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Short Communication

A one-pot three-component synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones (DAPMs) using atomized sodium in THF under sonic condition

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ABSTRACT

A simple and an efficient procedure for the synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones using atomized sodium/THF via a one-pot three-component Biginelli-like cyclocondensation of an aldehyde, a methyl ketone and urea under ultrasonic condition is developed. The method is mild and inexpensive; yields are high and the reactions go to completion within 10–15 min.

1. Introduction

In recent years synthesis of biological active compounds is in great demand. Pyrimidine nucleus is found in many natural bioactive products possessing multiple biological and medical properties [1]. Some of these compounds serve as antihypertensive, antibacterial, and anti-inflammatory agents [2]. The batzelladine alkaloids containing 3,4-DHPM isolated from marine sources inhibits the binding of HIV envelope protein gp-120 to human CD_4 cells [3]. Such properties make these pyrimidones highly important. There are only a few methods reported for the synthesis of DHPMs, wherein a one-pot cyclocondensation of methyl ketones with aldehydes and urea instead of 1,3-diketones is reported [4]. Heravi et al. used TMSCl and sulfamic acid as catalysts for the synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones (DAPMs) [5], Khosropour et al. used $Bi(TFA)_3$ immobilized on [nbpy]$FeCl_4$ [6], and Cai-hui used con. HCl in ionic liquid [BMIM][BF_4] to promote this reaction [7]. Other reported methods for the synthesis of DAPMs are from urea and 1,3-diphenylpropenone or 1,3-diphenylpropynone or 1,3-diphenylpropane-1,3-dione in the presence of NaOEt/Et_3N or cyanouric trichloride/CF_3SO_3Zn as catalysts [8–11]. The above reported methods suffer from drawbacks such as use of stoichiometric amounts of catalysts, expensive reagents, prolonged reaction time, and varying yields of the products.

In recent days multi-component reactions are preferred for the syntheses of a variety of organic molecules because of ease of execution, non-isolation of intermediates, short reaction times, and quantitative yields of the products [12]. In the last decade, sonochemistry has gained tremendous popularity in the field of organic syntheses for the reason that, it is more convenient, high yielding, reactions go to completion in short durations when compared to the traditional methods. Also, the number of steps involved are less and cruder reagents can be used under sonic conditions [13–21]. Richards and Loomis were the first to report the use of ultrasound in organic syntheses in the year 1927 [22]. In continuation of the work from our laboratory to develop new methods for the synthesis of various biologically important molecules such as *N*,*N*-disubstituted ureas/thioureas [23], azines [24], β-acetamido-β-arylpropiophenones [25] and aryl-14*H*-dibenzo[*a*,*j*]xanthenes [26], we are reporting an effective protocol for the synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones using atomized sodium in THF *via* a one-pot three-component cyclocondensation of methyl ketones and urea with various substituted aryl aldehydes under ultrasonic condition. This one-pot route is mild, energy efficient, and inexpensive. The yields are high (up to 90%) and the reactions go to completion within 10–15 min as shown in Scheme 1.

2. Results and discussion

We started our investigations with various catalysts, in various solvents under different reaction conditions in order to develop a new method for the synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones

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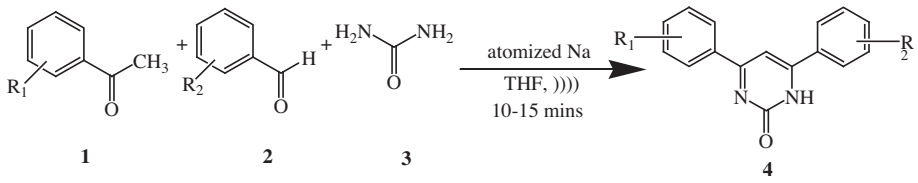

Scheme 1. Cyclocondensation of benzaldehyde, acetophenone, and urea using atomized sodium in THF under sonic condition.

via a one-pot three-component Biginelli-type cyclocondensation reaction. To standardize the conditions all the reactions were carried out with 4-(*N*,*N*-dimethylamino)benzaldehyde, acetophenone and urea as model substrates. Initially we selected a series of metals and conducted the above reaction to get high yield of the desired product with atomized sodium in THF as a solvent under sonic condition within 10–20 min. It was found that, at silent condition in-order to get the same yield we require to stir for 360–480 min as shown in Table 1 (entry b). It was also found that, with commercial LR grade sodium the yield is only 70%, which can be attributed to the presence of hydroxide/oxide layer on the surface of the metal, which requires cleaning first and then participation in the reaction. As the surface area of the atomized sodium is large the reaction with atomized sodium is fast and gives high yield of the product under sonic condition. Hence, pre-treatment of the commercial sodium to get atomized sodium is essential to get the high yield of the product in short reaction duration. However, the pre-treatment of commercial sodium with ultrasound for about 10 min followed by the reaction with the substrates did not improve the yield of the reaction, which may be due to the non-atomization of the sodium metal under sonic condition.

In order to verify whether THF is a suitable solvent or not for the above reaction, the reaction was carried out in various solvents and under solvent-free condition. The results of these studies are presented in Table 2; and it was found that, atomized sodium in THF is ideal in terms of yield and the time of the completion of the reaction. As I_{max} (maximum cavitation intensity) and $T_{I_{max}}$ (the temperature at which I_{max} is reached) of any solvent has a profound effect in sonochemical reactivity, I_{max} of THF may be responsible for the increase in the reaction rate when compared to the other solvents [27]. Hence, the rate of the reaction under sonic condition gets accelerated substantially in THF when compared to other solvents.

For optimizing the amount of atomized sodium, we worked with different amounts of atomized sodium required for the reaction and the results of this study are given in Table 3. From this Table it is clear that, equivalent amounts (2 mg atom) of the metal is essential for the present reaction.

In order to find the minimum amount of THF required to get maximum yield in short duration, the reaction was carried out in different volumes of THF and it was found that, the minimum amount of THF required to get the maximum yield of the product is 2 mL. The results of this study are presented in the Table 4. From the data provided in Tables 1–4 it is also clear that, synthesis of 4,6-diarylpyrimidin-2(1*H*)-one (4f) using atomized sodium/THF *via* a three-component one-pot Biginelli-type cyclocondensation under the influence of ultrasound at 35 kHz is efficient and gives high yield of the product in short duration.

In order to find the generality of the use of atomized sodium in THF for the cyclocondensation of an aldehyde with acetophenone and urea under sonic condition, different substituted aromatic aldehydes and methyl ketones were selected and the reactions were carried out in a sonic bath working at 35 kHz (constant frequency, 80 W) maintained at 25 °C by circulating water. The results of this study are presented in Table 5. It can be seen that, the reaction is not influenced by the presence of neither electron donating nor electron withdrawing substituents on the aromatic ring of aldehydes and ketones at different positions.

Table 1
A comparative study on the synthesis of 4f with different metals in THF.[a]

Entry	Metal[b]	Time (min)		Yield (%)[c,d]
		Silent	US	
a	Sodium	360–480	30–40	70
b	Atomized sodium	360–480	10–20	90
c	Aluminium	360–480	12–20	10
d	Zinc	360–480	12–20	15
e	Iron	360–480	12–20	20
f	Copper	360–480	12–20	05

[a] 5 mL.
[b] 2 mg atom.
[c] Isolated yields.
[d] Compared with authentic sample on TLC.

Table 2
Effect of nature of solvent on the synthesis of 4f in the presence of sodium.[a]

Entry	Solvent[b]	Time (min)			Yield (%)[c,d]
		Silent 25 °C	MW	US	
a	No solvent	360–480	60–80	60–80	5
b	Acetone	360–480	60–80	50–60	10
c	Acetonitrile	360–480	60–80	50–60	5
d	Chloroform	360–480	60–80	50–60	25
e	DCE	360–480	60–80	50–60	25
f	DCM	360–480	60–80	50–60	25
g	Ethanol	360–480	60–80	60–80	25
h	Ether	360–480	60–80	60–70	10
i	Hexane	360–480	60–80	30–40	20
j	Methanol	360–480	60–80	60–80	20
k	THF	360–480	60–80	10–20	90
l	Xylene	360–480	60–80	50–60	5

[a] 4-(*N*,*N*-dimethylamino)benzaldehyde (2.0 mmol), acetophenone (2.0 mmol), urea (3.0 mmol) and atomized sodium (2 mg atom) under sonic condition (35 kHz).
[b] 5 mL.
[c] Isolated yields.
[d] Compared with authentic sample on TLC.

Table 3
Optimisation of the amount of sodium required for the synthesis of 4f.

Entry	Amount of atomized sodium (mg atom)	Time (min) US	Yield (%)[a,b]
a	0.0	50	ND
b	0.5	30	70
c	1.0	20	75
d	1.5	20	80[c]
e	2.0	10	90
f	2.5	10	90
g	3.0	10	90

[a] Isolated yields; after silica gel column chromatography.
[b] Compared with authentic sample on TLC; ND: not detected.
[c] 4-(*N*,*N*-dimethylamino)benzaldehyde (2.0 mmol), acetophenone (2.0 mmol), and urea (3.0 mmol) in THF (5.0 mL) under sonic condition (35 kHz).

Table 4
Amount of THF required for the synthesis of 4f.

Entry	THF (mL)	Time (min) US	Yield (%)[a,b,c]
a	0.5	50	80
b	1.0	50	80
c	1.5	50	85
d	2	50	90
e	2	40	90
f	2	35	90
g	2	30	90
h	2	25	90
i	2	20	90
j	2	15	90
k	2	10	90
l	2	08	90
m	2.5	10	90
n	2.5	15	90
o	5	10	90
p	10	10	86

[a] Isolated yields.
[b] Compared with authentic sample on TLC.
[c] 4-(*N*,*N*-dimethylamino)benzaldehyde (2.0 mmol), acetophenone (2.0 mmol), atomized sodium (2 mg atom) and urea (3.0 mmol) under sonic condition (35 kHz).

2.1. Effect of ultrasound on the reaction

Sonochemistry is a unique and distinctive chemistry, in which the physical properties of the medium will have a decisive effect on the chemical reactivity. The reactions carried out under the influence of ultrasound are considered to be clean and the method is green as it involves use of an energy efficient technique [30,31]. The present reaction is an example of a three-phase system: the liquid phase (reagents in solvents), solid phase (atomized sodium and solid substrates), and the gas phase (dissolved gases in the liquids and gases on the inner-surface of the vessel) [31]. When sound waves pass through liquid medium, they induce vibrational motion to the medium, the solvent molecules then compress, stretch and oscillate around their mean position due to time-varying pressure, at a point when the intensity of the sonic waves is higher enough to break the intermolecular forces existing between the solvent molecules it breaks down and a cavity is formed. The process of generating cavitational bubble is called acoustic cavitation [30,32]; and the bubble collapse then becomes non-spherical near the solid surface *i.e.,* near the surface of the solid atomized sodium and the surface of the vessel, which drags the liquid high-speed jets near the surface creating shockwaves which can activate the surface of the metal. The formation of the micro-jets and shockwaves create the localized erosion responsible for most of the sonochemical effects in the present heterogeneous reaction. The phenomenon of acoustic cavitation attributes to the accomplishment of the organic reactions under sonic condition [32]. The primary chemical reactions are due to the transient state of immense temperature, pressure and extraordinary heating rates which are generated due the cavitation bubble collapse [21]. The other effects are considered to be physical rather than chemical and judged to be '*false*' sonochemical effects [33].

3. Experimental

3.1. Materials and methods

All the chemicals used were commercially available reagents. All the solvents were distilled before use. THF was distilled and dried over sodium. All the reactions were studied using SIDILU Indian make sonic bath working at 35 kHz (constant frequency, 80 W) maintained at 25 °C (by circulating water). The completion of the reaction was monitored on TLC (eluent: 8–10% ethyl acetate in light petrol), by comparison with the authentic samples. Melting points of the obtained products were determined using a Büchi apparatus. Nuclear magnetic resonance spectra were obtained on a 400 MHz Bruker AMX spectrometer in DMSO-d_6 using TMS as a standard. GC–Mass spectra were obtained using a Shimadzu GC–MS QP 5050A instrument equipped with a 30 m length and 0.32 mm dia BP-5 column with the column temperature 80–15–250 °C. Infrared spectra were recorded using Shimadzu FT-IR-8400s Spectrophotometer as KBr pellets for solids.

Typical experimental procedure for the synthesis of 4f: A mixture 4-(*N*,*N*-dimethylamino)benzaldehyde (0.274 g, 2.0 mmol), acetophenone (0.24 g, 2.0 mmol), urea (0.18 g, 3.0 mmol), atomized sodium (2.0 mg atom), THF (2 mL) were sonicated in a sonic bath working at 35 kHz (constant frequency, 80 W) maintained at 25 °C (by circulating water) for 10 min. At the end of the reaction, liquefied reaction mixture suddenly becomes solid, to which water was added and shaken for few minutes. This was filtered through a sintered funnel to afford the crude product, which was further purified by recrystallization using absolute ethanol.

4. Spectral data

4.1. 4,6-Diphenyl-pyrimidin-2(1H)-one (4a)

m.p. 235 °C; IR (KBr): ν 3358, 3159, 2960, 1612, 1502 cm^{-1}; ^1H NMR (DMSO, 300 MHz): δ 7.1–7.15 (d, 2H, J = 15 Hz, HAr and CH), 7.3–7.6 (m, 7H, J = 90 Hz, HAr), 7.88–7.92 (d, 2H, J = 12 Hz, HAr), 7.99 (s, 1H, NH) ppm; MS (70 eV), *m/z*: 248 [M^+]

Table 5
Three-component cyclocondensation of aldehyde, urea with aromatic ketones by atomized sodium in dry THF under sonic condition (35 kHz constant frequency at 25 °C).

Entry	Ketone	Aldehyde	Product	Time (min)	Yield (%)[a,b]	Melting point (°C)	
	R_1 (**1**)	R_2 (**2**)	(**4**)	US		Found	Reported[c,d]
a	H	H	**4a**	10	90	235	233–240[c]
b	H	4-CH_3	**4b**	14	88	288	287–290[c]
c	H	4-Cl	**4c**	13	86	255–257	258–260[c]
d	H	4-OH	**4d**	13	86	258–261	260–263[c]
e	H	4-OCH_3	**4e**	12	88	258	258–260[c]
f	H	4-*N*,*N*-$(CH_3)_2$	**4f**	10	90	292	290–293[d]
g	4-Cl	H	**4g**	10	88	250–253	251–254[c]
h	4-NO_2	4-Cl	**4h**	11	88	310	308–310[c]
i	4-OCH_3	4-Cl	**4i**	11	87	311	312–314[c]
j	4-OCH_3	4-*N*,*N*-$(CH_3)_2$	**4j**	10	90	285–289	280–290[c]

[a] Isolated yields.
[b] All the products are known and were characterized by comparison of their spectroscopic and physical data with the authentic samples.
[c] References [28,29].
[d] References [28,29].

4.2. 4-(4′-Toluyl)-6-phenyl-pyrimidin-2(1H)-one (4b)

m.p. 288 °C; IR (KBr): ν 3449, 3099, 2923, 1620, 1513, 1460 cm^{-1}; ^1H NMR (DMSO, 300 MHz): δ 2.39 (s, 3H, CH_3), 7.36 (d, 2H, J = 7.5 Hz, HAr), 7.53–7.57 (m, 4H, H-5 and HAr), 8.07 (d, 2H, J = 7.6 Hz, HAr), 8.15 (d, 2H, J = 5.75 Hz, HAr) ppm; MS (70 eV), *m/z*: 262 [M^+].

4.3. 4-(4′-Chlorophenyl)-6-phenyl-pyrimidin-2(1H)-one (4c)

m.p. 255–257 °C; IR (KBr): ν 3429, 3230, 3083, 1605, 1549, 1509 cm^{-1}; ^1H NMR (DMSO, 500 MHz): δ 7.59 (d, 2H, J = 7.33 Hz, HAr), 7.60–7.69 (m, 4H, HAr and H-5), 8.16 (d, 2H, J = 7.45 Hz, HAr), 8.22 (d, 2H, J = 8.32 Hz, HAr) ppm; MS (70 eV), *m/z*: 282 [M^+]

4.4. 4-(4′-Hydroxyphenyl)-6-phenyl-pyrimidin-2(1H)-one (4d)

m.p. 258–261 °C; IR (KBr): ν 3389, 2922, 1613, 1515, 1450 cm^{-1}; ^1H NMR (DMSO, 500 MHz): δ 6.96–7.04 (m, 3H, NH and H-4), 7.52–7.71 (m, 5H, HAr and H-5), 8.05–8.16 (m, 5H, HAr), 11 (s, 1H, OH) ppm; MS (70 eV), *m/z*: 264 [M^+]

4.5. 4-(4′-Methoxyphenyl)-6-phenyl-pyrimidin-2(1H)-one (4e)

m.p. 258 °C; IR (KBr): ν 3440, 3095, 2930, 1608, 1514, 1458 cm^{-1}; ^1H NMR (DMSO, 400 MHz): δ 3.9 (s, 3H, CH_3), 7.2–8.2 (m, 10H, H-5 and HAr), 11.95 (S, 1*H*, NH) ppm; MS (70 eV), *m/z*: 278 [M^+]

4.6. 4-(4′-N,N-Dimethylphenyl)-6-phenyl-pyrimidin-2(1H)-one (4f)

m.p. 292 °C; IR (KBr): ν 3448, 3095, 2925, 1619, 1509 cm^{-1}; ^1H NMR (DMSO, 300 MHz): δ 3.04 (d, 6H, J = 6.63 Hz, CH_3), 6.79–6.81 (d, 2H, J = 6 Hz, HAr), 7.36 (s, 1H, CH), 7.52–7.58 (m, 3H, J = 18 Hz, HAr), 8.06–8.12 (m, 4H, J = 18 Hz, HAr), 11.89 (br s, 1H, NH) ppm; MS (70 eV), *m/z*: 291 [M^+]

4.7. 4-(4′-Chlorophenyl)-6-phenyl-pyrimidin-2(1H)-one (4g)

m.p. 250–253 °C; IR (KBr): ν 3320, 1608, 1532, 1488 cm^{-1}; ^1H NMR (DMSO, 300 MHz): δ 7.56 (d, 3H, J = 5.90 Hz, HAr), 7.82 (s, 1H, H-5), 8.17 (d, 2H, J = 6.62 Hz), 8.35 (d, 2H, J = 8.25 Hz, HAr), 8.46 (d, 2H, J = 8.16 Hz, HAr) ppm; MS (70 eV), *m/z*: 282 [M^+]

4.8. 4-(4′-Chlorophenyl)-6-(4″-nitrophenyl)-pyrimidin-2(1H)-one (4h)

m.p. 310 °C; IR (KBr): ν 3412, 3192, 2921, 1612, 1548, 1515, 1456, 1348 cm^{-1}; ^{1}HNMR (DMSO, 300 MHz): δ 6.90–7.83 (m, 9H, H-5 and HAr), 9.98 (s, 1H, NH) ppm; MS (70 eV), *m/z*: 327 [M^+]

4.9. 4-(4′-Chlorophenyl)-6-(4″-methoxyphenyl)–pyrimidin-2(1H)-one (4i)

m.p. 311 °C; IR (KBr) ν = 3446, 3105, 2926, 1614, 1513 cm^{-1}; ^1H NMR (DMSO, 500 MHz): δ 3.86 (s, 3H, OCH_3), 7.09 (d, 2H, J = 8.85 Hz, HAr), 7.57 (s, 1H, H-5), 7.61 (d, 2H, J = 8.55 Hz, HAr), 8.16 (d, 2H, J = 8.16 Hz, HAr), 8.21 (d, 2H, J = 8.15 Hz, HAr), 11.96 (br s, 1H, NH) ppm;
MS (70 eV), *m/z*: 312 [M^+]

4.10. 4-(4′-N,N-Dimethylphenyl)-6-(4″-methoxyphenyl)-pyrimidin-2(1H)-one (4j)

m.p. 285–289 °C; IR (KBr): ν 3454, 3113, 2932, 1627, 1528 cm^{-1}; ^1H NMR (DMSO, 500 MHz): δ 1.23 (d, 6H, J = 6.63 Hz, CH_3), 3.88 (s, 3H, OCH_3), 7.101 (d, 2H, J = 8.86 Hz, HAr), 7.62 (s, 1*H*, H-5), 7.63 (d, 2H, J = 8.56 Hz, HAr), 8.19 (d, 2H, J = 8.17 Hz, HAr), 8.23 (d, 2H, J = 8.16 Hz, HAr), 11.97 (br s, 1*H*, NH) ppm; MS (70 eV), *m/z*: 321 [M^+]

5. Conclusions

We have developed an efficient synthesis of 4,6-diarylpyrimidin-2(1*H*)-ones by a one-pot three-component cyclocondensation reaction between an aldehyde, a methyl ketone and urea using atomized sodium in THF under sonic condition. This new protocol has advantages such as: (i) the use of cheap, easy to handle and commercially available sodium metal; (ii) short reaction times (10–15 min); and (iii) high yields (91–96%).

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