, ³J_{HH} = 8.8 Hz, CH-3), 8.44 (1H, s, CH-10), 8.86 (1H, d, ³J_{HH} = 7.6 Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): δ = 21.6, 27.5, 31.2, 111.7, 120.4, 127.0, 127.3, 128.3, 133.7, 136.2, 137.7, 139.8, 144.0 ppm.

Pyrimido[2',1':2,3][1,3]thiazino[4,5-b]quinolin-1(6

H)-one: (C₁₄H₉N₃OS) (5c). White powder (87%), m.p.: 295-297 °C; FT-IR (KBr): $v_{\text{max}} = 1482$, 1666, 126.6, 126.9 cm⁻¹. ¹H NMR (500 MHz, CDCl₃): $\delta = 4.30$ (2H, s, CH₂), 6.43 (1H, d, ³ $J_{\text{HH}} = 7.8$ Hz, CH-16), 7.66 (1H, t, ³ $J_{\text{HH}} = 8.1$ Hz, CH-1), 7.83 (1H, t, ³ $J_{\text{HH}} = 7.2$ Hz, CH-2), 7.90 (1H, d, ³ $J_{\text{HH}} = 8.1$ Hz, CH-6), 8.07 (1H, d, ³ $J_{\text{HH}} = 8.1$ Hz, CH-3), 8.17 (1H, s, CH-10), 8.92 (1H, d, ³ $J_{\text{HH}} = 7.8$ Hz, CH-17) ppm; ¹³C NMR (125 MHz, DMSO-d₆): $\delta = 29.7$, 126.6, 126.9, 127.4, 128.8, 130.8, 130.9, 132.0, 132.5, 133.7, 136.9, 142.7, 148.4 ppm.

9-Chloro-4,4a-dihydropyrimido[2',1':2,3][1,3] thiazino[4,5-b]quinolin-1(6H)-one: (C₁₄H₈ClN₃OS) (5d). White powder (77%), m.p.: 283-285 °C, FT-IR (KBr): $v_{max} = 1474$, 1646 cm⁻¹, ¹H NMR (400 MHz, DMSO-d₆): $\delta = 4.57$ (2H, s, CH₂), 6.30 (1H, d, ³ $J_{HH} = 8.0$ Hz, CH-16), 7.87, 7.89 (1H, dd, ³ $J_{HH} = 2.4$ Hz, ³ $J_{HH} = 8.0$ Hz, CH-2), 8.06 (1H, d, ³ $J_{HH} = 9.2$ Hz, CH-6), 8.25 (1H, d, ³ $J_{HH} = 2.0$ Hz, CH-3), 8.51 (1H, s, CH), 8.85 (1H, d, ³ $J_{HH} = 8.0$ Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): $\delta = 27.4$, 111.8, 121.6, 127.0, 128.0, 130.6, 132.0, 132.2, 136.2, 139.7, 134.9, 147.7, 161.5, 166.9 ppm.

9-Bromopyrimido[2',1':2,3][1,3]thiazino[4,5-b] quinolin-1(6*H***)-one: (C₁₄H₈BrN₃OS) (5e). White powder (80%), m.p.: 279-281 °C; FT-IR (KBr): v_{max} = 1650, 1795, 2922 cm⁻¹; ¹H NMR (400 MHz, DMSO-d₆): δ = 4.57 (2H, s, CH₂), 6.3 (1H, d, ³J_{HH} = 8.0 Hz, CH-16), 7.98 (2H, d, ³J_{HH} = 1.6 Hz, CH-2,6), 8.4 (1H, s, CH-3), 8.5 (1H, s, CH-10), 8.85 (1H, d, ³J_{HH} = 8.0 Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): δ = 27.4, 111.8, 120.8, 121.6, 128.5, 130.3, 130.7, 134.5, 136.0, 139.7, 144.1, 147.8, 161.5, 166.9 ppm.**

9-Methoxypyrimido[2',1':2,3][1,3]thiazino[4,5-b] quinolin-1(6*H***)-one: (**C₁₅H₁₁N₃O₂S**) (5f).** White powder (89%), m.p.: 244-246 °C; FT-IR (KBr): v_{max} = 1415, 1597, 2850, 2919 cm⁻¹; ¹H NMR (400 MHz, DMSO-d₆): δ = 4.07 (3H, s, CH₃), 4.63 (2H, s, CH₂), 6.3 (1H, d, ³ J_{HH} = 8.0 Hz, CH-16), 7.90 (1H, d, ³ J_{HH} = 4.8 Hz, CH-2), 8.07 (1H, d, J = 9.2 Hz, CH-3), 8.6 (1H, s, CH-6), 8.83 (1H, d, J = 8.0 Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): δ = 27.5, 57.5, 111.8, 114.6, 119.0, 122.1, 126.0, 129.0, 132.2, 139.7, 140.8, 146.1, 153.7, 161.4, 166.9 ppm.

Benzo[h]pyrimido[2',1':2,3][1,3]thiazino[4,5-b] quinolin-1(6H)-one: (C₁₈H₁₁N₃OS) (5g). White powder

Synthesis of Functionalized Dihydropyrimidothiazinoquinoline/Org. Chem. Res., Vol. 6, No. 2, 173-178, September 2020.

R = Me, OMe, H, Cl, Br, Benzo

Scheme 1. Synthesis of 3-(Chloromethyl)-2-chloroquinolines 3a-g

Scheme 1. The model reaction for the synthesized 5a

(85%), m.p.: 292-294 °C; FT-IR (KBr): v_{max} = 1609, 1740 cm⁻¹. ¹H NMR (400 MHz, DMSO-d₆): δ = 4.61 (2H, s, CH₂), 6.35 (1H, d, ³ J_{HH} = 7.6 Hz, CH-16), 7.82-7.85 (2H, m, CH-20,21), 7.97 (1H, d, ³ J_{HH} = 8.8 Hz, CH-6), 8.06 (1H, d, ³ J_{HH} = 8.8 Hz, CH-1), 8.10-8.13 (1H, m, CH-22), 8.59 (1H, s, CH-10), 9.16-9.19 (1H, m, CH-19), 9.22 (1H, d, ³ J_{HH} = 7.6 Hz, CH-17) ppm; ¹³C NMR (100 MHz, DMSO-d₆): δ = 27.2, 111.9, 120.3, 124.6, 125.2, 125.6, 128.0, 128.7, 128.8, 129.5, 130.2, 134.1, 137.1, 139.9, 143.6, 146.4, 161.5, 167.1 ppm.

RESULTS AND DISCUSSION

2-Chloroquinoline-3-carbaldehydes 1 was reduced to the corresponding alcohols 2 using NaBH₄ (Scheme 1). Then, alcohols 2 converted to the corresponding 2-chloro-3-(chloromethyl)quinolines 3 with SOCl₂ [36].

Next, 2-chloro-3-(chloromethyl)-8-methylquinoline 3a reacted with 2-thioxo-2,3-dihydropyrimidin-4(1H)-one 4 in the presence of K_2CO_3 under reflux in DMF for 5 hours. Surprisingly, fused polycyclic 11- methylpyrimido[2',1':2,3] [1,3]thiazino[4,5-b]quinolin-1(6H)-one 5a was obtained as the only product (Scheme 2).

To find the optimal reaction conditions, the effects of solvent, promoter and temperature were examined in a

model reaction involving 2-chloro-3-(chloromethyl)-8-methylquinoline 3a and 2-thioxo-2,3-dihydropyrimidin-4(1*H*)-one 4. The results are shown in Table 1. The best result was obtained when the reaction was performed in the presence of K₂CO₃ in DMF under the reflux condition, that product 5a was synthesized in 81% yield in 5 h (Table 1, entry 1). Other solvents such as EtOH, MeOH, H₂O, DMSO, CH₃CN, toluene and dioxane provided the desired products 5a in 35-75% yields (Table 1).

The ¹H NMR spectrum of 5a exhibited the singlet picks at $\delta = 2.74$ for CH₃ and at $\delta = 4.56$ for CH₂. Two peaks as a doublet in 6.32 and 8.97, respectively, are related to hydrogens of *alpha* and *beta* positions of carbonyl. The aromatic protons of quinoline ring appeared at 7.59 to 7.91 ppm as a doublet and a triplet and the singlet peak of quinoline ring at 8.50 ppm.

The FT-IR spectrum of 5a showed absorption bands related to C=N group at 1480 and carbonyl groups at 1500 cm⁻¹.

The ¹³C NMR spectrum showed 14 signals in agreement with the suggested structure. The signals related to newly formed bonds, C=N and C=O appeared in 161.6 and 167.0 ppm, respectively, and all data verified the molecular structure of 5a.

Relied on the optimal reaction conditions, the scope of

Table 1. Examination of Various Conditions in the Reaction of 2-Chloro-3-(chloromethyl)-8-methylquinoline 3a and 2-Thioxo-2,3-dihydropyrimidin-4(1*H*)-one 4a

Entry	Catalyst	Solvent	Temp.	Time (h)	Isolated yield (%)
1	K ₂ CO ₃	DMF	Reflux	5	81
2	K_2CO_3	МеОН	Reflux	10	64
3	K_2CO_3	H_2O	Reflux	20	10
4	K_2CO_3	EtOH	Reflux	10	30
5	K_2CO_3	DMSO	135	5	75
6	K_2CO_3	CH ₃ CN	Reflux	7	72
7	K_2CO_3	toluene	Reflux	6	75
8	K_2CO_3	Dioxane	Reflux	7	70
9	Cs_2CO_3	DMF	Reflux	6	61
10	L-proline	DMF	Reflux	12	35
11	None	DMF	Reflux	20	0
12	K_2CO_3	DMF	r.t.	20	0

Table 2. Diversity in the Synthesis of Dihydropyrimidothiazinoquinolinones 5a-g

R ²		CI + CI	HN NH	K ₂ CO ₃ DMF reflux	R ² N N N Sa-g		
Entry	R^1	R^2	Product	Time (h)	Yield (%)	m. p. (°C)	
1	Me	Н	5a	5	81	289-291	
2	Н	Me	5b	6	85	317-319	
3	Н	Н	5c	4	87	295-297	
4	Н	Cl	5d	8	77	283-285	
5	Н	Br	5e	9	80	279-281	
6	Н	OMe	5f	8	89	244-246	
7	-CH=CH- CH=CH-	Н	5g	7	85	292-294	

Scheme 3. A suggested probable mechanism for the synthesis of 5a

this reaction was examined using various 6-(un)substituted-2-chloro-3-(chloromethyl)quinolines 3a-d. As shown in Scheme 1, 3a-d containing electron-donating and halogen substituents and 2-thioxo-2,3-dihydropyrimidin-4(1*H*)-one 4 were applied and afforded the new dihydropyrimidothiazinoquinolinone derivatives 5a-g. The results are summarized in Table 2. Electron-donating and electron-withdrawing groups such as halogen on the aryl substituents worked well under aforementioned conditions. To the best of our knowledge, no analogous products have been reported in the literature so far.

A plausible mechanism for the synthesis of 5a is shown in Scheme 3. Reaction is initiated with hydrogen abstraction of N-H of 2-thioxo-2,3-dihydro-1*H*-pyrimidin-4-one with base, as shown in Scheme 3. After removal of hydrogen and then tautomerization with C=S, sulfur atom attacks to CH₂Cl of 2-chloro-3- chloromethyl-8-methyl-quinoline (3a) to generate intermediate I. Second hydrogen abstraction of N-H of I and then tatumerization prepared N as a nucleophile to intramolecular cyclization to form the desired product 5a (Scheme 3).

CONCLUSIONS

In summary, we have developed an efficient method for the synthesis of substituted, functionalized dihydropyrimidothiazinoquinolinone derivatives by the reaction of 3-(chloromethyl)-2-chloroquinolines and 2-thioxo-2,3-dihydropyrimidin-4(1H)-one in the presence of K_2CO_3 in DMF under the reflux condition. Some advantages of this method are good yields, mild reaction conditions, short reaction times and high selectivity. This method affords a simple and an effective route for the synthesis of novel dihydropyrimidothiazinoquinolines.

ACKNOWLEDGMENTS

We are thankful to Alzahra University and the Iran National Science Foundation (INSF) for the financial support.

REFERENCES

- [1] A. Shirai, O. Miyata, N. Tohnai, M. Miyata, D.J. Procter, D. Sucunza, T. Naito, J. Org. Chem. 73 (2008) 4464.
- [2] T. Shigeyama, K. Katakawa, N. Kogure, M. Kitajima, H. Takayama, Org. Lett. 9 (2007) 4069.
- [3] G.D. Henry, Tetrahedron 60 (2004) 6043.
- [4] E. Ebenso, I. Obor, Murulana, L. Int. J. Electrochem. Sci. 5 (2010) 1574.
- [5] E. Ballesteros, D. Moreno, T. Gomez, T. Rodriguez, J. Rojo, M. Garcia-Valverde, T. Torroba, Org. Lett. 11 (2009) 1269.
- [6] R. Musiol, J. Jampilek, V. Buchta, L. Silva, H. Niedbala, B. Podeszwa, A. Palka, K. Majerz-Maniecka, B. Oleksyn, J. Polanski, Bioorg. Med. Chem. 14 (2006) 3592.
- [7] M. Jain, S.I. Khan, B.L. Tekwani, M.R. Jacob, S. Singh, P.P. Singh, R. Jain, Bioorg. Med. Chem. 13 (2005) 4458.
- [8] a) Y.L. Zhao, Y.L. Chen, F.S. Chang, C.C.Tzeng, Eur. J. Med. Chem. 40 (2005) 792.
- [9] Y.L. Chen, C.J. Huang, Z.Y. Huang, C.H. Tseng, F.S. Chang, S.H. Yang, S.R. Lin, C.C. Tzeng, Bioorg. Med. Chem. 14 (2006) 3098.
- [10] a) M. G. Kayirere, A. Mahamoud, J. Chevalier, J.C. Soyfer, A. Cremieux, J. Barbe, Eur. J. Med. Chem. 33 (1998) 55; b) M. Kidwai, K.R. Bhushan, P. Sapra,

- R.K. Saxena, R. Gupta, Bioorg. Med. Chem. 8 (2000) 69
- [11] a) J.H. Burckhalter, W.H. Edgerton, J. Am. Chem. Soc. 73 (1951) 4837; b) D.M. Bailey, E.M. Mount, J. Siggins, J.A. Carlson, A. Yarinsky, R.G. Slighter, J. Med. Chem. 22 (1979) 599.
- [12] a) R. Klingenstein, P. Melnyk, S.R. Leliveld, A. Ryckebusch, C. Korth, J. Med. Chem. 49 (2006) 5300; b) Y.L. Chen, Y.L. Zhao, C.M. Lu, C.C. Tzeng, J.P. Wang, Bioorg. Med. Chem. 14 (2006) 4373; c) P. Benedetti, R. Mannhold, G. Cruciani, G. Ottaviani, Bioorg. Med. Chem. 12 (2004) 3607.
- [13] a) S.E. Denmark, S. Venkatraman, J. Org. Chem. 71 (2006) 1668; b) M.E. Theoclitou, L.A. Robinson, Tetrahedron Lett. 43 (2002) 3907.
- [14] O. Nitidandhaprabhas, Nature 212 (1966) 504.
- [15] F.W. Bergstrom, Chem. Rev. 35 (1944) 77.
- [16] E.J. Cragoe, C.M. Robb, Org. Synth. 5 (1973) 635.
- [17] J.L.J. Born, Org. Chem. 37 (1972) 3952.
- [18] B.R. McNaughton, B.L. Miller, Org. Lett. 5 (2003) 4257.
- [19] A. Srivastava, R.M. Singh, Indian J. Chem. 44B (2005) 1868.
- [20] S. Paul, M. Gupta, R. Gupta, A. Loupy, Tetrahedron Lett. 42 (2001) 3827.
- [21] A. Chandra, B. Singh, R.S. Khanna, R. Singh, J. Org. Chem. 74 (2009) 5664.
- [22] M. Gupta, Bioorg. Med. Chem. Lett. 21 (2011) 4919.
- [23] V. Nadaraj, S.T. Selvi, J. Chem. Pharm. Res. 4 (2012) 2850.
- [24] S.M. Roopan, F.R. Nawaz Khan, B.K. Mandal, Tetrahedron Lett. 51 (2010) 2309.

- [25] M. Gund, S.M. Roopan, F.R.N. Khan, J.S. Jin, R. Kumar, A.S. Kumar, Res. Chem. Intermed. 38 (2012) 1111.
- [26] Z. Tanbakouchian, M.A. Zolfigol, B. Notash, M. Ranjbar, M. Shiri, Appl. Organometal. Chem. 33 (2019) 5024.
- [27] M. Shiri, M.A. Zolfigol, H.G. Kruger, Z. Tanbakouchian, Advances in Heterocyclic Chemistry, ed. Katritzky A. R., Academic, Oxford, 185 (2011) 139.
- [28] Z. Faghihi, H.A. Oskooie, M.M. Heravi, M. Tajbakhsh, M. Shiri, Monatsh. Chem. 148 (2017) 315.
- [29] M. Shiri, M. Ranjbar, Z. Yasaei, F. Zamanian, B. Notash, Org. Biomol. Chem. 15 (2017) 10073.
- [30] M. Shiri, R. Pourabed, V. Zadsirjan, E. Sodagar, Tetrahedron Lett. 57 (2016) 5435.
- [31] M. Shiri, M.A. Zolfigol, M. Pirveysian, R. Ayazi-Nasrabadi, H.G. Kruger, T. Naicker, I. Mohammad Poor Baltork, Tetrahedron 68 (2012) 6059.
- [32] M. Shiri, M. Fathollahi-Lahroud, Z. Yasaei, Tetrahedron 73 (2017) 2501.
- [33] M. Shiri, M. Heydari, V. Zadsirjan, Tetrahedron 73 (2017) 2116.
- [34] M. Shiri, Z. Faghihi, H.A. Oskooei, M.M. Heravi, Sh. Fazelzadeh, B. Notash, RSC Adv. 6 (2016) 92235.
- [35] Z. Faghihi, M. Shiri, R. Pourabed, M.M. Heravi, V. Zadsirjan, Polycycl. Aromat. Compd. 28 (2019) 1.
- [36] S.S. Sonar, S.A. Sadaphal, R.U. Pokalwar, B.B. Shingate, M.S. Shingare, J. Heterocycl. Chem. 47 (2010) 441.