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# NOVEL SYNTHESIS OF [1,2,4]-TRIAZOLO-QUINAZOLINONE AND PYRIMIDINE DERIVATIVES MEDIATED BY CERIC AMMONIUM NITRATE (CAN)

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**Abstract:** A highly efficient, clean and simple protocol has been established for the synthesis of [1,2,4]-triazolo-quinazolinone and [1,2,4]-triazolo-pyrimidine derivatives in the presence of CAN. This CAN was effective for the one-pot multi-component reaction of aromatic aldehyde, dimedone or ethyl acetoacetate and 3-amino-1,2,4-triazole in acetonitrile under mild reaction conditions. The present work shows attractive features, such as the short reaction time, excellent yield, mild reaction condition, easily isolated the product and no need of chromatographic separation.

***Keywords*:** Triazoloquinazolinone; 3-Amino-1,2,4-Triazole; aromatic aldehyde; CAN; Mild reaction condition

## Introduction

Nitrogen containing heterocyclic compound are important parts that often exist in biologically active natural products and medicinal interest of synthetic compounds.[1,2] Among them 1,2,4-triazoloquinazolinone

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derivatives are known for diverse biological and pharmaceutical activities as derivatives are known for diverse biological and pharmaceutical activities as anti-HIV,[3] antihistaminic,[4–6] analgesic,[7] anti-inflammatory,[8] anticancer,[9] anti-bacteria,[10-12] anti-malarial.[13] Alternatively, one pot synthetic strategies of multi-component reaction (MCRs) has been facilitated to the growth of organic synthesis. Because of their advantages over the multistep synthesis such as they generate less waste, minimize isolation of intermediates, save times and minimize cost.[14] In addition, MCRs are eco-friendly, highly a tom economic and they avoid protection–deprotection steps with minimum synthetic effort and time.[15-16]

Due to their wide range of synthetic, industrial and pharmacological application, the synthesis of 1,2,4-triazoloquinazolinone derivatives has become a focus of intense research in recent years. Several synthetic methodologies have been developed for the synthesis of 1,2,4-triazoloquinazolinone derivatives. Among these methods are the condensation of dimedone, various aldehydes with 3-amino-1,2,4-triazole in the presence of Nafion-H®,[17] molecular iodine,[18] Amberlyst-15® in PEG,[19] DMF (microwave assisted),[20] $H_6P_2W_{18}O_{62}$ .$18H_2O$,[21] acetic acid,[22] 1-*n*butyl-3-methylimidazolium tetrafluoroborate ([Bmim] $BF_4$),[23] p-toluenesulfonic acid monohydrate,[24] sulfonic acid functionalized nanoporous silica (SBA-Pr-$SO_3H$),[25] anthranilic acid,[26] Sulfamic acid.[27]

However, many of these methods suffer from one or more of the limitations such as requirement of strong acidic conditions, longer reaction times, low yields, tedious work-up procedures, excess amount of catalyst, and the use of toxic reagents, catalysts or solvents. Therefore, there is a strong demand for a highly efficient and environmentally benign method for the synthesis of these heterocycles.

In recent literatures, CAN is convenient and widely used reagent for the organic transformation due to the many advantages such as excellent solubility in water, eco-friendly nature, high reactivity, cost-effectiveness, low toxicity and easy work up the procedure. Although, CAN is able to catalyze not only based on its electron transfer capacity, but also with its Lewis acidic property for various organic transformation.[28] CAN have used as an important reagent for the formation of C-C and carbon-heteroatom bonds.[29,30]

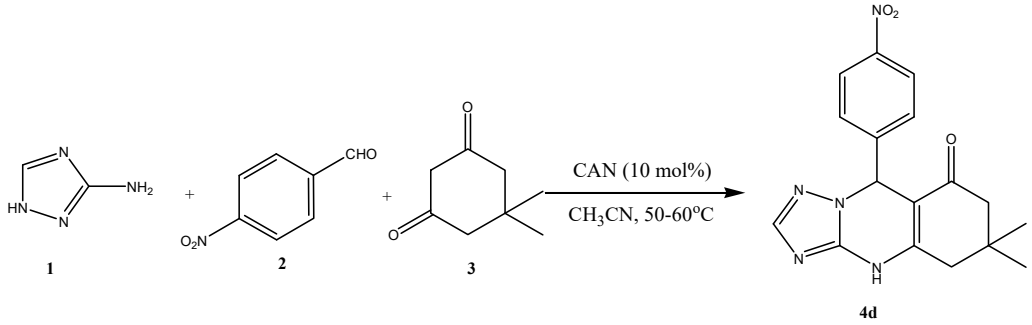
In continuation of our ongoing research work to develop novel methodologies in synthetic chemistry,[31-34] Herein, we report CAN as an efficient, low cost and environmentally benign protocol for the synthesis of 1,2,4-triazoloquinazolinone derivatives under mild reaction conditions at 50–60 $^{o}$C.

**Results and Discussion**

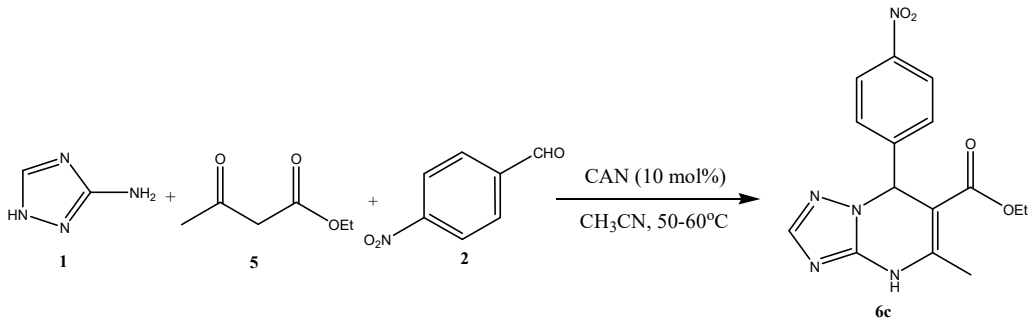
To explore the use of CAN as a catalyst, for the reaction of dimedone or ethyl acetoacetate, 4-nitrobenzaldehydes with 3-amino-1,2,4-triazole for the preparation of 1,2,4-triazoloquinazolinone and 1,2,4-triazolo[1,5-a]pyrimidine-6-carboxylate derivatives compound **4d** and **6c** was considered as a standard model reaction (Scheme 1 and Scheme 2). Model reaction carried out in the absence of catalyst did not lead to formation of preferred product. It means the initiation of reaction was must required the involvement of catalyst. Initially, we find out the exact requirement of amount of catalyst for this transformation. During this study, we consider the model reaction and examine requirement of catalyst concentration using different concentrations of CAN (Table 1). After this study, we have observed that 10 mol% CAN show to be an efficient catalyst

to bring out the reaction smoothly. With these optimized reaction conditions, effect of different solvents such as water, methanol, ethanol, aqueous ethanol and acetonitrile was investigated (Table 1). Among the tested solvents, acetonitrile was found to be better over the other tested solvents in terms of both yield of the product and reaction time (Table 1 Entry 8) for this transformation.

As a results, further set of experiments, in order to make the generality of the reaction, various aromatic aldehydes having both electron-donating as well as electron-withdrawing substituent's were transformed into 1,2,4-triazoloquinazolinone derivatives in high to excellent yields. The entire results are summarized in (Table 2 and Table 3).




**Scheme 1**

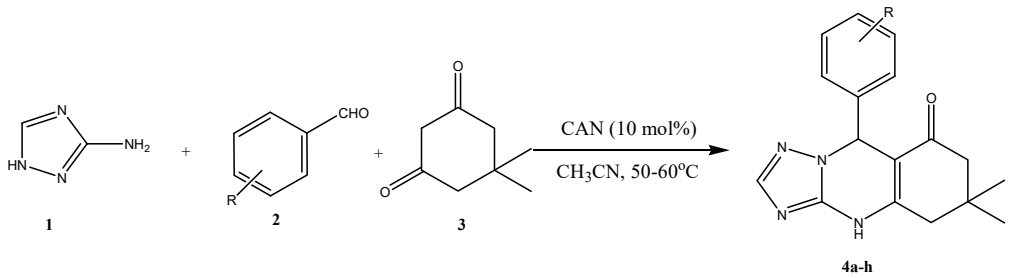



**Scheme 2**

**Table 1.** Optimization of solvent and catalyst effect.

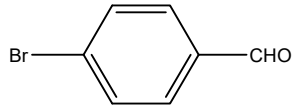
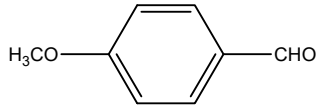
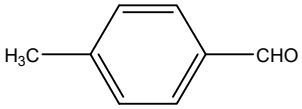
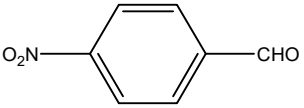
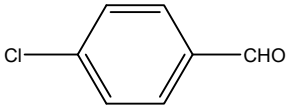
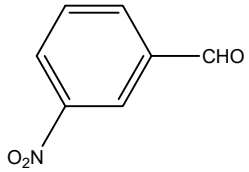
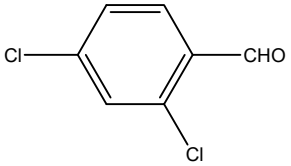
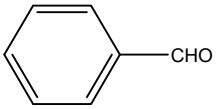
| Entry | Solvent | Catalyst (mol %) | Time (min.) | Yield[b] (%) |
|---|---|---|---|---|
| 1 | - | - | 60 | No reaction |
| 2 | $H_2O$ | 10 | 50 | 45 |
| 3 | $CH_3OH$ | 10 | 50 | 72 |
| 4 | EtOH | 10 | 50 | 80 |
| 5 | EtOH:$H_2O$ | 10 | 50 | 68 |
| 6 | $CH_3CN$ | 5 | 60 | 75 |
| **7** | $CH_3CN$ | 7 | 40 | 82 |
| 8 | **$CH_3CN$** | **10** | **17** | **96** |
| 9 | $CH_3CN$ | 15 | 17 | 96 |

[a]**Reaction conditions:** Dimedone (1 mmol), 4-Nitrobenzaldehyde (1 mmol), 3-amino-1,2,4 triazole (1 mmol), CAN (10 mol%) in acetonitrile (5 mL) at 50-60 °C. [b]Isolated yield.

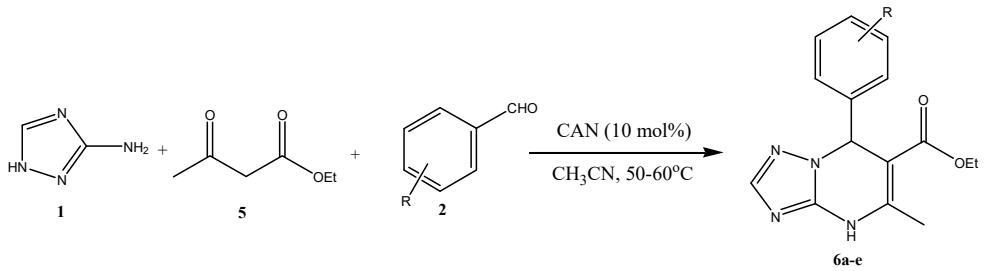



**Scheme 3**

**Table 2.** Synthesis of 1,2,4-triazoloquinazolinone derivatives using CAN under mild reaction conditions[a]

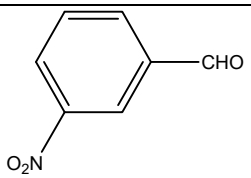
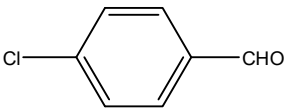
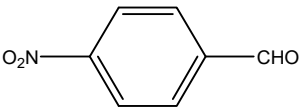
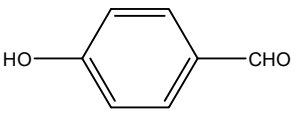
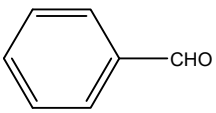
| Entry | Aldehyde | Time (min) | Yield (%)[b] | Melting point $^{o}$C |
|---|---|---|---|---|
| **4a** |  | 20 | 90 | 284-286 |
| **4b** |  | 20 | 94 | 225-227 |
| **4c** |  | 21 | 92 | 264-266 |
| **4d** |  | 17 | 96 | 302-304 |
| **4e** |  | 18 | 95 | 301-303 |
| **4f** |  | 18 | 93 | 265-268 |
| **4g** |  | 25 | 84 | 322-324 |
| **4h** |  | 20 | 94 | 250-252 |

[a]**Reaction conditions:** Dimedone (1 mmol), Aromatic aldehydes (1 mmol), 3-amino-1,2,4 triazole (1 mmol), CAN (10 mol%) in acetonitrile (5 mL) at 50-60 $^{o}$C. [b]Isolated yield.




**Scheme 4**

**Table 3.** Synthesis of **[**1,2,4]triazolo[1,5-a]pyrimidine derivatives using CAN under mild reaction conditions[a]

| Entry | Aldehyde | Time (min) | Yield (%)[b] | Melting point °C |
|---|---|---|---|---|
| **6a** |  | 10 | 92 | 207-210 |
| **6b** |  | 7 | 94 | 252-254 |
| **6c** |  | 5 | 95 | 262-264 |
| **6d** |  | 10 | 92 | 272-275 |
| **6e** |  | 7 | 92 | 191-192 |

[a]Reaction conditions: Ethyl acetoacetate (1 mmol), Aromatic aldehydes (1 mmol), 3-amino-1,2,4 triazole (1 mmol), CAN (10 mol%) in acetonitrile (5 mL) at 50-60 °C. [b]Isolated yield.

In order to find out the efficiency and greenness of the method, we compared our obtained results for the synthesis of

1,2,4-triazoloquinazolinone derivatives with the pre-eminent of the data from the literature as shown in the following Table 4, many of the formerly reported methodologies experience from one or more disadvantages such as necessity of excess amount of catalyst, high temperature or ultrasound irradiation, prolonged reaction time, use of volatile and toxic organic solvents. We believe that the present method helps to keep away from the disadvantages within the formerly reported methodologies.

**Table 4.** Comparison of the ability of various catalysts with CAN.

| Entry | Catalyst / Reaction condition | Time (min) | Yield (%)[b] | [Ref.] |
|---|---|---|---|---|
| 1 | Acetic acid (5 mL)/ 60 $^{o}$C | 25 | 95 | 22 |
| 2 | Anthranilic acid(30 mol%)/EtOH, Reflux, 80$^{o}$C | 360 | 95 | 26 |
| 3 | *p*-TsOH.$H_2O$ (15 mol%)/$CH_3CN$, 40–50 $^{o}$C | 30 | 96 | 24 |
| 4 | $NH_2SO_3H$/ $CH_3CN$, reflux, 80 $^{o}$C | 30 | 95 | 27 |
| 5 | $H_6P_2W_{18}O_{62}$ .$18H_2O$/ $CH_3CN$, 80 $^{o}$C | 30 | 95 | 21 |
| 6 | CAN (10 mol%)/ $CH_3CN$, 50-60 $^{o}$C | 20 | 96 | Present work |

[b]Isolated Yield

## Experimental

All the basic chemicals, reagents and solvents were purchased from S. D. Fine, Spectrochem, Alfa Aesar, and Loba Chemical companies and used further without purification. We have determined melting points by an open capillary tube method and are uncorrected. Progress of the reaction was tested by using alumina TLC plates (Merck 60 $F_{250}$). $^{1}$HNMR and $^{13}$CNMR spectra of synthesized heterocyclic compounds were tested by 500

MHz and 125 MHz Bruker Avance spectrometer respectively in DMSO solvents and using tetramethylsilane (TMS) as an internal standard and the value of chemical shift is in the δ scale and *J* value is in hertz (Hz). Mass spectra analyses were performed with electrospray ionization (ESI) method.

*General procedure for the synthesis of 1,2,4-triazolo-quinazolinone derivatives and [1,2,4]triazolo[1,5-a]pyrimidine-6-carboxylate*

In round bottom flask a mixture of 3-amino-1,2,4-triazole (1.0 mmol), aromatic aldehyde (1.0 mmol), dimedone (1.0 mmol) or ethylacetoacetate (1.0 mmol), 3 mL acetonitrile and 10 mol% of CAN was stirred for the 5-25 min at 50-60 $^{o}$C. Progress of the reaction was monitored by TLC. After the formation of product, then reaction mixture was cooled to room temperature. The solid products were separated by filtration washed with ethanol. The synthesized pure compounds were characterized by spectroscopic methods.

*Selected spectral data:*

*6,6-dimethyl-9-(4-bromophenyl)-5,6,7,9-tetrahydro[1,2,4]-triazolo[5,1b] quinazolin-8 (4H)-one (Table 2, entry 4a)*

Pale yellow solid; IR(KBr): 765, 835, 1252, 1364, 1580, 1642, 2886, 2956, 3082 cm$^{-1}$; $^{1}$H NMR (500 MHz, DMSO-d6): δ 0.96 (s, 3H, $CH_3$), 1.09 (s, 3H, $CH_3$), 2.20 (q, J = 10.38, 16.43 Hz, 2H, –$CH_2$), 2.55 (s, 2H, –$CH_2$), 6.21 (s, 1H, –CH), 7.14-7.16 (d, J = 8.30 Hz, 2H, Ar-H), 7.48-7.50 (d, J = 8.30 Hz, 2H, Ar-H), 7.71 (s, 1H, Ar-H), 11.19 (s, 1H, NH); $^{13}$C NMR (125 MHz, DMSO-d6): δ 27.40, 28.86, 32.69, 50.24, 57.94, 105.64, 121.33, 129.73, 131.69, 141.43, 147.29, 150.69, 151.07, 193.49; MS m/z (ESI): 373 $[M+H]^{+}$.

*6,6-dimethyl-9-(4-methoxyphenyl)-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1b] quinazolin-8 (4H)-one (Table 2, entry 4b)*

Colourless solid; IR (KBr): 765, 829, 1252, 1364, 1580, 1635, 2950, 3095 $cm^{-1}$. $^{1}H$ NMR (DMSO-$D_6$, 500 MHz): δ 0.75 (s, 3H). 0.82 (s, 3H), 2.07(d, *J* = 16Hz, 1H), 1.83 (d, *J* = 16Hz, 1H), 1.98 (d, *J* =16Hz, 1H), 2.28-2.37 (m, 2H), 3.47 (s, 3H), 5.93 (s, 1H), 6.60 (d, *J* = 8Hz, 2H), 6.87 (d, *J* = 8Hz, 2H), 7.45 (s, 1H), 10.87 (s, 1H), $^{13}C$ NMR (DMSO-$D_6$, 125 MHz): δ 26.79, 28.49, 32.15, 49.78, 54.99, 57.30, 105.71, 113.54, 128.09, 133.82, 146.72, 150.14, 158.66, 192.94; ESI-MS: *m/z* 325 $[M+H]^+$.

*6,6-Dimethyl-9-p-tolyl-5,6,7,9-tetrahydro-[1,2,4]triazolo[5,1-b]quinazolin-8(4H)-one (Table 2, entry 4c)*

White solid; IR (KBr): 756, 1253, 1368, 1581, 1649, 2924, 3091 $cm^{-1}$; $^{1}H$ NMR (500 MHz, DMSO-d6): δ 0.96 (s, 3H, $CH_3$), 1.04 (s, 3H, $CH_3$), 2.20 (d, J = 11.52 Hz, 2H, $CH_2$), 2.39 (s, 3H, $–CH_3$), 2.50-2.58 (m, 2H, $–CH_2$), 6.16 (s, 1H, –CH), 7.07 (S, 4H, Ar-H), 7.67 (s, 1H, Ar-H), 11.10 (s, 1H, NH); $^{13}$CNMR (125 MHz, DMSO-$d_6$): δ 19.7, 26.0, 37.7, 31.2, 59.1, 59.9, 105.2, 125.7, 127.7, 136.0, 137.3, 145.8, 148.6, 148.9, 192.2; MS m/z (ESI); 309 $[M+H]^+$.

*6,6-dimethyl-9-(4-nitrophenyl)-5,6,7,9-tetrahydro[1,2,4]triazolo[5,1b] quinazolin-8-(4H)-one (Table 2, entry 4d*)

Pale yellow solid; IR (KBr): 852, 1252, 1346, 153, 1643, 2961, 3080, 3105 $cm^{-1}$. $^{1}H$ NMR (DMSO-$D_6$, 500 MHz): δ 0.96(s, 3H). 1.05 (s, 3H), 2.07(d, *J* = 16Hz, 1H), 2.21 (d, *J* = 16Hz, 1H), 2.57 (d, *J* =16Hz, 1H), 2.50 (d, *J* = 16Hz, 1H), 6.37 (s, 1H), 7.50 (d, *J* = 8Hz, 2H), 7.74 (s, 1H), 8.17 (d, *J* = 8Hz, 2H), 11.31 (s, 1H), $^{13}$CNMR (DMSO-$D_6$,125 MHz): δ 27.45, 28.78, 32.72, 50.17, 58.01, 105.24, 124.06, 128.97, 147.33, 147.43, 148.90, 150.92, 151.48, 193.52; ESI-MS: *m/z* 340 $[M+H]^+$.

*6,6-dimethyl-9-(4-Chlorophenyl)-5,6,7,9-tetrahydro[1,2,4]-triazolo[5,1b] quinazolin-8 (4H)-one (Table 2, entry 4e)*

Pale yellow solid, IR (KBr): 795, 1253, 1367, 1579, 1649, 2962, 3088, 3124 $cm^{-1}$; $^1H$ NMR (DMSO-$D_6$, 500 MHz): δ = 0.96 (s, 3H, -$CH_3$), 1.08 (s, 3H, -$CH_3$), 2.07(d, *J* = 16Hz, 1H,-$CH_2$), 2.27 (d, *J* = 16Hz, 1H,-$CH_2$), 2.50-2.58 (d, *J* =16Hz, 2H,-$CH_2$), 6.22 (s, 1H, -CH), 7.19–7.37 (m, 4H, Ar–H) 7.71 (s, 1H, NH) 11.19 (s, 1H, NH); $^{13}$CNMR (DMSO-$D_6$, 125 MHz) δ 27.40, 28.86, 31.73, 32.69, 50.24, 57.86, 105.69, 128.76, 129.38, 132.77, 141.01, 147.29, 150.68, 151.06, 193.48; MS m/z (ESI): 329 $[M+H]^+$.

*6,6-dimethyl-9-phenyl-5,6,7,9-tetrahydro [1,2,4]-triazolo[5,1b]quinazolin-8(4H)-one (Table 2 , entry 4h)*

Pale yellow solid, IR (KBr): 721, 1252, 1373, 1594, 1650, 2962, 3090 $cm^{-1}$; $^1H$ NMR (DMSO-$D_6$, 500 MHz): δ = 0.95 (s, 3H, -$CH_3$), 1.03 (s, 3H, -$CH_3$), 2.05(d, *J* = 16Hz, 1H,-$CH_2$), 2.19 (d, *J* = 16Hz, 1H,-$CH_2$), 2.52-2.59 (m, 2H,-$CH_2$), 6.19 (s, 1H, -CH), 7.17-7.29 (m, 5H, Ar–H) 7.68 (s, 1H, NH) 11.14 (s, 1H, NH); $^{13}$C NMR (DMSO-$D_6$, 125 MHz) δ 26.77, 28.45, 32.16, 49.74, 57.89, 105.55, 126.92, 127.69, 128.23, 141.55, 146.82, 150.24, 150.39, 192.96; MS m/z (ESI): 295 $[M+H]^+$.

*Ethyl-4,7-dihydro-5-methyl-7-(4-chlorophenyl)[1,2,4]-triazolo[1,5a]pyrimidine-6 carboxylate (Table 3, entry 6b)*

Pale yellow solid; IR (KBr): 779, 829, 1246, 1372, 1586, 1691, 2866, 2984, 3095 $cm^{-1}$; $^1H$ NMR (500 MHz, DMSO-d6): δ 1.06 (t, 3H, $CH_3$), 2.42 (s, 3H, $CH_3$), 3.92-3.99 (q, 2H, –$CH_2$), 6.28 (s, 1H, –CH), 7.24-7.26 (d, 2H, J = 8.30 Hz, Ar-H), 7.36-7.38 (d, J = 8.30 Hz, 2H, Ar-H), 7.67 (s, 1H, Ar-H), 10.86 (s, 1H, NH); $^{13}$CNMR (125 MHz, DMSO-d6): 14.3, 18.9, 59.3, 59.8, 97.2, 128.8, 129.0, 129.1, 129.4, 132.9, 141.5, 147.3, 147.5, 150.7, 165.4; MS m/z (ESI): 319 $[M+H]^+$.

*Ethyl-4,7-dihydro-5-methyl-7-(4-hydroxy phenyl)-[1,2,4]-triazolo[1,5-a] pyrimidine-6-carboxylate (Table 3, entry 6d)*

Pale yellow solid; IR(KBr): 731, 821, 1252, 1372, 1586, 1691, 2866, 2976, 3151 $cm^{-1}$; $^{1}H$ NMR (500 MHz, DMSO-d6): δ 1.07 (t, 3H, $CH_3$), 2.07 (s, 3H, $CH_3$), 3.92-3.99 (q, 2H, $–CH_2$), 4.0 (s, 1H, –OH), 6.16 (s, 1H, –CH), 6.65-6.99 (d, 2H, J = 8.30 Hz, Ar-H), 7.02 (d, J = 8.30 Hz, 2H, Ar-H), 7.63 (s, 1H, Ar-H), 10.71 (s, 1H, NH); $^{13}$CNMR (125 MHz, DMSO-d6): 14.4, 18.8, 59.4, 59.7, 98.0, 115.7, 128.6, 133.1, 146.6, 147.3, 150.4, 157.4, 165.6; MS m/z (ESI); 301 $[M+H]^{+}$.

## Conclusions

In summary, we have developed highly efficient protocol for the synthesis of [1,2,4]-triazoloquinazolinone and [1,2,4]-triazolo[1,5-a] pyrimidine derivatives from the condensation reaction of dimedone or ethyl acetoacetate, aromatic aldehyde and 3-amino-1,2,4 triazole using CAN (10 mol%) in acetonitrile at 50-60 $^{o}$C. Present protocol offers many advantages such as short reaction time, easy isolation, simple procedure, inexpensive, mild reaction condition and no need of chromatographic separation.

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