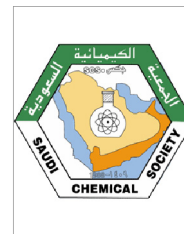




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ORIGINAL ARTICLE

Synthesis of *N*-arylacetamides via amination of aryltriazenes with acetonitrile under metal-free and mild conditions



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Abstract A transition-metal-free synthetic strategy has been developed for the synthesis of *N*-aryl amides. The present amination reaction of aryltriazenes with acetonitrile was carried out with acetonitrile as nitrogen donor, stable aryltriazenes as aryl precursors by the cleavage of C–N bond, water as oxygen source, and Brønsted acidic ionic liquids (BAILs) as potential promoter under ambient conditions. Remarkably, the practicality of this method was further proved through the reusability of the promoter BAILs. The environmentally benign nature, stable and readily available starting materials make the protocol more attractive for preparing *N*-aryl amides than traditional methods.

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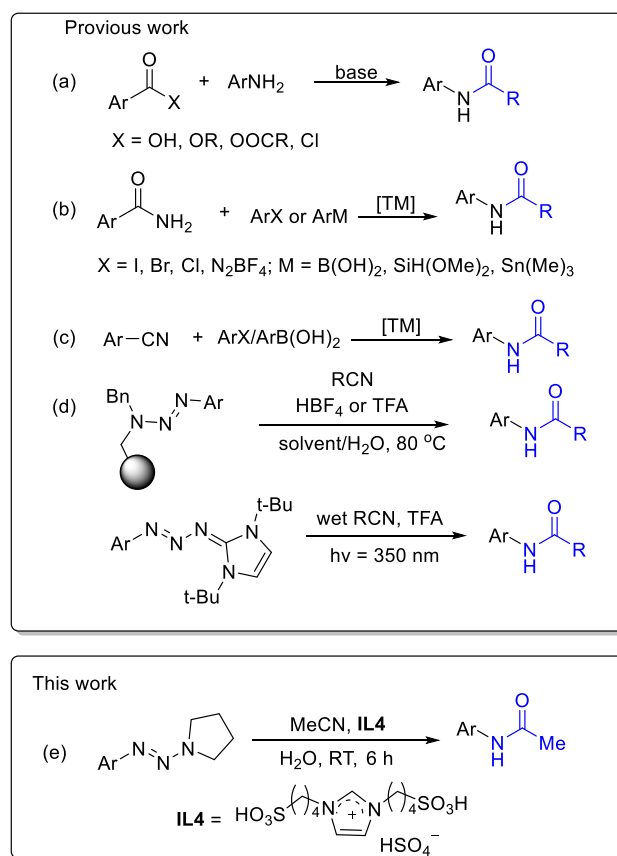
1. Introduction

Amides are indispensable in fine chemical industries and organic synthesis, including the production of medicines, functional materials, and bioactive molecules synthesis (Alcaide et al., 2007; Valeur and Bradley, 2009; Allen and Williams, 2011; Zhang et al., 2012; Garcia-Alvarez et al., 2013; Gu et al., 2016; Álvarez-Perez et al., 2019). In particular, *N*-arylacetamides are significant intermediates for the synthesis of medicinal, agrochemical, and pharmaceutical compounds (Beccalli et al., 2007; Valeur and Bradley, 2009; Allen and Williams, 2011; Garcia-Alvarez et al., 2013). Therefore, to develop efficient reaction for the synthesis of *N*-aryl amides

has attracted the attention of organic chemist (Montalbetti and Falque, 2005; Valeur and Bradley, 2009; Allen and Williams, 2011; de-Figueiredo et al., 2016). To date, several classical methods have been paved to access *N*-aryl amides. For instance (Scheme 1): (a) acylation of anilines with carboxylic acid or its derivatives in the presence of base (Wamser and Yates, 1989; Ueda and Nagasawa, 2009; Shi et al., 2010; Choudhary and Dumbre, 2011; Majumdar and Ganai, 2013; Mirza-Aghayan, et al., 2016; Ben-Halima et al., 2017; Guo et al., 2017; Kalla et al., 2017; Sonawane, et al., 2017); (b) cross coupling reactions of primary amide with arylating agents using transition metal (TM) catalyst, arylating agents including aryl halides (Chandrasekhar, et al., 2006; Jammi et al., 2009; Teo, 2009; Yao and Wei, 2010; Zheng et al., 2012), arylboronic acids (Jammi et al., 2009; Islam et al., 2012), arylbismuth compounds (Fedorov and Finet, 1999), aryl(trialkyl)stannanes (Lam et al., 2002), aryllead compounds (Lopez-Alvarado et al., 1995), diaryliodonium salts (Kang et al., 2000; Bhojane et al., 2014) and arylsiloxanes (Lam et al., 2001; Lin et al., 2009); (c) TM catalyzed domino coupling of nitriles with aryl halides or aryl boronic acids (Prakash et al., 2009; Lee et al., 2010; Hsieh et al., 2012; Garcia-Alvarez et al., 2013; Wang et al., 2013; Xiang et al., 2013; Chen et al., 2014; Saikia et al., 2016; Guo et al., 2017; Qiao et al., 2017); (d) Ritter-type reaction *via* cleavage of immobilized aryltriazenes and π -conjugated aryltriazenes (Wippert et al., 2019; Barragan et al., 2020). To date, the classical method for the synthesis of *N*-aryl amides is the cross coupling reactions of amides with aryl halides. However, involving of noble or other transition metal catalysts, toxic reagents and harsh reaction conditions impede their extensive use. In particular, some reactions involve harsh, anaerobic, anhydrous reaction conditions and use toxic solvents, which do not meet the concept of green organic synthesis. Furthermore, the use of TM catalyst limits their application in food additives and pharmaceutical synthesis. Therefore, to develop mild, convenient and metal-free protocol for the synthesis of *N*-aryl amides is still highly desired.

Recently, aryltriazenes were predominantly employed as highly powerful and diversity building blocks to access organic functional molecular compounds, because this compounds has advantage of good stability, easy preparation and mild reaction condition (Gampbell and Day, 1951; Vaughan and Stevens, 1978; Julliard et al., 1980; Kimball and Haley, 2002; Zhang et al., 2015ab). As a stable and safe aryldiazonium salt surrogate, aryltriazenes are expected to supplant aryldiazonium salts to construct C—C and C-heteroatom bonds under mild conditions (Ku and Barrio, 1981; Bräse and Schroein, 1999; Kimball et al., 2002a, 2002b, 2002c; Li et al., 2004; Liu and Knochel, 2007; Döbele et al., 2010; Goeminne et al., 2010; Romanato et al., 2010, 2011; Hafner and Bräse, 2012, 2013; Zhu and Yamane, 2012; Kirk et al., 2013; Yang et al., 2013; Liu et al., 2014, 2019; Li and Wu, 2015; Zhang et al., 2015ab, 2017, 2018; Cao et al., 2016; Wippert et al., 2019).

In recent years, from sustainable and environmental protection points of view, the use of ionic liquids (ILs) as reaction media or catalysts in organic synthesis has attracted much attention due to its nontoxic, commonly reusable, environmentally benign and unique physical/chemical properties, as compared with volatile organic solvents (Kim et al., 2002; Jadhav et al., 2015; Zhang et al., 2018; Liu et al., 2019). Especially, acidic-functionalized ionic liquids (AFILs) have been treated



Scheme 1 Different approaches for the synthesis of *N*-aryl amide.

as a fascinating dual class of solvent and catalysts, which have both the catalytic performance of Brønsted acid and uncommon properties of ILs (Hajipour and Rafiee, 2010; Amarasekara, 2016; Song et al., 2016; Kalla et al., 2017; Mahato et al., 2017; Liu et al., 2019). However, to the best of our knowledge, there are no reports comprising of the ionic liquids activation of aryltriazene C—N bond cleavage and subsequent for the synthesis of *N*-arylacetamides. In continuation of our recent interest to develop green organic synthesis methodology with aryltriazenes by using ILs as promoter (Cao et al., 2016; Zhang et al., 2017, 2018; Liu et al., 2019), in an attempt to use aryltriazenes with an aim to promote amination of acetonitrile, we used aryltriazene as the aryldiazonium salt surrogate due to its good stability and easy preparation. Herein, we report the synthesis of *N*-arylacetamides by the reaction of aryltriazene with acetonitrile with water as oxygen source and ionic liquids as promoter at room temperature (Scheme 1e).

2. Experimental

Unless otherwise stated, all commercial reagents were purchased from Adamas-beta or Energy Chemical and used without further purification. Aryltriazenes were synthesized by the literature method with some modification, all of these compounds are known (Gross et al., 1993). All reactions were performed in air. Thin layer chromatography was performed with

GF254 thin layer chromatography plate. ^1H NMR and ^{13}C NMR spectra were obtained on Varian Inova-400 MHz NMR spectrometer using CDCl_3 as solvent and tetramethylsilane as an internal standard. Chemical shifts for protons are reported in parts per million downfield and are referenced to residual protium in the NMR solvent ($\text{CHCl}_3 = \delta$ 7.26). Chemical shifts for carbon are reported in parts per million downfield and are referenced to the carbon resonances of the solvent ($\text{CDCl}_3 = \delta$ 77.0). Data are represented as follows: chemical shift, multiplicity (br = broad, s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet), coupling constants in Hertz (Hz), integration. High-resolution mass spectra (HRMS) were recorded on Thermo Fisher Scientific Ultimate 3000/Q-Exacte instrument. The melting point was recorded on a BÜCHI (M-560) melting point apparatus.

2.1. General procedure for the synthesis of *N*-Arylacetamides

The synthesis of *N*-arylacetamide **3a** given here is representative. A mixture of **IL4** (43.9 mg, 0.1 mmol), 1-(phenyldiazenyl)pyrrolidine **1a** (17.5 mg, 0.1 mmol), CH_3CN (1 mL), H_2O (0.5 mL) were added to a 10 mL clear and dried reaction tube. The reaction mixture was stirred at RT for 6 h. After completion of the reaction (monitored by TLC), 5 mL of water was added to the mixture and extracted with ethyl acetate (3×5 mL). The combined organic layer was washed with water (20 mL) and dried over anhydrous sodium sulfate, after filtration through celite, all the volatile solvents were concentrated under vacuum. The crude residue was subjected to a preparative GF254 thin layer chromatography plate (a mixture of petroleum ether and ethyl acetate as eluent) to purify and obtain the desired *N*-arylacetamide **3a**.

***N*-phenylacetamide (3a).** Yellow solid. 54% yield. m.p.: 112–114 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 8.50 (br, 1H), 7.52 (d, $J = 7.6$ Hz, 2H), 7.27 (t, $J = 7.6$ Hz, 2H), 7.08 (t, $J = 7.6$ Hz, 1H), 2.13 (s, 3H). ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 169.2, 137.9, 128.7, 124.2, 120.2, 24.2. HRMS (ESI) calcd. for $\text{C}_8\text{H}_{10}\text{NO}$ ($[\text{M} + \text{H}]^+$): 136.0757, found: 136.0755.

***N*-(*p*-tolyl)acetamide (3b).** White solid. 62% yield. m.p.: 149–151 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.36 (d, $J = 8.0$ Hz, 2H), 7.18 (br, 1H), 7.11 (d, $J = 8.0$ Hz, 2H), 2.30 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.2, 135.3, 134.0, 129.5, 120.0, 24.5, 20.8.

***N*-(4-ethylphenyl)acetamide (3c).** Brown solid. 52% yield. m.p.: 93–95 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.39 (d, $J = 8.4$ Hz, 2H), 7.15 (d, $J = 8.4$ Hz, 2H), 7.14 (br, 1H), 2.61 (q, $J = 7.6$ Hz, 2H), 2.17 (s, 3H), 1.21 (t, $J = 7.6$ Hz, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.2, 140.4, 135.4, 128.3, 120.1, 28.3, 24.6, 15.6.

***N*-(4-*iso*-propylphenyl)acetamide (3d).** Brown solid. 49% yield. m.p.: 108–110 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.40 (d, $J = 8.4$ Hz, 2H), 7.18 (d, $J = 8.4$ Hz, 2H), 7.14 (br, 1H), 2.92–2.82 (m, 1H), 2.17 (s, 3H), 1.23 (d, $J = 6.8$ Hz, 6H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.1, 145.1, 135.4, 126.9, 120.1, 33.6, 24.6, 24.0.

***N*-(4-*tert*-butylphenyl)acetamide (3e).** Brown solid. 53% yield. m.p.: 170–172 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.41 (d, $J = 8.8$ Hz, 2H), 7.33 (d, $J = 8.8$ Hz, 2H),

7.31 (br, 1H), 2.16 (s, 3H), 1.30 (s, 9H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.3, 147.3, 135.2, 125.8, 119.8, 34.3, 31.3, 24.5.

***N*-(4-methoxyphenyl)acetamide (3f).** Brown solid. 48% yield. m.p.: 129–131 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.39 (d, $J = 9.2$ Hz, 2H), 7.10 (s, 1H), 6.86 (d, $J = 9.2$ Hz, 2H), 3.80 (s, 3H), 2.17 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.1, 156.5, 130.9, 121.9, 114.1, 55.5, 24.4.

***N*-(4-chlorophenyl)acetamide (3g).** Yellowish solid. 38% yield. m.p.: 180–181 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.45 (d, $J = 8.8$ Hz, 2H), 7.28 (d, $J = 8.8$ Hz, 2H), 7.25 (br, 1H), 2.17 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.3, 136.4, 129.3, 129.0, 121.0, 24.7.

***N*-(4-iodophenyl)acetamide (3h).** Brown solid. 50% yield. m.p.: 185–187 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.60 (d, $J = 8.8$ Hz, 2H), 7.28 (d, $J = 8.4$ Hz, 2H), 7.25 (br, 1H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.2, 137.9, 137.6, 121.6, 87.4, 24.6.

***N*-(4-(trifluoromethyl)phenyl)acetamide (3i).** Yellowish solid. 50% yield. m.p.: 104–105 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.64 (d, $J = 8.8$ Hz, 2H), 7.58 (d, $J = 8.4$ Hz, 2H), 7.30 (br, 1H), 2.22 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.4, 140.8, 126.3, 124.00 (q, $J = 269$ Hz), 119.2, 24.7.

Methyl 4-acetamidobenzoate (3j). Yellowish solid. 23% yield. m.p.: 112–114 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 8.01 (d, $J = 8.0$ Hz, 2H), 7.59 (d, $J = 8.0$ Hz, 2H), 7.34 (br, 1H), 3.90 (s, 3H), 2.21 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.3, 166.6, 142.0, 130.9, 125.6, 118.7, 52.0, 24.8.

***N*-(*m*-tolyl)acetamide (3l).** Yellowish solid. 47% yield. m.p.: 63–64 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.46 (br, 1H), 7.35 (s, 1H), 7.27 (d, $J = 7.6$ Hz, 1H), 7.18 (t, $J = 7.6$ Hz, 1H), 6.92 (d, $J = 7.6$ Hz, 1H), 2.33 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.4, 138.8, 137.8, 128.7, 125.1, 120.6, 117.0, 24.6, 21.4.

***N*-(3-methoxyphenyl)acetamide (3m).** Yellowish solid. 45% yield. m.p.: 103–104 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.30 (br, 1H), 7.27 (s, 1H), 7.20 (t, $J = 8.0$ Hz, 1H), 6.96 (d, $J = 8.0$ Hz, 1H), 6.66 (d, $J = 8.0$ Hz, 1H), 3.79 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.4, 160.1, 139.1, 129.6, 111.9, 110.0, 105.6, 55.3, 24.7.

***N*-(3-chlorophenyl)acetamide (3n).** Brown solid. 32% yield. m.p.: 77–78 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.62 (s, 1H), 7.33 (d, $J = 8.0$ Hz, 1H), 7.29 (br, 1H), 7.23 (t, $J = 8.0$ Hz, 1H), 7.08 (d, $J = 8.0$ Hz, 1H), 2.18 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.3, 138.9, 134.6, 130.0, 124.3, 119.9, 117.7, 24.7.

***N*-(*o*-tolyl)acetamide (3p).** Yellowish solid. 44% yield. m.p.: 109–110 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.72 (d, $J = 8.0$ Hz, 1H), 7.19 (m, 2H), 7.08 (m, 2H), 2.25 (s, 3H), 2.19 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.4, 135.6, 130.4, 129.5, 126.7, 125.3, 123.6, 24.2, 17.8.

***N*-(3,5-dimethoxyphenyl)acetamide (3r).** Brown solid. 45% yield. m.p.: 157–158 °C. ^1H NMR (400 MHz, CDCl_3): δ (ppm) 7.35 (br, 1H), 6.76 (s, 1H), 6.75 (s, 1H), 6.23 (s, 1H), 3.77 (s, 6H), 2.16 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3): δ (ppm) 168.4, 161.0, 139.6, 98.0, 96.5, 55.4, 24.7.

3. Results and discussion

The reaction of 1-phenyltriazene (**1a**) and acetonitrile (**2a**) was used as the model reaction for the synthesis of *N*-phenylacetamide (**3a**) to screen the reaction conditions, including promoters, reaction time, temperature and the amount of water and acetonitrile, in order to obtain the optimal conditions (Table 1, entries 1–15). Initially, we examined the influence of the promoters (Table 1, entries 1–7). To our delight, the results indicated that the strong acidic ionic liquid **IL4** was found to be the most efficient promoter (entry 4), which produced the desired product *N*-phenylacetamide **3a** in 49% isolated yield. We also examined other promoter such as *p*-TSA, but this promoter was found to be ineffective in our reaction (entry 7). Furthermore, no desired product **3a** was detected in the absence of a promoter (entry 8). The effect of reaction time was then studied by variation of the reaction time (entries 4, 9), it turns out that 6 h is enough to finish the reaction (entry 9). Considering that water plays an important role in this reaction, the amount of water was also tested (entries 9–13), and the optimum amount of water was proved to be 0.5 mL. Furthermore, reducing the amount of acetonitrile or enhancing the reaction temperature could not improve the yield (entry 14, 15). The optimal reaction condition are therefore as follows: a mixture aryltriazene (0.1 mmol), acetonitrile (1 mL), and 1,3-Bis(4-sulfobutyl)-1*H*-imidazol-3-ium hydrogen sulfate (**IL4**) (0.1 mmol) in water (0.5 mL) stirred at room temperature for 6 h under air.

After obtaining the initial optimal conditions, we next investigated the mask group of 1-phenyltriazenes (Table 2). Both cyclic and non-cyclic mask group were investigated comprehensively. The expansion of protect group ring size did not improve the yield (entries 1 and 2). The ring containing an additional nitrogen atom, such as morpholinyl group, only afforded a low yield (entry 3). In addition, two separate cyclohexyl substituted substrate diminished the yield to 22% (entry 4). After the investigation of cyclic substituents, the acyclic substituents were then screened. Both linear and branched chains substituted aryltriazenes, including methyl, methoxy, ethyl, propyl, butyl and isopropyl. However, none of above substrates was revealed to be efficient for this transformation, which only afforded products in 20–40% yields (entries 5–10). Considering that the free OH groups might have hydrogen bonding effect and might be beneficial for enhancing solubility of aryltriazene in water, the ethyl alcohol substituted 1-phenyltriazene was introduced into the reaction, however, just achieved a low yield (entry 11). Finally, the diversified reactive substitutions were screened to improve the yield, such as allyl and benzyl substituted compounds were performed, but do not seem to improve the yield (entries 12 and 13). Besides the sp^3 substituent groups, the sp^2 ones were also studied, which failed to increase yields (entry 14). Therefore, pyrrolidinyl was chosen as the optimum mask group for this amination reaction.

With the optimized mask groups of 1-aryltriazenes in hand, the aryl substitutions of aryltriazenes were then explored comprehensively (Table 3). Reactions of various 1-aryltriazenes were carried out with acetonitrile to obtain the corresponding products *N*-aryl amides. The unsubstituted and electron-neutral substituted compounds gave moderate yields (**3a–3e**). The substrates with electron-donating groups at *para*-positions of phenyl were effective in this reaction (**3f**). How-

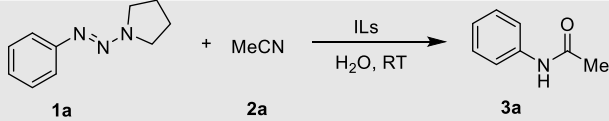
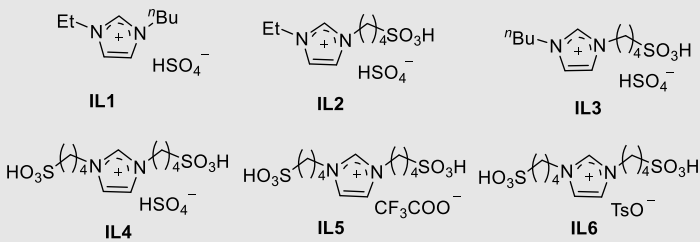
ever, aryltriazenes with electron-deficient substituents *para*-positions of phenyl were not well-tolerated during this transformation (**3g–3k**). Weak electron-deficient groups Cl, I, CF_3 , and ester at the C4 aryl *para*-position of phenyl provided desired product in 23–50% yields. The strong electron-deficient group nitro led to no reaction (**3k**). This appearance could probably be explained by the fact that electron-deficient groups not conducive to the formation of the stable aryl cation intermediate. The electron-neutral and electron-donating group in *meta*-position of phenyl of aryltriazenes also furnished the product in slightly lower yields (**3l**, **3m**). Moreover, the weak electron-withdrawing group in *meta*-position resulted in relatively lower yields of products (**3n**), and the strong electron-withdrawing nitro substituted aryltriazene was also failed to react with acetonitrile under standard reaction (**3o**). Notably, halogen substitutions at *para*- and *meta*-positions of aryltriazene were well tolerated in the present transformation (**3g**, **3h**, **3n**), with no detection of dehalogenative, which demonstrate that this strategy has good potential in further transformation. In addition, sterically hindered substrates, i.e., *ortho*- (**3p**) and 3,5-dimethoxy- (**3r**) substituted substrate were also tolerated in this reaction. Although the electron-withdrawing group ketone at *ortho*-position was investigated under optimized reaction condition, our efforts were proved to be futile and no desired product **3q** was obtained.

One of the advantages of ILs is its ability to function as a recyclable reagent and solvent. To confirm this IL can be used as a recyclable promoter, the recycling of the **IL4** was explored under the optimal reaction conditions (Fig. 1). Following each cycle, the reaction mixture was washed with H_2O and EtOAc, and the promoter **IL4** was extracted from the aqueous layer and reused for the next reaction after drying under reduced pressure. The above results confirmed that our promoter can be recycled at least four times with almost the same efficiency. The yield decreased gradually after four cycles, the desired product was still obtained with 33% in the eighth cycle. These results suggested that the **IL4** promoter was stable in this amination reaction.

To further gain the reaction mechanistic insights, a series of control experiments were performed. Firstly, to explore the source of oxygen in the final product and the role of water, the reaction between 1-phenyltriazene (**1a**) and acetonitrile (**2a**) was carried out under standard conditions in the presence of H_2O^{18} (Scheme 2a), the corresponding ^{18}O -labeled *N*-phenylacetamide (**3a- O^{18}**) was obtained, which was confirmed by HRMS analysis. The ^{18}O exchange experiment indicated that ^{16}O amide did not exchange an oxygen atom with $H_2^{18}O$, which was consistent with the $H_2^{18}O$ labelled experiment. Therefore, water might be involved in the reaction and provided the O atom.

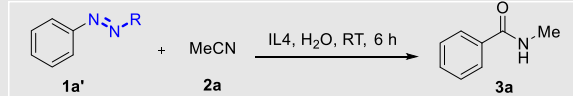
According to relevant reports (Prakash et al., 2009; Lee et al., 2010; Ramanathan and Liu, 2015; Hu et al., 2017; Zhang et al., 2018; Liu et al., 2019) and the results above, we proposed a possible reaction pathway (Scheme 3). At first, **1a** was activated by promoter **IL4** to produce ammonium salt **A**. Then, intermediate **A** was decomposed into aryl cation by the release molecule of pyrrolidine and N_2 respectively. Then, nucleophilic attack to the aryl cation by the acetonitrile results in the formation of a nitrilium ion intermediate **B**, which is captured by water to produce intermediate **C**. Finally, product

Table 1 Optimization of the reaction conditions.

				
				
Entry ^a	ILs	H ₂ O/mL	Time/h	Yield/% ^b
1	IL1	0.5	48	Trace
2	IL2	0.5	48	21
3	IL3	0.5	12	41
4	IL4	0.5	12	49
5	IL5	0.5	12	35
6	IL6	0.5	12	47
7	<i>p</i>-TSA	0.5	12	25
8	-	0.5	12	ND ^c
9	IL4	0.5	6	54
10	IL4	0.1	6	40
11	IL4	0.2	6	40
12	IL4	0.3	6	44
13	IL4	0.4	6	48
14 ^d	IL4	0.5	6	Trace
15 ^e	IL4	0.5	6	52

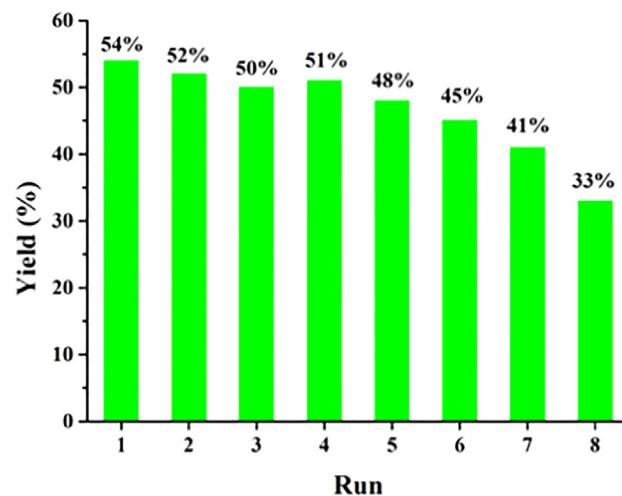
^aConditions: **1a** (0.1 mmol), **2a** (1 mL), **IL** (0.1 mmol), H₂O (0.5 mL), at room temperature.^bIsolated yields.^cND = No detection.^d0.1 mmol **2a** used.^eReaction was performed at 50 °C.

Table 2 Optimization of the mask group of 1-phenyltriazenes.

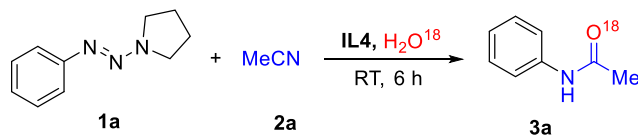
					
Entry ^a	R	yield ^b	Entry ^a	R	yield ^b
1		54%	8		40%
2		50%	9		35%
3		41%	10		40%
4		22%	11		Trace
5		22%	12		20%
6		20%	13		43%
7		25%	14		20%

^aConditions: **1a** (0.1 mmol), **2a** (1 mL), **IL4** (0.1 mmol), H₂O (0.5 mL), at room temperature.

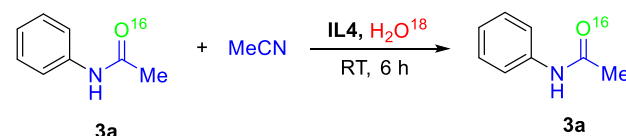
^bIsolated yields.

**Fig. 1** Recycling of **IL4** for the synthesis *N*-phenylacetamide.

a) H₂¹⁸O-labeled experiment



b) ¹⁸O-exchange experiment

**Scheme 2** Control experiments.

3a was afforded by the deprotonation and isomerization of **C**, and **IL4** was regenerated at same time.

4. Conclusions

In summary, we have developed a novel method for the synthesis of *N*-arylacetamides from aryltriazenes and acetonitrile at room temperature. Notably, the promoter **IL4** could be easily recycled and reused with the similar efficacies for at least four cycles. The easy availability of starting materials and the fairly mild reaction conditions without using volatile organic solvents make this reaction attractive for organic synthesis.

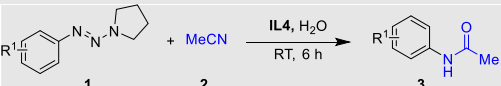
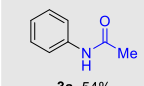
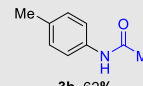
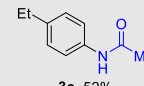
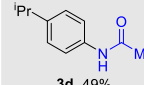
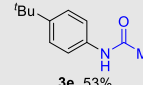
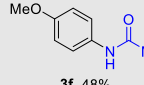
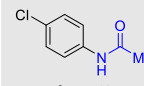
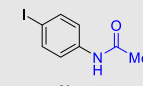
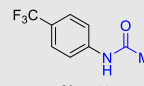
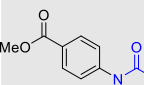
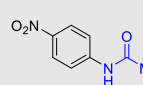
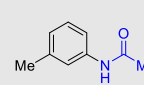
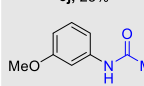
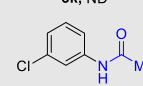
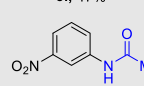
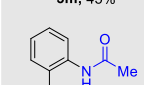
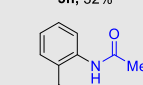
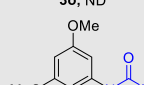
Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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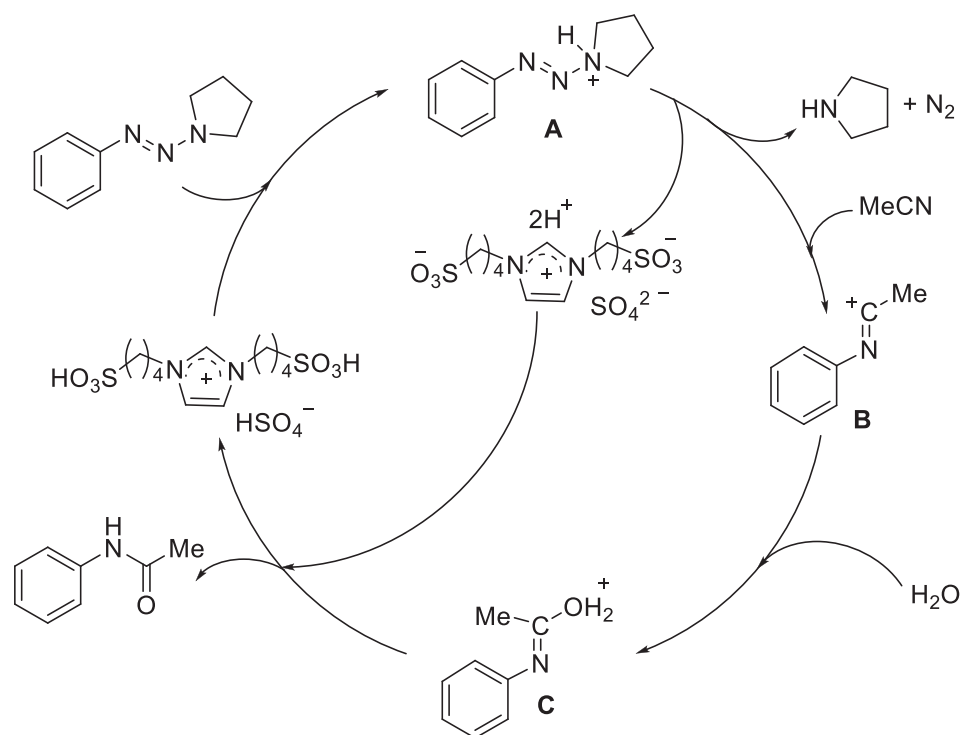
Table 3 Substrate scope of aryltriazenes.

^aConditions: **1a** (0.1 mmol), **2a** (1 mL), **IL4** (0.1 mmol), H₂O (0.5 mL), at room temperature.

^bIsolated yields.

^cReacted for 24 h.



Scheme 3 Plausible mechanism for *N*-phenylacetamide.

dition of Xinjiang Uyghur Autonomous Region (2017D01C035 and 2017D01C075).

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.arabjc.2021.103158>.

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