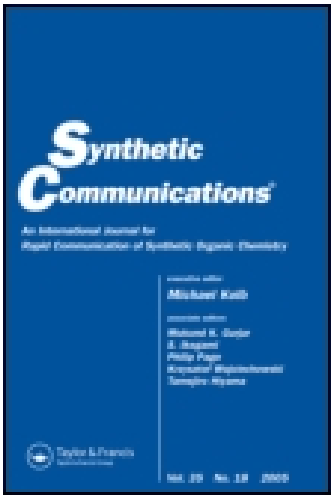


Synthetic Communications: An International Journal for Rapid Communication of Synthetic Organic Chemistry

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/lsyc20

Facile Synthesis of Tetrahydroimidazolpyridinones via an MCR Involving 6-Cl-PMNI, Aldehydes, and Meldrum's Acid

Bin Hu [a], Yi-Yue Zhou [a], Wenting Du [a], Xiaoyong Xu [a], Zhong Li [a] & Wei-Ping Deng [a b]

[a] School of Pharmacy, East China University of Science and Technology, Shanghai, China

[b] Department of Pharmacy, Zhejiang Medical College, Hangzhou, China

Published online: 21 Mar 2011.

To cite this article: Bin Hu, Yi-Yue Zhou, Wenting Du, Xiaoyong Xu, Zhong Li & Wei-Ping Deng (2011) Facile Synthesis of Tetrahydroimidazolpyridinones via an MCR Involving 6-Cl-PMNI, Aldehydes, and Meldrum's Acid, Synthetic Communications: An International Journal for Rapid Communication of Synthetic Organic Chemistry, 41:8, 1112-1118, DOI: 10.1080/00397911003797791

To link to this article: http://dx.doi.org/10.1080/00397911003797791

Synthetic Communications®, 41: 1112–1118, 2011

ISSN: 0039-7911 print/1532-2432 online
DOI: 10.1080/00397911003797791

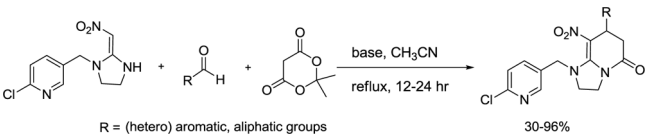
Taylor & Francis
Taylor & Francis Group

FACILE SYNTHESIS OF TETRAHYDROIMIDAZOLPYRIDINONES VIA AN MCR INVOLVING 6-Cl-PMNI, ALDEHYDES, AND MELDRUM'S ACID

Bin Hu,[1] Yi-Yue Zhou,[1] Wenting Du,[1] Xiaoyong Xu,[1] Zhong Li,[1] and Wei-Ping Deng[1,2]

[1]*School of Pharmacy, East China University of Science and Technology, Shanghai, China*
[2]*Department of Pharmacy, Zhejiang Medical College, Hangzhou, China*

GRAPHICAL ABSTRACT



Abstract *Tetrahydroimidazolpyridinones* **2***, which are new, highly bioactive potential insecticides, were prepared in 30–96% yield through a multicomponent reaction (MCR) of 6-Cl-PMNI, aldehydes, and Meldrum's acid using catalytic* Et_3N *or* K_2CO_3 *in anhydrous* CH_3CN *at refluxing temperature.*

Keywords 6-Cl-PMNI; insecticide; Meldrum's acid; multicomponent reaction; tetrahydroimidazolpyridinone

INTRODUCTION

Nitromethylene neonicotinoids **1** (Fig. 1) were recently found in our laboratory to be potential new insecticides possessing greater activity against imidacloprid-resistant brown planthopper than imidacloprid.[1] Its synthesis proceeded through Michael addition of β-nitroketeneaminal (6-Cl-PMNI) **3** to α,β-unsaturated aldehydes and subsequent ring closure.[1,2] However, this reaction suffers from relatively poor yield, low reactivities to cinnamic aldehyde and its analogs, and limited availability of corresponding α,β-unsaturated aldehydes. On the other hand, because of the chemical instability of **1** arising from the dehydration of the 2-hydroxyl group, we envisaged that a more stable tetrahydroimidazolpyridinone **2** might be a potential skeleton as a potent insecticide. It has been reported[3] that tetrahydroimidazolpyridinones can be prepared through Michael addition of

Received December 13, 2009.

Address correspondence to Wenting Du, School of Pharmacy, East China University of Science and Technology, 130 Meilong Road, Shanghai 200237, China. E-mail: duwt@ecust.edu.cn

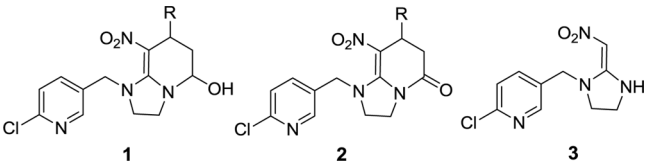


Figure 1. The structures of neonicotinoids **1**, tetrahydroimidazolpyridinone **2**, and β-nitroketeneaminal **3**.

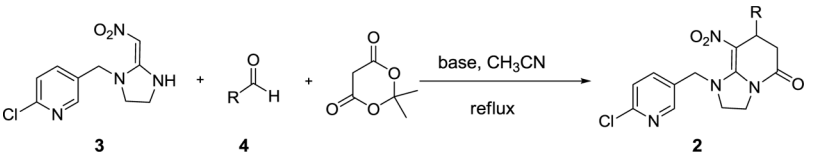
2-(nitromethylene)imidazolidine with ethyl acrylate and subsequent ring closure. However, the reaction of *N*-methyl-2-(nitromethylene)imidazolidine with methyl acrylate provided the corresponding product in 32% yield,[4] and notably no other β-substituted α,β-unsaturated substrates were reported to perform this reaction, probably because of the low reactivity of both *N*-substituted 2-(nitromethylene)imidazolidine and β-substituted α,β-unsaturated carboxylate. As a matter of fact, the reaction of **3** with methyl cinnamate (or crotonate) turned out to give only a trace amount of the desired products under the same reaction condition.[5] Therefore, to further develop our newly discovered insecticide and its analogs, a versatile and efficient synthetic protocol was highly demanded.

Recently, Yu and coworkers[6] reported a Meldrum's acid–involved one-pot reaction of heterocyclic ketene aminals (HKAs) and aldehydes that smoothly provided a similar imidazolpyridinone skeleton, presumably via an aza-ene pathway. It is noted that, in this case, *N*-unprotected HKAs with α,β-unsaturated ketone or ester were used, which are normally more reactive than *N*-protected HKAs, such as the 6-Cl-PMNI used in our case. However, it was still believed that the more reactive alkylidene Meldrum's acids generated in situ facilitate the Michael addition to the *N*-protected HKAs. Therefore, we describe herein our preliminary results on the synthesis of **2** by a multicomponent reaction (MCR) of Meldrum's acid, aldehydes, and **3**.

RESULTS AND DISCUSSION

For nitromethylene neonicotinoids **1**, insecticidal activity was found to be optimal when R is methyl group. Therefore, we initially employed acetaldehyde as substrate to test the MCR. Unfortunately, no desired product was found under the optimal reaction condition reported by Yu et al. We then turned our attention to using aromatic aldehyde to test our idea. A series of aromatic aldehydes were thus employed for this reaction using catalytic (0.1 equiv.) triethylamine in anhydrous acetonitrile at refluxing temperature, and the results are summarized in Table 1. It is encouraging that (hetero) aromatic aldehydes with both electron-withdrawing and electron-donating groups proceeded smoothly and resulted in good to excellent yields without apparent difference (entries 1–11). Substrate salicylaldehyde **4h** with an unprotected OH group also gave corresponding product **2h** in 78% yield (entry 8). With such a positive result with aromatic aldehyde, we came back to solving the problem with aliphatic aldehydes. According to the mechanism proposed by Yu et al., the key intermediate would be enzylidene–Meldrum's acid, and the role

Table 1. Three-component reaction of various aldehydes, Meldrum's acid, and **3**



3 **4** **2**

Entry	Aldehyde	R	Product	Base (amount)	Reaction time (h)	Yield (%)[a]
1	**4a**	Ph	**2a**	Et_3N (0.1 eq)	12	89
2	**4b**	4-BrPh	**2b**	Et_3N (0.1 eq)	12	85
3	**4c**	4-NO_2Ph	**2c**	Et_3N (0.1 eq)	12	90
4	**4d**	2-NO_2Ph	**2d**	Et_3N (0.1 eq)	12	91
5	**4e**	2-MePh	**2e**	Et_3N (0.1 eq)	12	96
6	**4f**	4-MeOPh	**2f**	Et_3N (0.1 eq)	12	90
7	**4g**	3,4-CH_2O_2Ph	**2g**	Et_3N (0.1 eq)	12	94
8	**4h**	2-OHPh	**2h**	Et_3N (0.1 eq)	12	78
9	**4i**	3-Indole	**2i**[b]	Et_3N (0.1 eq)	12	85
10	**4j**	2-Furan	**2j**[b]	Et_3N (0.1 eq)	12	79
11	**4k**	2-thiophene	**2k**	Et_3N (0.1 eq)	12	72
12	**4l**	Ethyl	**2l**[b]	Et_3N (0.1 eq)	12	Trace
13	**4l**	Ethyl	**2l**[b]	Et_3N (1 eq)	24	Trace
14	**4l**	Ethyl	**2l**[b]	Et_3N (2 eq)	24	16
15	**4l**	Ethyl	**2l**[b]	K_2CO_3 (2 eq)	24	53
16	**4m**	$BnOCH_2$	**2m**	Et_3N (2 eq)	24	15
17	**4m**	$BnOCH_2$	**2m**	K_2CO_3 (2 eq)	24	30
18	**4n**	Me	**2n**	K_2CO_3 (2 eq)	24	31

[a]Yield of isolated and purified products.
[b]Showing high insecticidal activities against pea aphids.[5]

of base is to form the key intermediate. Therefore, a base screening was carried out to find the proper reaction condition for aliphatic aldehydes. It is interesting that 16% yield of corresponding product **4l** was obtained, when the amount of triethylamine was increased to 2 equiv. (entries 12–14). To our amazement, further optimization of various bases showed that 2 equiv. of K_2CO_3 was the optimal condition and gave corresponding products in moderate yields (entries 15, 17, and 18, up to 53%). On the other hand, the reaction of preprepared benzylidene–Meldrum's acid with **3** under the same reaction condition resulted in comparable yield (90%), as expected, which further provided experimental evidence for the aza–ene mechanism.[6]

CONCLUSIONS

In summary, we have developed a facile synthesis of tetrahydroimidazolpyridinones **2** in 30–94% yields through an MCR of 6-Cl-PMNI, aldehydes, and Meldrum's acid using catalytic Et_3N or K_2CO_3 in anhydrous CH_3CN at refluxing temperature. Taking account of the high efficiency of this MCR and the ready availability of various aldehydes, we believe that our synthetic protocol would be of great

help in synthesizing a library of tetrahydroimidazolpyridinones with various substituents to seek out new potent insecticides. Further work is currently ongoing in our laboratory to extend the synthetic application of this new method and to test the insecticidal activities against insects, especially the resistant strains.

EXPERIMENTAL

All solvents were distilled under standard procedures prior to use under a nitrogen atmosphere. (For example, CH_3CN was distilled from CaH_2). ^1H (400 MHz) chemical shifts are reported in $CDCl_3$ 7.27 ppm for ^1H, and standards and coupling constants (J) are reported in hertz (Hz). The following abbreviations are used to designate signal multiplicity: s, singlet; d, doublet; t, triplet; q, quartet; m, multiplet; and br, broad.

General Procedure for the MCR of Various Aldehydes, Meldrum's Acid, and 6-Cl-PMNI 3

Compound **3** was prepared according to previously reported procedures.[2,7] Compound **3** (0.39 mmol), Meldrum's acid (0.47 mmol), and aldehyde **4** (0.47 mmol) were added to anhydrous acetonitrile (1 mL) respectively with Et_3N (0.1 eq) or K_2CO_3 (2 eq) as a catalyst under the nitrogen atmosphere. After stirring under reflux for 12 or 24 h, the reaction mixture was concentrated in vacuo, and the residue was purified via column chromatography (EtOAc–petroleum ether).

All products gave satisfactory analytical data.

Representative Data

All products gave satisfactory analytical data.

Characterization data of product 2a (entry 1, R = Ph). Mp 219–220 °C. IR (KBr): 3440.38, 1701.39, 1590.15, 1392.21, 1319.92, 1273.09, 1216.87 cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): $\delta = 8.27$ (d, $J = 2.4$ Hz, 1H), 7.49 (dd, $J = 2.4$, 8.0 Hz, 1H), 7.27–7.26 (m, 3H), 7.18 (d, $J = 8.0$ Hz, 1H), 7.11–7.09 (m, 2H), 4.90–4.81 (m, 3H), 4.15–4.09 (m, 1H), 3.96–3.79 (m, 2H), 3.73–3.67 (m, 1H), 3.06 (d, $J = 4.4$ Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): $\delta = 167.62$, 151.72, 151.53, 149.11, 140.98, 138.61, 129.32, 128.95, 127.37, 125.95, 124.69, 110.23, 51.90, 50.14, 41.12, 38.36, 37.33. HRMS (EI): m/z [M^+] calcd. for $C_{19}H_{17}ClN_4O_3$: 384.0989; found: 384.0990.

Characterization data of product 2b (entry 2, R = 4-BrPh). Mp 108–111 °C. ^1H NMR (400 MHz, $CDCl_3$): δ 8.27 (s, 1H), 7.51 (d, $J = 8.4$ Hz, 1H), 7.37 (d, $J = 8.4$ Hz, 2H), 7.21 (d, $J = 8.4$ Hz, 1H), 6.95 (d, $J = 8.4$ Hz, 2H), 4.88–4.79 (m, 3H), 4.12–4.08 (m, 1H), 3.95–3.84 (m, 2H), 3.76–3.73 (m, 1H), 3.04 (m, 2H).

Characterization data of product 2c (entry 3, R = 4-NO_2Ph). Mp 233–237 °C. ^1H NMR (400 MHz, DMSO-d_6): δ 8.30 (d, $J = 1.6$ Hz, 1H), 8.07 (d, $J = 8.8$ Hz, 2H), 7.74 (dd, $J = 2.8$, 2.4 Hz, 1H), 7.40 (d, $J = 8.4$ Hz, 1H), 7.21

(d, $J = 8.4$ Hz, 2H), 4.90 (s, 2H), 4.74 (d, $J = 6.8$ Hz, 1H), 4.05–3.86 (m, 4H), 3.25 (dd, $J = 8.0$, 7.2 Hz, 1H), 2.80 (d, $J = 17.2$ Hz, 1H).

Characterization data of product 2d (entry 4, R = 2-NO_2Ph). Mp 107–109 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.39 (s, 1H), 7.91 (s, 1H), 7.68 (d, $J = 7.2$ Hz, 1H), 7.41 (s, 2H), 7.28 (d, $J = 6.0$ Hz, 1H), 6.81 (s, 1H), 5.27 (d, $J = 8.8$ Hz, 1H), 4.90 (dd, $J = 15.6$ Hz, 2H), 4.14–3.89 (m, 4H), 3.24 (dd, $J = 9.2$, 8.8 Hz, 1H), 2.93 (d, $J = 17.6$ Hz, 1H).

Characterization data of product 2e (entry 5, R = 2-MePh). Mp 122–125 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.32 (s, 1H), 7.60 (s, 1H), 7.19–6.90 (m, 4H), 6.42 (s, 1H), 4.80 (s, br, 3H), 4.03–3.72 (m, 4H), 2.98 (d, $J = 6.4$ Hz, 1H), 2.61 (d, $J = 16.3$ Hz, 1H), 2.34 (s, 3H).

Characterization data of product 2f (entry 6, R = 4-MeOPh). Mp 164–166 °C. IR (KBr): 3440.38, 1704.76, 1596.77, 1292.07, 1224.58 cm^{-1}. ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.27 (s, 1H), 7.50 (d, $J = 8.0$ Hz, 1H), 7.19 (d, $J = 8.4$ Hz, 1H), 6.97 (d, $J = 8.4$ Hz, 2H), 6.78 (d, $J = 8.8$ Hz, 2H), 4.89–4.80 (m, 3H), 4.13–4.07 (m, 1H), 3.96–3.81 (m, 2H), 3.79 (s, 3H), 3.74–3.69 (m, 1H), 3.01 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 167.68, 158.74, 151.55, 151.48, 149.16, 138.64, 132.98, 129.51, 127.03, 124.60, 114.23, 110.50, 55.28, 51.85, 50.33, 41.14, 37.73, 37.56. HRMS (EI): m/z [M^+] calcd. for $C_{20}H_{19}ClN_4O_4$: 414.1095; found: 414.1097.

Characterization data of product 2g (entry 7, R = 3,4-CH_2O_2Ph). Mp 218–221 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.32 (s, 1H), 7.78 (d, $J = 6.4$ Hz, 1H), 7.44 (d, $J = 7.2$ Hz, 1H), 6.68 (d, $J = 6.8$ Hz, 1H), 6.57 (s, 1H), 6.30 (d, $J = 6.80$ Hz, 1H), 5.96 (s, 2H), 4.89 (s, 2H), 4.52 (d, $J = 4.4$ Hz, 1H), 3.94–3.82 (m, 4H), 3.11 (dd, $J = 5.6$ Hz, 1H), 2.71 (d, $J = 16.4$ Hz, 1H).

Characterization data of product 2h (entry 8, R = 2-OHPh). Mp 239–240 °C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.69 (s, 1H), 8.39 (s, 1H), 7.84 (d, $J = 8.0$ Hz, 1H), 7.49 (dd, $J = 8.4$ Hz, 1H), 7.01 (t, $J = 7.2$ Hz, 1H), 6.77 (d, $J = 7.6$ Hz, 1H), 6.52 (t, $J = 7.2$ Hz, 1H), 6.30 (d, $J = 7.2$ Hz, 1H), 4.94 (s, 2H), 4.71 (d, $J = 7.6$ Hz, 1H), 4.03–3.75 (m, 4H), 3.08 (dd, $J = 7.6$, 8.0 Hz, 1H), 2.61 (d, $J = 16.4$ Hz, 1H).

Characterization data of product 2i (entry 9, R = 3-Indole). Mp 206–211 °C. IR (KBr): 3417.24, 1693.19, 1602.56, 1398.14, 1284.36, 1216.86 cm^{-1}. ^{1}H NMR (100 MHz, DMSO-d_6): δ = 10.86 (s, 1H), 8.37 (s, 1H), 7.67 (dd, $J = 2.0$ Hz, 8.0 Hz, 1H), 7.57 (d, $J = 8.0$ Hz; 1H), 7.35 (d, $J = 8.0$ Hz, 1H), 7.26 (d, $J = 8.4$ Hz, 1H), 7.09 (t, $J = 7.6$ Hz, 1H), 6.97 (t, $J = 7.6$ Hz, 1H), 6.83 (s, 1H), 4.98–4.80 (m, 3H), 3.98–3.75 (m, 4H), 3.16 (dd, $J = 6.4$ Hz, 1H), 2.90–2.86 (m, 1H); ^{13}C NMR (100 MHz; DMSO-d_6): δ = 168.59, 153.21, 149.86, 149.66, 139.46, 137.19, 131.40, 126.24, 124.27, 121.82, 121.57, 119.14, 119.08, 115.33, 112.01, 108.95, 51.82, 50.50, 41.71, 38.29, 32.03. HRMS (EI): m/z [M^+] calcd. for $C_{21}H_{18}ClN_5O_3$: 423.1098; found: 423.1105.

Characterization data of product 2j (entry 10, R = 2-Furan). Mp 86–90 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.28 (s, 1H), 7.53 (d, $J = 8.4$ Hz, 1H),

7.27–7.24 (m, 1H), 6.28 (s, 1H), 6.57 (s, 1H), 6.03 (d, $J = 2.8$ Hz, 1H), 4.92–4.89 (m, 2H), 4.81 (d, $J = 15.6$ Hz, 1H), 4.13–4.08 (m, 1H), 3.92–3.84 (m, 2H), 3.72–3.69 (m, 1H), 2.99 (d, $J = 3.2$ Hz, 2H).

Characterization data of product 2k (entry 11, R = 2-Thiophene). Mp 86–88 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.28 (d, $J = 1.6$ Hz, 1H), 7.53 (d, $J = 2.0$ Hz, 1H), 7.23 (d, $J = 8.0$ Hz, 1H), 7.16 (d, $J = 5.2$ Hz, 1H), 6.91 (t, $J = 3.6$, 5.2 Hz, 1H), 6.78 (d, $J = 3.2$ Hz, 1H), 5.08 (t, $J = 4.8$, 3.2 Hz, 1H), 4.84 (s, 2H), 4.11 (m, 1H), 3.90–3.83 (m, 2H), 3.66–3.65 (m, 1H), 3.08 (d, $J = 5.6$ Hz, 2H).

Characterization data of product 2l (entry 12, R = Ethyl). Mp 132–135 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.33 (s, 1H), 7.73 (d, $J = 8.4$ Hz, 1H), 7.34 (d, $J = 8.4$ Hz, 1H), 4.89 (dd, $J = 15.2$, 15.6 Hz, 2H), 4.10–4.06 (m, 1H), 3.94–3.84 (m, 2H), 3.75–3.72 (m, 1H), 3.47 (dd, $J = 6.0$, 6.8 Hz, 1H), 2.76 (dd, $J = 6.4$, 6.8 Hz, 1H), 2.63 (d, $J = 17.2$ Hz, 1H), 1.47–1.36 (m, 2H), 0.84 (t, $J = 7.2$ Hz, 3H).

Characterization data of product 2m (entry 16, R = $BnOCH_2$). Mp 53–55 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.21 (s, 1H), 7.52 (dd, $J = 2.4$ Hz, 1H), 7.37–7.27 (m, 5H), 7.13 (d, $J = 8.0$ Hz, 1H), 4.77 (dd, $J = 15.6$ Hz, 2H), 4.41 (dd, $J = 11.2$ Hz, 2H), 3.93 (m, 1H), 3.80–3.75 (m, 2H), 3.61–3.57 (m, 2H), 3.47–3.44 (m, 2H), 2.86–2.84 (m, 1H), 2.72 (d, $J = 17.2$ Hz, 1H).

Characterization data of product 2n (entry 19, R = Methyl). Mp 204–207 °C. IR (KBr): 3436.53, 1706.69, 1585.20, 1396.21, 1309.43, 1197.58, 1141.65 cm^{-1}. ^{1}H NMR (100 MHz, $CDCl_3$): δ = 8.33 (d, $J = 2.4$ Hz, 1H), 7.72 (dd, $J = 2.4$, 8.4 Hz, 1H), 7.35 (d, $J = 8.0$ Hz, 1H), 4.89 (d, $J = 15.6$ Hz, 1H), 4.78 (d, $J = 15.6$ Hz, 1H), 4.16–4.05 (m, 1H), 3.94–3.82 (m, 2H), 3.73–3.64 (m, 2H), 2.81 (dd, $J = 6.8$, 17.2 Hz, 1H), 2.53 (dd, $J = 1.6$, 17.2 Hz, 1H), 1.12 (d, $J = 7.2$ Hz, 3H). ^{13}C NMR (100 MHz, $CDCl_3$): δ = 167.79, 151.61, 150.48, 149.00, 138.77, 129.68, 124.62, 111.58, 52.03, 50.08, 41.10, 38.50, 29.31, 19.19. HRMS (EI): m/z [M^+] calcd. for $C_{14}H_{15}ClN_4O_3$: 322.0833; found: 322.0836.

ACKNOWLEDGMENTS

This work was financially supported by the Shanghai Committee of Science and Technology (No. 064319022), the New Century Excellent Talents in University, the Ministry of Education, China (No. NCET-07-0283), the Natural Science Foundation of China (No. NSFC 20602011), and the "111" Project (No. B07023).

REFERENCES

1. Shao, X.; Zhang, W.; Peng, Y.; Li, Z.; Tian, Z.; Qian, X. cis-Nitromethylene neonicotinoids as new nicotinic family: Synthesis, structural diversity, and insecticidal evaluation of hexahydroimidazo[1,2-α]pyridine. *Bioorg. Med. Chem. Lett.* **2008**, *18*, 6513.
2. Tian, Z.; Shao, X.; Li, Z.; Qian, X.; Huang, Q. Synthesis, insecticidal activity, and QSAR of novel nitromethylene neonicotinoids with tetrahydropyridine fixed *cis* configuration and exo-ring ether modification. *J. Agric. Food Chem.* **2007**, *55*, 2288.

3. Tokumitsu, T. Reaction of β-nitroketeneaminal with olefins bearing electron-withdrawing group and aldehydes. *Bull. Chem. Soc. Jpn.* **1990**, *63*, 1921.
4. (a) Huang, Z.-T.; Tzai, L.-H. Synthesis of ketene aminals with an imidazolidine ring by condensation of 4,5-dihydro-2-(methylthio)-1*H*-imidazoles with active methylene compounds and some addition and cyclocondensation reactions. *Chem. Ber.* **1986**, *119*, 2208; (b) Huang, Z.-T.; Wang, M.-X. Heterocyclic ketene aminals. *Heterocycles* **1994**, *37*, 1233.
5. Zhou, Y.-Y. Studies towards the synthesis of chiral neonicotine compounds. MS thesis, 2009.
6. Yu, C.-Y.; Yang, P.-H.; Zhao, M.-X.; Huang, Z.-T. A novel one-pot reaction of heterocyclic ketene aminals: Synthesis of a small library of tetrahydropyridinone-fused 1,3-diazaheterocycles. *Synlett* **2006**, 1835.
7. (a) Li, Z.; Qian, X.; Shao, X.; Xu, X.; Tian, Z.; Huang, Q. Preparation method and use of compounds having high biocidal activities. PCT Int. Appl. WO Patent 2007101369, **2007**; *Chem. Abstr.* **2007**, *147*, 344093.