Chinese Chemical Letters xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Chinese Chemical Letters

journal homepage: www.elsevier.com/locate/cclet

Original article

Tandem intramolecular Diels–Alder/retro-Diels–Alder cycloaddition of 2*H*-chromen-2-one as dienes with the expulsion of CO_2

Jian Zhang, Jin-Long Wu *

Laboratary of Asymmetry Catalysis and synthesis, Department of Chemistry, Zhejiang University, Hangzhou 310027, China

ARTICLE INFO

Article history:
Received 15 February 2016
Received in revised form 2 March 2016
Accepted 7 March 2016
Available online xxx

Keywords:
2*H*-Chromen-2-one
Intramolecular Diels–Alder
Retro-Diels–Alder
Benzo[*f*]isoindol-1-one
Dihydroxylation

ABSTRACT

To study the intramolecular Diels–Alder cycloadditon of 2*H*-chromen-2-one as a diene, a series of chiral *N*-allyl-*N*-benzylamides that contain a 2*H*-chromen-2-one moiety were designed for the synthesis of benzo[*f*]isoindol-1-ones *via* an intramolecular Diels–Alder and a subsequent retro-Diels–Alder cycloaddition with the expulsion of CO_2. Both the yield (80%–89%) and absolute stereocontrol of the tandem reaction were high when an electron-withdrawing group was attached to the dienophile. The double bond in the styrene substructure remained in the products could be further derivatized by dihydroxylation.

© 2016 Chinese Chemical Society and Institute of Materia Medica, Chinese Academy of Medical Sciences. Published by Elsevier B.V. All rights reserved.

1. Introduction

Because of the stability of an aromatic ring, the exocyclic double bond in styrene commonly reacts as a dienophile in [4 + 2] cycloaddition reactions [1] and it is quite difficult for styrene to participate as a diene in thermal Diels–Alder cycloaddition unless extremely reactive dienophiles are employed [2]. 2*H*-pyran-2-one and its derivatives are a class of important electron-rich diene in the Diels–Alder reaction [3] and are widely used in organic synthesis [4]. There is a feature of this type of dienes in the Diels–Alder reaction: Retro-Diels–Alder reaction will happen under certain conditions by eliminating one molecule of CO_2, which introduces a carbon–carbon double bond in the ring [5]. Several reports claimed that 2*H*-chromen-2-one, a kind of 2*H*-pyran-2-one fused with a benzene ring, can be used in the Diels–Alder cycloaddition as a dienophile [6]. However, only one case where it was used as a diene was reported in the tandem intramolecular Diels–Alder (IMDA) and retro-Diels–Alder cycloaddition with acetylene as shown in Scheme 1 [7]. The reaction must be carried out in a sealed tube placed in a 300 °C bath, and surprisingly, the acetylenic hydrogen replaced by an electron-withdrawing group (EWG), carboethoxy, **1e** failed to cyclize to form **2e**. In our continuous works on synthesis of polycyclic compounds using IMDA reaction [8], herein, we report the tandem IMDA and retro-Diels–Alder reaction of chiral *N*-(4-methoxybenzyl)-2-oxo-*N*-(1-phenylbut-3-en-2-yl)-2*H*-chromene-3-carboxamides for stereocontrolled synthesis of 3-benzyl-2-(4-methoxybenzyl)-2,3,3a,4-tetrahydro-1*H*-benzo[*f*]isoindol-1-ones under relatively mild conditions.

2. Experimental

2.1. General procedure for the synthesis of *4a–g*

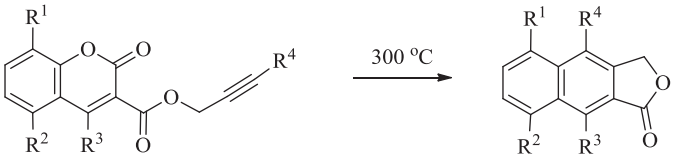
A solution of **3** (0.1 mmol) in CH_3CN (2 mL) in a sealed vial was heated in an oil bath at 160 °C. After the reaction completed (monitored by TLC analysis), the solvent was removed under reduced pressure. The residue was purified by chromatography on silica gel (ethyl acetate/petroleum ether = 1/3) to give **4** (Scheme 2).

Ethyl (3*S*,3a*S*,4*R*)-3-benzyl-2-(4-methoxybenzyl)-4-methyl-1-oxo-2,3,3a,4-tetrahydro-1*H*-benzo[*f*]isoindole-4-carboxylate **4a**: White crystalline solid, yield 82%, mp 204–205 °C (EtOAc–hexane); $[\alpha]_D^{25}$ +13.9 (*c* 1.0, $CHCl_3$); R_f = 0.20 (20% EtOAc in hexane); IR (film, cm^{-1}): 3061, 3028, 2979, 2933, 2836, 1721, 1685, 1611, 1512, 1447, 1409, 1298, 1247, 1174, 1093, 1031, 909, 755, 731, 700; ^1H NMR (400 MHz, $CDCl_3$): δ 7.29–7.21 (m, 6H), 7.19 (d, 1H, *J* = 3.2 Hz), 7.11–7.09 (m, 2H), 6.94–6.90 (m, 3H), 6.78 (d, 2H, *J* = 8.8 Hz), 5.34 and 3.91 (ABq, 2H, *J* = 14.8 Hz), 4.32 (dq, 1H, *J* = 10.8, 7.2 Hz), 4.19 (dq, 1H, *J* = 10.8, 7.2 Hz), 3.79 (s, 3H), 3.76 (dt,

* Corresponding author.
E-mail address: wyz@zju.edu.cn (J.-L. Wu).

http://dx.doi.org/10.1016/j.cclet.2016.03.034
1001-8417/© 2016 Chinese Chemical Society and Institute of Materia Medica, Chinese Academy of Medical Sciences. Published by Elsevier B.V. All rights reserved.

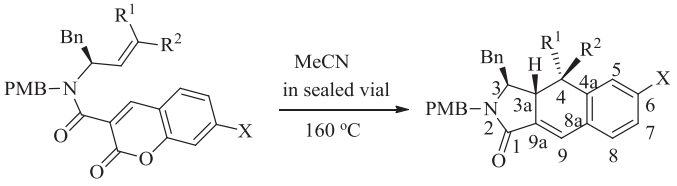
Please cite this article in press as: J. Zhang, J.-L. Wu, Tandem intramolecular Diels–Alder/retro-Diels–Alder cycloaddition of 2*H*-chromen-2-one as dienes with the expulsion of CO_2, Chin. Chem. Lett. (2016), http://dx.doi.org/10.1016/j.cclet.2016.03.034



1a: R^1 = H; R^2 = H; R^3 = H; R^4 = H
1b: R^1 = MeO; R^2 = H; R^3 = H; R^4 = H
1c: R^1 = H; R^2 = MeO; R^3 = H; R^4 = H
1d: R^1 = H; R^2 = H; R^3 = MeO; R^4 = H
1e: R^1 = H; R^2 = H; R^3 = H; R^4 = CO_2Et

2a: 58%
2b: 61%
2c: 51%
2d: 47%
2e: 0%

Scheme 1. Tandem IMDA and retro-Diels–Alder reaction of 2*H*-chromen-2-one with acetylene.



3a-g **4a-g**

Scheme 2. Synthesis of compounds **4a–g**.

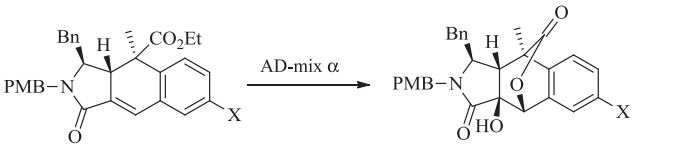
1H, *J* = 5.2, 3.6 Hz), 3.65 (dd, 1H, *J* = 4.8, 3.2 Hz), 3.12 (dd, 1H, *J* = 15.2, 3.2 Hz), 2.66 (dd, 1H, *J* = 15.2, 5.6 Hz), 1.31 (t, 3H, *J* = 7.2 Hz), 1.11 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 175.8, 167.0, 159.0, 139.6, 136.4, 132.2, 131.6, 129.7, 129.6 (×2), 129.2 (×2), 129.1, 128.6 (×2), 127.9, 127.7, 126.8, 126.0, 125.4, 114.0 (×2), 61.6, 55.6, 55.2, 51.2, 44.2, 42.9, 38.2, 17.2, 14.2; HRMS (+EI) calculated for $C_{31}H_{31}NO_4^+$ (M^+) 481.2253; found 481.2246.

(3*S*,3a*R*)-3-Benzyl-2-(4-methoxybenzyl)-4,4-dimethyl-2,3,3a,4-tetrahydro-1*H*-benzo[*f*]isoindol-1-one **4e**: Colorless oil, yield 11%, $[\alpha]_D^{25}$ +7.6 (*c* 1.0, $CHCl_3$); R_f = 0.19 (25% EtOAc in hexane); IR (film, cm^{-1}): 3066, 3030, 3003, 2965, 2929, 2866, 2834, 1676, 1611, 1516, 1453, 1409, 1304, 1239, 1174, 1031, 834, 811, 751, 703; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.27–7.19 (m, 8H), 7.09–7.04 (m, 4H), 6.85 (d, 2H, *J* = 8.8 Hz), 5.40 and 4.01 (ABq, 2H, *J* = 14.8 Hz), 3.81 (s, 3H), 3.67–3.63 (m, 1H), 3.02–2.92 (m, 2H), 2.78–2.75 (m, 1H), 1.03 (s, 3H), 0.74 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 167.4, 159.1, 144.8, 136.6, 133.7, 132.5, 129.8 (×2), 129.5, 129.4 (×2), 128.9, 128.7 (×2), 128.1, 126.9, 126.8, 126.7, 123.6, 114.0 (×2), 56.2, 55.3, 46.0, 44.1, 40.1, 37.5, 24.3, 21.6; HRMS (+EI) calculated for $C_{29}H_{29}NO_2^+$ (M^+) 423.2198; found 423.2204.

2.2. General procedure for the synthesis of ***5a–c***

To a solution of **4** (0.1 mmol) in a mixed solvent of 2-methyl-2-propanol (1 mL) and water (1 mL) were added methanesulfonamide (0.1 mmol) and AD-mix α (145 mg). After stirring at 0 °C for 10–24 h, the reaction mixture was quenched with saturated aqueous $Na_2S_2O_3$ (5 mL) and stirred at room temperature for another 20 min, then the mixture was extracted with ethyl acetate (10 mL × 3). The combined organic phase was dried over anhydrous Na_2SO_4, filtrated and concentrated under reduced pressure. The residue was purified by chromatography on silica gel (EtOAc/PE = 1/1) to give **5** (Scheme 3).

(1*S*,3a*S*,4*R*,9*R*,9a*S*)-1-Benzyl-3a-hydroxy-2-(4-methoxybenzyl)-9-methyl-3a,4,9,9a-tetrahydro-1*H*-4,9-(epoxymethano)benzo[*f*]-isoindole-3,10(2H)-dione **5a**: White crystalline solid, yield 90%; mp 102–107 °C (EtOAc–hexane); $[\alpha]_D^{25}$ −4.2 (*c* 1.0, $CHCl_3$); R_f = 0.22



4a-c

5a X= H, t =10 h, yiled 90%
5b X= Cl, t =12 h, yiled 83%
5c X= NO_2, t =24 h, yiled 87%

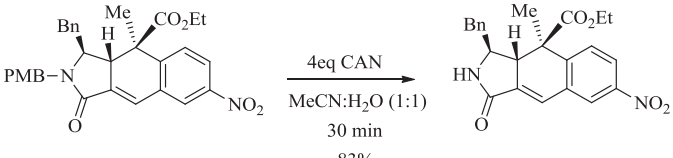
Scheme 3. Synthesis of compounds **5a–c**.

(50% EtOAc in hexane); IR (film, cm^{-1}): 3387, 3063, 3033, 2980, 2938, 2840, 1757, 1676, 1513, 1453, 1373, 1245, 1043, 1004, 843, 778, 745, 701; ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.48–7.46 (m, 1H), 7.41–7.34 (m, 2H), 7.33–7.25 (m, 3H), 6.99–6.94 (m, 3H), 6.67 (d, 2H, *J* = 8.8 Hz), 6.23 (d, 2H, *J* = 8.8 Hz), 5.64 (s, 1H), 4.78 and 3.72 (ABq, 2H, *J* = 14.8 Hz), 3.83 (s, 3H), 3.15 (dd, 1H, *J* = 13.2, 3.6 Hz), 2.73 (ddd, 1H, *J* = 9.6, 3.6, 2.4 Hz), 2.55 (dd, 1H, *J* = 13.2, 9.6 Hz), 2.34 (d, 1H, *J* = 2.4 Hz), 0.76 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 173.3, 170.7, 159.0, 135.5, 135.3, 134.3, 129.8, 129.7 (×2), 129.4 (×2), 129.0 (×2), 128.1, 127.4, 125.4, 125.3, 123.9, 114.1 (×2), 82.1, 81.5, 58.1, 55.3, 47.8, 47.6, 43.8, 39.8, 12.6; HRMS (+EI) calculated for $C_{29}H_{27}NO_5^+$ (M^+) 469.1889; found 469.1881.

2.3. Synthesis of ***6c*** [9]

Some biological studies on the approved and developing γ-lactam drugs showed that the N-H type lactams are more active than the corresponding *N*-substituted one [11], therefore we completed the transformation of **4c** to **6c** as an example using ceric (IV) ammonium nitrate (CAN). To a solution of **4c** (72 mg, 0.137 mmol) in a mixed solvent of CH_3CN (3 mL) and H_2O (1 mL) was added ceric ammonium nitrate (298 mg, 0.544 mmol) in one portion. After stirring for 30 min at room temperature, H_2O (3 mL) was added and then the mixture was extracted with ethyl acetate (10 mL × 3). The combined organic layer was washed with saturated aqueous $NaHCO_3$ (1.5 mL × 3) and brine (1.5 mL). The organic phase was dried over anhydrous Na_2SO_4, filtrated and concentrated under reduced pressure. The residue was purified by chromatography on silica gel (ethyl acetate/petroleum = 1/1) to give **6c** (46 mg, 83%) as a colorless oil (Scheme 4).

Ethyl (3*S*)-3-benzyl-4-methyl-7-nitro-1-oxo-2,3,3a,4-tetrahydro-1*H*-benzo[*f*]isoindole-4-carboxylate **6c**: Yield 83%; $[\alpha]_D^{15}$ +2.1 (*c* 1.0, $CHCl_3$); R_f = 0.29 (50% EtOAc in hexane); IR (film, cm^{-1}): 3405, 3214, 3066, 3024, 2979, 2935, 2873, 1730, 1697, 1608, 1584, 1519, 1456, 1346, 1292, 1239, 1090, 1050, 1016, 909, 825, 736, 698; ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.14–8.12 (m, 2H), 7.35 (t, 2H, *J* = 7.2 Hz), 7.29 (d, 1H, *J* = 7.2 Hz), 7.25 (d, 1H, *J* = 3.2 Hz), 7.21 (d, 1H, *J* = 7.2 Hz), 7.19 (d, 2H, *J* = 8.0 Hz), 6.18 (br s, 1H), 4.47–4.34 (m, 2H), 3.98–3.93 (m, 1H), 3.70 (dd, 1H, *J* = 6.0, 3.6 Hz), 2.98 (dd, 1H, *J* = 13.6, 2.8 Hz), 2.60 (dd, 1H, *J* = 13.6, 10.0 Hz), 1.39 (t, 3H, *J* = 7.2 Hz), 1.38 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$): δ 174.7, 167.1, 147.3, 146.1, 136.4, 135.3, 132.8, 129.1 (×2), 129.0 (×2), 127.3, 126.7, 124.32, 124.29, 123.4, 62.3, 54.6, 51.0, 46.4, 42.0, 17.8, 14.2; HRMS (+EI) calcd. for $C_{23}H_{22}N_2O_5^+$ (M^+) 406.1529; found 406.1538.



4c **6c**

Scheme 4. Transformation from **4c** to **6c** by removing PMB with CAN.

Please cite this article in press as: J. Zhang, J.-L. Wu, Tandem intramolecular Diels–Alder/retro-Diels–Alder cycloaddition of 2*H*-chromen-2-one as dienes with the expulsion of CO_2, Chin. Chem. Lett. (2016), http://dx.doi.org/10.1016/j.cclet.2016.03.034

Download English Version:

https://daneshyari.com/en/article/5143212

Download Persian Version:

https://daneshyari.com/article/5143212

Daneshyari.com