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1-METHYLIMIDAZOLIUM TRIFLUOROACETATE EFFICIENTLY CATALYZED THREE COMPONENT SYNTHESIS OF DIAZASPIRO-[5.5]UNDECANE-1,5,9-TRIONE DERIVATIVES UNDER SOLVENT-FREE CONDITIONS

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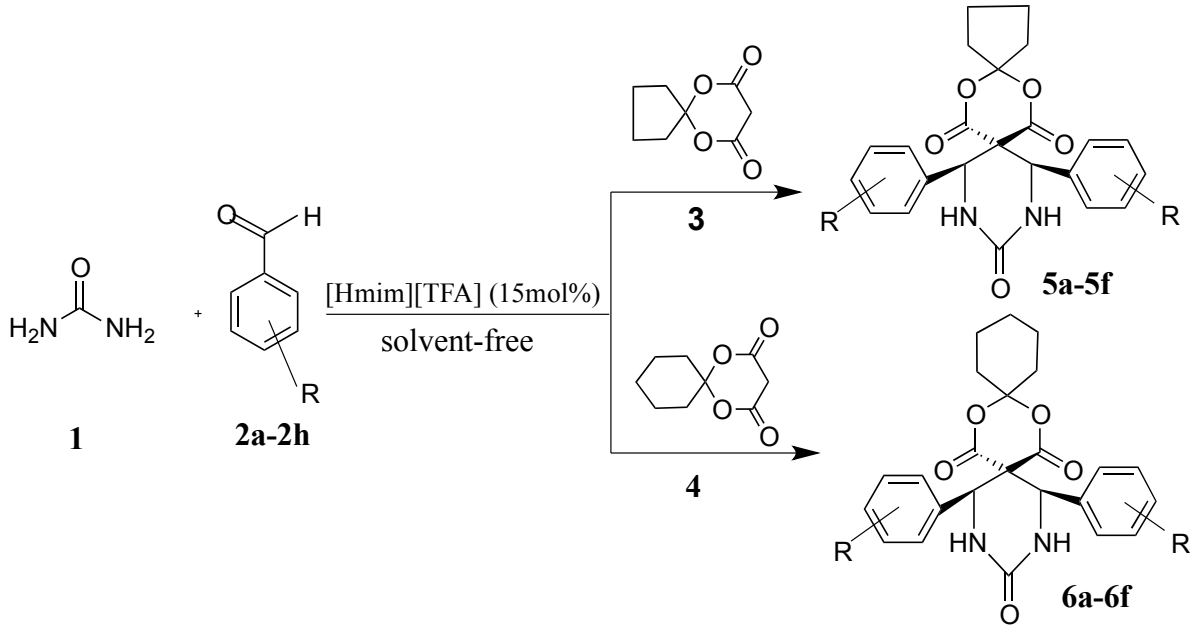
Abstract – A simple, green and efficient method for the synthesis of 2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione derivatives has been developed by the reaction of aromatic aldehydes with urea and 1,3-dioxane-4,6-dione catalyzed by 1-methylimidazolium trifluoroacetate under solvent-free conditions.

INTRODUCTION

Nitrogen-containing heterocyclic compounds have attracted considerable attention[1] owing to their potential involvement as key component for various pharmacological activities.[2,3] Among the various spiroheterobicyclic compounds, 2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione derivatives exhibit antiviral, antitumor, antihypertensive, anti-HIV, narcotic and analgesic.[4-7] In addition, there are also spiroheterocyclic units in several bioactive natural products.[8-10] Therefore the preparation of spiroheterobicyclic derivatives is of much current importance.

Diverse synthetic methodologies available for the synthesis of 2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione derivatives have been developed. One type of spiroheterocycles was generally synthesized from the reaction of aromatic aldehydes, 2,2-dimethyl-1,3-dioxane-4,6-dione and urea in the presence of either Brønsted acids (cellulosesulfuric acid,[11] $H_7P_2W_{18}VO_{62}$,[12] and *p*-toluenesulfonic acid[13]) or Lewis acids (iodine,[14] TMSCl,[15,16] $SnCl_2$,[17] $NiCl_2$,[18] NBS/AIBN,[19] and boric acid[20]), or without any catalyst,[21] or microwave irradiation.[22] However, many of these methodologies have not been entirely satisfactory, suffering drawbacks from low yields, long reaction times, high catalyst loading, environmentally unfavorable solvents, and tedious work-up, to waste production. Thus, an efficient, and enviromentally friendly method using economical catalyst is actively desirable.

In recent years, ionic liquids (ILs) have attracted increasing interests and been successfully used in a variety of catalytic reactions due to their relatively low viscosities, low vapor pressure, and high thermal and chemical stability.[23-25] Protic ionic liquids, in particular, can play dual roles, as both solvent and catalyst,[26,27] in some reactions like Biginelli and Hantzsch reactions.[28,29] Herein, we report 1-methylimidazolium trifluoroacetate ([Hmim][TFA]) as an effective and reusable catalyst for the synthesis of 3,3-butylidene-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione derivatives. Moreover, this reaction condition can also be successfully applied to 2,2-pentamethylene-1,3-dioxane-4,6-dione system and furnish the desired products in good to excellent yields (Scheme 1).



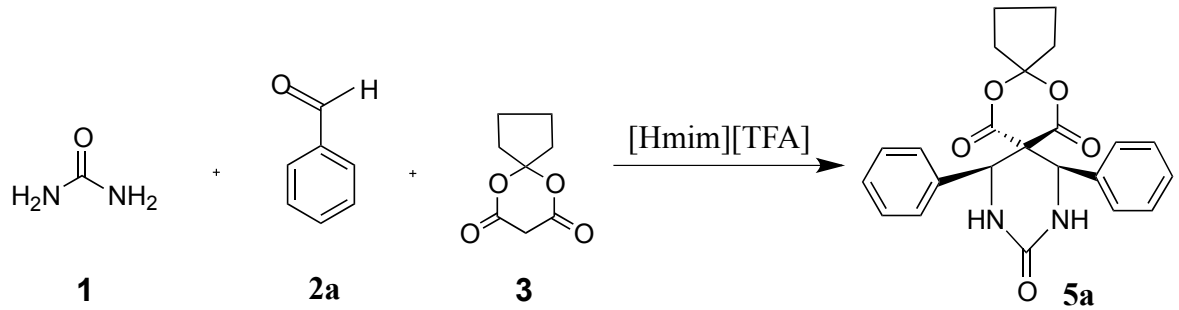
Scheme 1. Synthesis of **5** and **6**

RESULTS AND DISCUSSION

Reactions were started by examing the rection of benzaldehyde (10 mmol), urea and 2,2-butylidene-1,3-dioxane-4,6-dione as model reaction. As is shown in Table 1, without catalyst, only a trace amount of product was detected in glycerol, polyethylene glycol 400, ethylene glycol and water (Table 1, Entries 1-4). While the reaction proceeded sluggishly in DMF and DMSO, a huge improvement was obtained in toluene, EtOH and ethyl acetate, and the yield reached to 62% effectively under neat conditions (Table 1, Entries 5-10). However, under solvent-free conditions the highest yield was obtained in the persence of 15 mol% [Hmim]TFA (Table 1, Entry 12). The catalyst ([Hmim][TFA]) plays a crucial role in the success of the reaction in terms of the yields. Increasing the dosage of catalyst to 20 and 25 mol% resulted in improving reaction yields to 85% and 85% respectively at 80 ^{o}C (Table 1, Entries 12, 13). Use of just 15 mol% [Hmim][TFA] is sufficient to push the reaction forward. Higher amounts of the catalyst did not enhance yields. Thus, 15 mol% [Hmim][TFA] was chosen as a quantitative catalyst for

these reactions. The optimum reaction time and reaction temperature were also found respectively. The best result was obtained when the reaction was conducted at 80 ºC, for 4.0 h, in the presence of 15 mol% [Hmim][TFA] under neat conditions (Table 1, Entry 12). The possibility to recycle catalyst was also examined. The catalyst was reused four times with the yields of 85%, 85%, 83% and 80%, respectively (Table 1, Entry 12).

Table 1. Optimization of reaction conditions for the synthesis of **5a**[a]



1 **2a** **3** **5a**

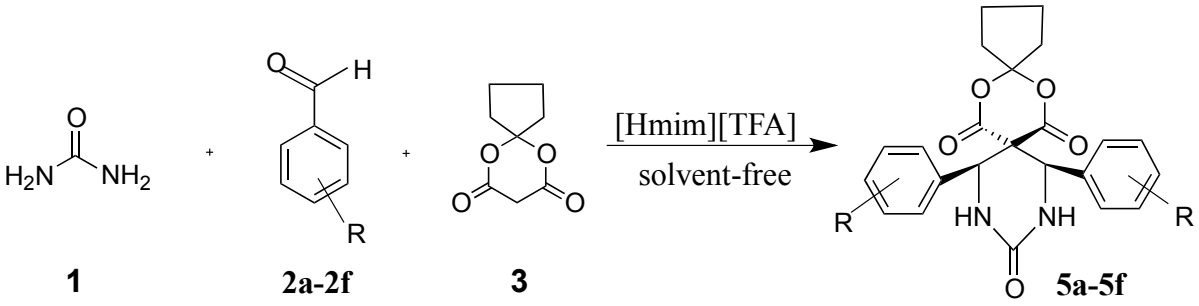
Entry	Solvent	Catalyst amounts (mol%)	Temp.(ºC)	Time(h)	yields (%)[b]
1	glycerol	none	80	8.0	<5
2	polyethylene glycol 400	none	80	8.0	<5
3	ethylene glycol	none	80	8.0	<5
4	H_2O	none	80	8.0	<5
5	DMF	none	80	8.0	21
6	DMSO	none	80	8.0	18
7	toluene	none	reflux	8.0	38
8	EtOH	none	reflux	8.0	42
9	EtOAc	none	reflux	8.0	46
10	none	none	80	5.0	62
11	none	CF_3CO_2H	80	5.0	70
12[c]	**none**	**[Hmim][TFA](15)**	**80**	**4.0**	**85**
12	none	[Hmim][TFA] (10)	80	5.0	72
13	none	[Hmim] [TFA] (20)	80	5.0	85
14	none	[Hmim] [TFA] (25)	80	4.0	85
15	none	[Hmim] [TFA] (15)	90	4.0	83
16	none	[Hmim] [TFA] (15)	70	5.0	78
17	none	[Hmim] [TFA] (15)	80	3.0	74

[a] Reaction conditions: benzaldehyde (10 mmol); urea (5 mmol); 2,2-butylidene-1,3-dioxane-4,6-dione (5 mmol) in 15 mL solvent or solvent-free. [b] Isolated yield. [c] The catalyst was reused four times with the yields of 85%, 85%, 83% and 80%, respectively.

Using the optimized reaction conditions in hand, various aromatic aldehydes were tested to investigate the generality of the reaction. The results were summarized in Table 2. The results exhibited various para-substituted benzaldehydes with electron withdrawing groups (-Cl, -NO_2,-F) and electron donating

groups (Me) have given good yields. 3-Chlorobenzaldehyde at the meta position also afforded the desired products in high yield. Conversely, it proceeded only up to Knoevenagel adducts, when electron-releasing para-substituted benzaldehydes were used (R=OMe, $N(Me)_2$).

Table 2. [Hmim][TFA]-catalyzed synthesis of **5**[a]



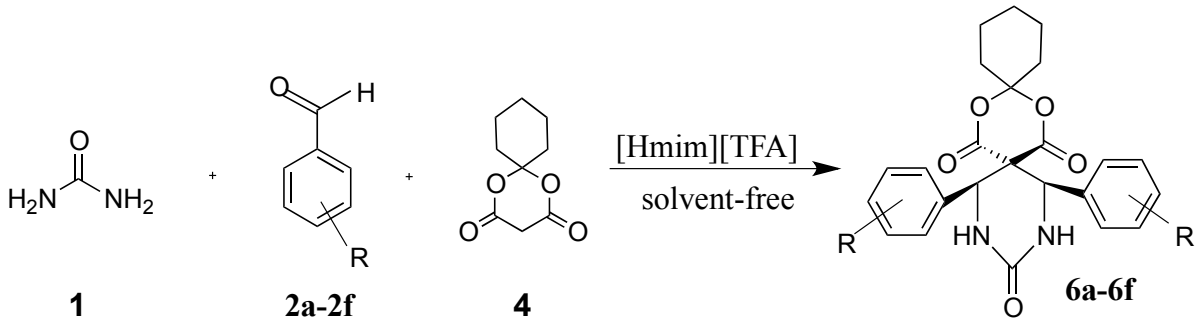
1 **2a-2f** **3** **5a-5f**

Entry	R	Time(h)	Product	yields (%)[b]
1	**2a**(R=H)	4.0	**5a**	85
2	**2b**(R=4-F)	3.0	**5b**	78
3	**2c**(R=4-Cl)	3.5	**5c**	81
4	**2d**(R=4-Me)	5.0	**5d**	71
5	**2e**(R=4-NO_2)	3.0	**5e**	75
6	**2f**(R=3-Cl)	4.0	**5f**	73

[a] Reaction conditions: aromatic aldehydes (**2a-2f**, 10 mmol); urea (5 mmol); 2,2-butylidene-1,3-dioxane-4,6-diones (5 mmol); [Hmim][TFA] (15 mol%); temperature 80 °C. [b] Isolated yield.

Inspired by the successful synthesis of **5a-5f** in the sustainable catalytic system consisting of [Hmim][TFA] without solvents, we then attempted to expand its application to the synthesis of **6a-6f.** Therefore, the desired products were obtained in 72-86% yields.

Table 3. [Hmim][TFA]-catalyzed synthesis of **6**[a]



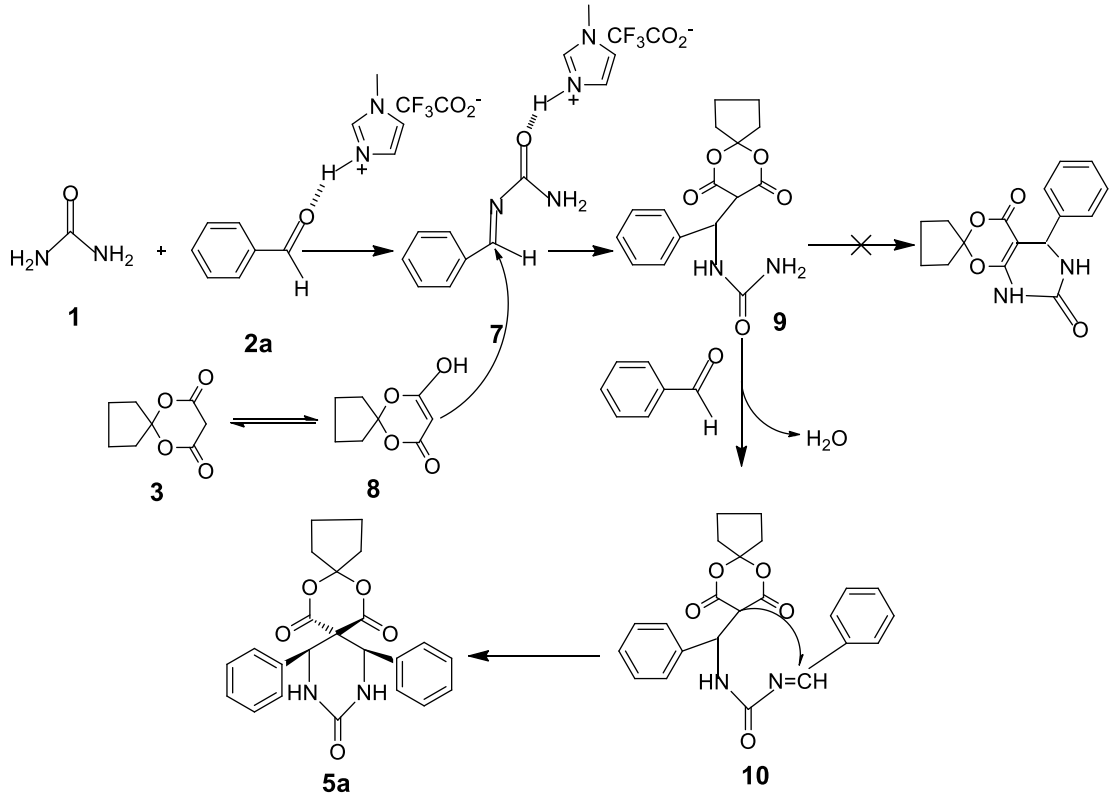
1 **2a-2f** **4** **6a-6f**

Entry	R	Time(h)	Product	yields (%)[b]
1	**2a**(R=H)	4.0	**6a**	86
2	**2b**(R=4-F)	3.5	**6b**	81
3	**2c**(R=4-Cl)	3.5	**6c**	83

4	**2d**(R=4-Me)	5.0	**6d**	75
5	**2e**(R=4-NO_2)	3.0	**6e**	76
6	**2f**(R=3-Cl)	5.0	**6f**	72

[a] Reaction conditions: aromatic aldhydes (**2a-2f**, 10 mmol); urea (5 mmol); 2,2-pentylidene-1,3-dioxane-4,6-diones (5 mmol); [Hmim][TFA] (15 mol%); temperature 80 °C. [b] Isolated yield.

From the above results and literatures,[7,21] a reasonable mechanism for the [Hmim][TFA]-catalyzed one-pot, three-component synthesis of the corresponding spiroheterocycle **5a** is depicted in Scheme 2. Firstly, benzaldehyde and urea would easily form the intermediate acylimine **7**, followed by a Michael-type addition reacting with **8** to give the open chain ureide **9**. Subsequently, intermediate **9** involved condensation of benzaldehyde to form intermediate **10** and ultimately cyclized to the spiroheterocyclic compound **5a**.



Scheme 2. Proposed mechanism for the synthesis of **5a** catalyzed by [Hmim][TFA]

EXPERIMENTAL

All chemicals were purchased from Aladdin, Aldrich and Fluka Chemical Companies and without further purification. Melting points were measured on XT-4 digital micro melting point apparatus and are uncorrected. IR spectra were taken on a Nicolet-360 FT-IR spectrometer by incorporating samples in KBr disks. ^{1}H NMR spectra were recorded on a BRUKER AVANCE 400 MHz spectrometer using DMSO as

the solvent and TMS as the internal standard. ^{13}C NMR data were collected on a BRUKER AVANCE 100 MHz instrument with DMSO as the solvent and TMS as the internal standard. The analytical MS of the compounds was performed on Agilent LC-MSD Trap VL Apparatus.

General procedure for the synthesis of 1-mthylimidazolium trifluoroacetate ([Hmim][TFA])

A solution of trifluoroacetic acid (11.4 g, 0.1 mol) in CH_2Cl_2 (50 mL) was added to *N*-methylimidazole (8.21 g, 0.1 mol)) in CH_2Cl_2 (40 mL) solution at 0-5 ºC. Then the mixture was stired at room temperature for 2 h. The solvent was evaporated at reduced pressure and the remaining product was washed with 2-methoxy-2-methylpropane (3×10 mL) to remove non-ionic residues and dried under vacuum.

1-Mthylimidazolium trifluoroacetate([Hmim][TFA]): white solid; IR (KBr, cm^{-1}): *v* 3154, 2968, 2871, 1680, 1589, 1424, 1286, 1205, 1135; ^1H NMR (DMSO-d_6) δ 3.82 (s, 3 H, CH_3), 7.62 (t, 1 H, J = 1.5 Hz), 7.66 (t, 1 H, J = 1.5 Hz), 8.95 (s, 1 H); ^{13}C NMR (DMSO-d_6) δ 35.13, 113.60, 116.1, 118.31, 120.15, 120.70, 122.95, 135.88, 158.20, 158.07, 158.33, 158.58, 158.81.

General procedure of the preparation of products 5a, 5b, 5c, 5d, 5e, and 5f

3,3-Butylidenediphenyl-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (5a): To a 50 mL tube equiped with a stirring bar were added urea **1** (300 mg, 5 mmol), 2,2-butylidene-1,3- dioxane-4,6-dione **3** (851 mg, 5 mmol), aromatic aldehyde **2a** (1060 mg, 10 mmol) and 1-mthylimidazolium trifluoroacetate ([Hmim][TFA]) 15 mol%. The vessel was then sealed with a screw cap and at 80 ºC for 4.0 h. Upon completion of the reaction, as confirmed by thin-layer chromatography (petroleum ether/EtOAc 2:1), the reaction mixture was treated with cold water. The aqueous layer consisting the acidic IL, was recovered after removal of water under reduce pressure and was reused for subsequent reactions. The crude solid product was filtered and then purified by recrystallization from EtOAc to afford the pure product **5a**; a white solid; mp 208-210 ºC. IR (KBr, cm^{-1}): *v* 3198, 3061, 1772, 1735, 1683; ^1H NMR (400 MHz, DMSO-d_6) δ 0.65-0.81 (m, 4 H), 1.29-1.33 (m, 4 H), 5.30 (s, 2 H), 7.20-7.22 (m, 4 H), 7.32 (s, 2 H, NH), 7.36-7.39 (m, 6 H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 22.21, 38.26, 58.98 (C_{spiro}), 61.98, 114.28, 128.10, 129.21, 129.75, 135.98, 155.71, 160.45, 166.30; ESI-MS *m*/*z*: 407.2 $[M+H]^+$.

3,3-Butylidene-bis(4-flurophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (5b): a white solid; mp 206-208 ºC. IR (KBr, cm^{-1}): *v* 3212, 3075, 1768, 1730, 1684; ^1H NMR (400 MHz, DMSO-d_6) δ 0.77-0.92 (m, 4 H), 1.34-1.40 (m, 4 H), 5.32 (s, 2 H), 7.24-7.28 (m, 8 H), 7.37 (s, 2 H, NH); ^{13}C NMR (100 MHz, DMSO-d_6) δ 22.37, 38.47, 59.06(C_{spiro}), 61.24, 114.44, 116.02,116.24, 130.28, 132.12, 132.16, 155.58, 160.53, 164.17, 166.23; ESI-MS *m*/*z*: 443.1 $[M+H]^+$.

3,3-Butylidene-bis(4-chlorophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione(5c): a white solid; mp 207-209 ºC. IR (KBr, cm^{-1}): *v* 3201, 3062, 1766, 1731, 1692; ^1H NMR (400 MHz, DMSO-d_6) δ 0.77-0.93 (m, 4 H), 1.36-1.39 (m, 4 H), 5.33 (s, 2 H), 7.21 (d, J = 8.0 Hz, 4 H), 7.42 (s, 2 H, NH),

7.48-7.63 (m, 4H); ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ 22.49, 38.63, 58.29 (C_{spiro}), 61.25, 114.82, 124.40, 129.81, 142.93, 148.52, 155.33, 160.09, 165.68; ESI-MS *m*/*z*: 475.1 $[M+H]^+$.

3,3-Butylidene-bis(4-methylphenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (5d): a white solid; mp 195-198 °C. IR (KBr, cm^{-1}): *v* 3226, 3081, 1773, 1735, 1686; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ 0.70-0.86 (m, 4 H), 1.22-1.31 (m, 4 H), 2.29 (s, 6 H), 5.25 (s, 2 H), 7.12 (d, *J* = 8.0 Hz, 4 H), 7.48 (d, *J* = 8.0 Hz, 4 H), 7.24 (s, 2 H, NH); ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ 22.33, 28.26, 38.47, 58.61 (C_{spiro}), 61.66, 114.78, 127.95, 129.35, 133.00, 139.01, 155.67, 156.11, 166.23; ESI-MS *m*/*z*: 435.2 $[M+H]^+$.

3,3-Butylidene-bis(4-nitrophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (5e): a light yellow solid; mp 201-202 °C. IR (KBr, cm^{-1}): *v* 3231, 3081, 1767, 1729, 1692; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ 0.72-0.90 (m, 4 H), 1.28-1.36 (m, 4 H), 5.56 (s, 2 H), 7.50 (d, *J* = 8.0 Hz, 4 H), 7.66 (s, 2 H, NH), 8.30 (d, *J* = 8.0 Hz, 4 H); ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ 22.49, 38.63, 58.29 (C_{spiro}), 61.25, 114.82, 124.40, 129.81, 142.93, 148.52, 155.33, 160.09, 165.68; ESI-MS *m*/*z*: 497.1 $[M+H]^+$.

3,3-Butylidene-bis(3-chlorophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (5f): a white solid; mp 192-194 °C. IR (KBr, cm^{-1}): *v* 3202, 3062, 1767, 1730, 1692; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ 0.80-0.90 (m, 4 H), 1.36-1.40 (m, 4 H), 5.34 (s, 2 H), 7.16 (d, *J* = 4.0 Hz, 2H), 7.23 (s, 2 H, NH), 7.42-7.49 (m, 6 H); ^{13}C NMR (100 MHz, DMSO-d$_6$)δ 22.37, 38.49, 58.70 (C_{spiro}), 61.23, 114.59, 127.02, 127.91, 129.84, 131.22, 133.86, 138.18, 155.42, 160.37, 165.95; ESI-MS *m*/*z*: 475.1 $[M+H]^+$.

General procedure of the preparation of products 6a, 6b, 6c, 6d, 6e, and 6f

3,3-Pentylidene-diphenyl-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (6a): To a 50 mL tube equiped with a stirring bar were added urea **1** (300 mg, 5 mmol), 2,2-pentylidene-1,3-dioxane-4,6-dione **4** (921 mg, 5 mmol), aromatic aldehyde **2a** (1060 mg, 10 mmol) and 1-mthylimidazolium trifluoroacetate ([Hmim][TFA]) 15 mol%. The vessel was then sealed with a screw cap and at 80 °C for 4.0 h. Upon completion of the reaction, as confirmed by thin-layer chromatography (petroleum ether/EtOAc 2:1), the reaction mixture was treated with cold water. The aqueous layer consisting the acidic IL, was recovered after removal of water under reduce pressure and was reused for subsequent reactions. The crude solid product was filtered and then purified by recrystallization from EtOAc to afford the pure product **6a**; a white solid; mp 218-220 °C. IR (KBr, cm^{-1}): *v* 3201, 3060, 1771, 1734, 1685; ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ 0.43 (s, 4 H), 1.07 (s, 6 H), 5.30 (s, 2 H), 7.21-7.23 (m, 4 H), 7.31 (s, 2 H, NH), 7.35-7.40 (m, 6 H); ^{13}C NMR (100 MHz, DMSO-*d*$_6$) δ 21.88, 23.28, 36.99, 59.06 (C_{spiro}), 62.01, 105.99, 128.16, 129.13, 129.72, 136.04, 155.72, 160.35, 166.17; ESI-MS *m*/*z*: 421.2 $[M+H]^+$.

3,3-Pentylidene-bis(4-flurophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (6b): a white

solid; mp 216-217 ºC; IR (KBr, cm^{-1}): ν 3207, 3074, 1768, 1734, 1683 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) δ 0.54 (s, 4 H), 1.12 (s, 6 H), 5.32 (s, 2 H), 7.22-7.28 (m, 8 H), 7.36 (s, 2 H, NH); ^{13}C NMR (100 MHz, DMSO-d_6) δ 21.87, 23.22, 37.16, 59.17 (C_{spiro}), 61.27, 106.11, 115.94, 116.16 130.34, 130.42, 132.20, 132.23, 155.55, 160.45, 161.75, 164.20, 166.12; ESI-MS *m*/*z*: 457.1 $[M+H]^+$.

3,3-Pentylidene-bis(4-chlorophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (6c): a white solid; mp 210-212 ºC. IR (KBr, cm^{-1}): *v* 3208, 3086, 1771, 1729, 1683; ^1H NMR (400 MHz, DMSO-d_6) δ 0.54 (s, 4 H), 1.12 (s, 6 H), 5.33 (s, 2 H), 7.22 (d, *J* = 6.0 Hz, 4 H), 7.41 (s, 2 H, NH), 7.49 (d, *J* = 4.0 Hz, 4 H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 21.89, 23.17, 37.15, 58.91 (C_{spiro}), 61.34, 106.17, 129.20, 130.09, 131.63, 134.89, 155.52, 160.36, 166.00; ESI-MS *m*/*z*: 489.1 $[M+H]^+$.

3,3-Pentylidene-bis(4-methylphenyl)-2,4-dioxa-8,10-diazaspiro[5.5]undecane-1,5,9-trione (6d): a white solid; mp 202-205 ºC. IR (KBr, cm^{-1}): *v* 3226, 3081, 1773, 1735, 1686; ^1H NMR (400 MHz, DMSO-d_6) δ 0.46 (s, 4 H), 1.08 (s, 6 H), 2.26 (s, 6 H), 5.23 (s, 2 H), 7.08 (d, *J* = 8.0 Hz, 4 H), 7.18 (d, *J* = 8.0 Hz, 4 H), 7.22 (s, 2 H, NH); ^{13}C NMR (100 MHz, DMSO-d_6) δ 21.11, 21.94, 23.31, 37.03, 59.15 (C_{spiro}), 61.87, 106.11, 128.03, 129.55, 133.07, 139.19, 155.78, 160.55, 166.35; ESI-MS *m*/*z*: 449.2 $[M+H]^+$.

3,3-Pentylidene-bis(4-nitrophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]-undecane-1,5,9-trione(6e): a light yellow solid; mp 211-212 ºC. IR (KBr, cm^{-1}): *v* 3229, 3078, 1769, 1727, 1691; ^1H NMR (400 MHz, DMSO-d_6) δ 0.49 (s, 4 H), 1.07 (s, 6 H), 5.57 (s, 2 H), 7.50 (d, *J* = 8.0 Hz, 4 H), 7.67 (s, 2 H, NH), 8.30 (d, *J* = 8.0 Hz, 4 H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 21.80, 23.07, 37.24, 58.48 (C_{spiro}), 61.37, 106.54, 124.32, 129.91, 143.01, 148.55, 155.34, 160.14, 165.62; ESI-MS *m*/*z*: 511.1 $[M+H]^+$.

3,3-Pentylidene-bis(3-chlorophenyl)-2,4-dioxa-8,10-diazaspiro[5.5]-undecane-1,5,9-trione(6f): a white solid; mp 196-198 ºC. IR (KBr, cm^{-1}): *v* 3201, 3064, 1771, 1734, 1689; ^1H NMR (400 MHz, DMSO-d_6) δ 0.55 (s, 4 H), 1.13 (s, 6 H), 5.34 (s, 2 H), 7.16 (d, *J* = 4.0 Hz, 2 H), 7.23 (s, 2H, NH), 7.41-7.49 (m, 6 H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 21.81, 23.16, 37.11, 58.84 (C_{spiro}), 61.28, 106.32, 127.11, 127.97, 129.80, 131.13, 133.90, 138.24, 155.42, 160.32, 165.86; ESI-MS *m*/*z*: 489.1 $[M+H]^+$.

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