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One Pot Synthesis and Characterization of Mono and Di-substituted Azo-Containing Amides

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Azo-containing amides and their derivatives were synthesized by the reaction of 4-(phenyldiazenyl)aniline with different substituted benzoyl chlorides. The characterization of these synthesized compounds were based on their IR, H^1 NMR spectra and elemental analysis with excellent yields.

Keywords: 4-(Phenyldiazenyl)aniline, Amidation, *N*-protection, *N*-acylation.

INTRODUCTION

Amides, ureas and sulfonamides are used everywhere for structural enhancement within drug design and discovery. The catalytically C-C and C-N bonds formation is a vigorous theme in the field of current organic synthesis[1-3]. Amides are among the great consequence functional groups in natural products, polymers and pharmaceuticals lead compounds[4,5]. Their value in organic, biological and fabric chemistry directive the development of more economical methods for their synthesis of these compounds[5,6]. In general, aliphatic, aromatic and heteroaromatic amides can be synthesized from the reaction between carboxylic acids or their derivatives with amines[7]. Rovis and Bode and their co-workers reported amidation with N-heterocyclic carbenes (NHCs) as catalyst[8]. Azo containing compounds, with two phenyl rings alienated by an azo (-N=N-) bond, are multipurpose molecules and highly acknowledged in research vicinity both fundamental and relevance. The strong electronic absorption utmost can be adapted by different ring substitution to fall ultraviolet to red-visible regions, allocating chemically fine-tuning of dyes[9-12]. This collective fact that azo groups are comparatively vigorous and chemically stable has provoked extensive study of azobenzene support structures as dyes as well as colorant[13-17]. In addition, the light stimulated interconversion permit the system integrating azo group to be used as photo switches, consequence rapid and reversible control over a diversity of chemical, electronic, mechanical and optical properties[18,19]. Because of the high quality of thermal constancy of azo compounds, one of the most essential applications of azo group containing compounds are optical data storage. Generally, phthalocyanine dyes, cyanine dyes and metaleazo complex dyes are used for DVD-R (digital versatile disc-recordable) as well as footage layer[20-22]. Phthalocyanine dyes also have disadvantages, such as poorly soluble and elevated expenditure than cyanine dyes[23-25]. In contrast, metal-azo composite and organic azo compounds are extra stable than cyanine dyes against light, offer uncomplicated control of the wavelength according to the different substituted groups and boast tremendous thermal reliability with a metal complex[26,27]. Based upon these reflections of beyond requirements, the synthesis of azo compounds engaged an important role in fabric chemistry[28-30]. Due to the significant importance of azo-containing compounds and prolongation of our interest in syntheses of azo-based compounds, we report herein the synthesis of new azo containing amides.

EXPERIMENTAL

General procedure for synthesis of 4a-g and 5a-g: The reaction was carried out in a two neck flask. 4-(Phenyldiazenyl)-aniline in Et_3N with different substituted aromatic benzoyl chlorides and aliphatic carbonyl chlorides at room temperature. On cooling the reaction mixture was diluted with chloroform and washed with 10 % HCl solution[31,32]. The organic layer dried over anhydrous Na_2SO_4 and concentrated under reduces pressure. The resulting residue was purified by column chromatography (silica gel, EtOAc/heptanes).

***N*-[4-(Phenyldiazenyl)phenyl]benzamide (4a):** Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), benzoyl

chloride (154 mg, 1.1 mmol), according to the general procedure A, **4a** was isolated as a redish solid (253 mg, 84 %). room temperature for 10 min. m.p. = 158-159 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 7.17-7.19 (m, 1H, ArH), 7.60-7.69 (m, 5H, ArH), 7.74-7.78 (m, 4H, ArH), 8.04 (d, *J* = 7.8 Hz, 2H), 8.17 (d, *J* = 7.5 Hz, 2H, ArH), 9.8 (bs, 1H). IR (KBr, ν_{max}, cm^{-1}): 3324 (NH), 1643 (CO). Anal. Calcd. for $C_{19}H_{15}N_3O$: C 75.73, H 5.02, N 13.94; found: C 75.61, H 4.99, N 13.86.

2-Methyl-*N*-(4-(phenyldiazenyl)phenyl)benzamide (4b): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), 2-methylbenzoyl chloride (169 mg, 1.1 mmol), according to the general procedure A, **4b** was isolated as a redish solid (249 mg, 79 %). Reaction at room temperature for 10 min. m.p. = 197-198 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.51 (s, 3H), 7.16-7.18 (m, 2H, ArH), 7.22-7.24 (m, 1H), 7.58-7.66 (m, 6H, ArH), 7.73 (d, *J* = 7.4 Hz, 2H), 8.15 (d, *J* = 7.6 Hz, 2H, ArH), 10.15 (bs, 1H). Elemental analysis: C, 76.17; H, 5.43; N, 13.32. IR (KBr, ν_{max}, cm^{-1}): 3322 (NH), 1641 (CO). Anal. Calcd. for $C_{20}H_{17}N_3O$: C 76.17, H 5.43, N 13.32; found: C 76.06, H 5.42, N 13.21.

3-Methyl-*N*-(4-(phenyldiazenyl)phenyl)benzamide (4c): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), 3-methylbenzoyl chloride (169 mg, 1.1 mmol), according to the general procedure A, **4c** was isolated as a redish solid (255 mg, 81 %). Reaction at room temperature for 10 min. m.p. = 193-194 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.36 (s, 3H), 7.21-7.23 (m, 2H, ArH), 7.34-7.36 (m, 1H, ArH), 7.43 (d, *J* = 7.7 Hz, 1H, ArH), 7.61-7.64 (m, 2H), 7.73 (d, *J* = 7.6 Hz, 2H), 7.78-7.81 (m, 2H, ArH), 7.84 (d, *J* = 7.6 Hz, 2H, ArH), 8.23 (d, *J* = 7.7 Hz, 2H, ArH), 10.14 (bs, 1H). IR (KBr, ν_{max}, cm^{-1}): 3325 (NH), 1645 (CO). Anal. Calcd. for $C_{20}H_{17}N_3O$: C 76.17, H 5.43, N 13.32; found: C 76.14, H 5.41, N 13.30.

4-Methyl-*N*-(4-(phenyldiazenyl)phenyl)benzamide (4d): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), 4-methylbenzoyl chloride (169 mg, 1.1 mmol), according to the general procedure A, **4d** was isolated as a redish solid (252 mg, 80 %). Reaction at room temperature for 10 min. m.p. = 197-198 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.31 (s, 3H, CH_3), 7.18-7.19 (m, 1H, ArH), 7.39-7.41 (m, 2H), 7.63-7.65 (m, 2H, ArH), 7.69 (d, *J* = 7.6 Hz, 2H), 8.13 (d, *J* = 7.8Hz, 2H, ArH), 10.14 (bs, 1H). IR (KBr, ν_{max}, cm^{-1}): 3319 (NH), 1640 (CO). Anal. Calcd. for $C_{20}H_{17}N_3O$: C 76.17, H 5.43, N 13.32; found: C 76.13, H 5.41, N 13.30.

2-Chloro-*N*-(4-(phenyldiazenyl)phenyl)benzamide (4e): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), 2-chlorobenzoyl chloride (192 mg, 1.1 mmol), according to the general procedure A, **4e** was isolated as a redish solid (298 mg, 89 %). Reaction at room temperature for 10 min. m.p. = 213-214 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 7.20-7.22 (m, 1H, ArH), 7.44-7.45 (m, 1H), 7.53-7.54 (m, 1H, ArH), 7.66-7.70 (m, 4H, ArH), 7.78-7.80 (m, 2H, ArH), 8.01-8.04 (m, 2H, ArH), 8.33-8.35 (m, 2H, ArH), 10.23 (bs, 1H). IR (KBr, ν_{max}, cm^{-1}): 3327 (NH), 1648 (CO). Anal. Calcd. for $C_{19}H_{14}N_3OCl$: C 67.96, H 4.20, N 12.51; found: C 67.92, H 4.17, N 12.43.

3-Phenyl-*N*-(4-((*E*)-phenyldiazenyl)phenyl)acrylamide (4f): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), cinnamyl chloride (183 mg, 1.1 mmol), according to the general procedure A, **4f** was isolated as a redish solid (271 mg, 83 %). Reaction at room temperature for 10 min. m.p. = 177-178 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 6.93(d, *J* = 17.5 Hz, 1H), 7.19-7.21 (m, 2H, ArH), 7.43 (d, *J* = 17.4 Hz, 1H), 7.37-7.39 (m, 2H, ArH), 7.57-7.61 (m, 4H, ArH), 7.96-7.98 (m, 2H, ArH), 7.79 (d, *J* = 7.8 Hz, 2H, ArH), 8.34 (d, *J* = 7.7 Hz, 2H, ArH), 10.01 (bs, 1H, NH). IR (KBr, ν_{max}, cm^{-1}): 3316 (NH), 1641 (CO). Anal. Calcd. for $C_{21}H_{17}N_3O$: C 77.04, H 5.23, N 12.84; found: C 77.01, H 5.19, N 12.71.

***N*-(4-(Phenyldiazenyl)phenyl)octanamide (4g):** Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), octanoyl chloride (178 mg, 1.1 mmol), according to the general procedure A, **4g** was isolated as a redish solid (252 mg, 78 %). Reaction at room temperature for 10 min. m.p. = 89-90 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 0.78 (t, *J* = 7.7Hz, 3H, $CH_3CH_2(CH_2)_6$), 1.27-1.34 (m, 8H, $CH_3(CH_2)_4(CH_2)_3$), 1.56-1.58 (m, 2H, $CH_3CH_2(CH_2)_6$), 2.37 (t, *J* = 7.4 Hz, 2H, $CH_3CH_2(CH_2)_6$), 7.14-7.16 (m, 1H, ArH), 7.61-7.63 (m, 2H, ArH), 7.72 (d, *J* = 7.6 Hz, 2H, ArH), 7.97-7.99 (m, 2H, ArH), 8.25 (d, *J* = 7.8 Hz, ArH), 10.03 (bs, 1H). IR (KBr, ν_{max}, cm^{-1}): 3324 (NH), 1643 (CO). Anal. Calcd. for $C_{20}H_{25}N_3O$: C 74.27, H 7.79, N 12.99; found: C 74.21, H 7.73, N 12.92.

***N*-Benzoyl-*N*-(4-(phenyldiazenyl)phenyl)benzamide (5a):** Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), benzoyl chloride (308 mg, 2.2 mmol), according to the general procedure A, **5a** was isolated as a redish solid (336 mg, 83 %). Reaction temperature 45 °C for 4 h. m.p. = 136 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 7.23-7.24 (m, 1H, ArH), 7.60-7.98 (m, 14H, ArH), 8.03 (m, 2H, ArH), 8.26 (m, 2H, ArH). IR (KBr, ν_{max}, cm^{-1}): 1641 (CO). Anal. Calcd. for $C_{26}H_{19}N_3O_2$: C 77.02, H 4.72, N 10.36; found: C 77.00, H 4.69, N 10.31.

2-Methyl-*N*-(2-methylbenzoyl)-*N*-(4-(phenyldiazenyl)-phenyl)benzamide (5b): Starting with 4-(phenyldiazenyl)-aniline (**2**) (197 mg, 1 mmol), 2-methylbenzoyl chloride (339 mg, 2.2 mmol), according to the general procedure A, **5b** was isolated as a redish solid (329 mg, 76 %). Reaction temperature 45 °C for 4 h. m.p. = 148 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.46 (s, 6H, 2 × CH_3), 7.13-7.16 (m, 4H, ArH), 7.23-7.24 (m, 1H, ArH), 7.55-7.57 (m, 2H, ArH), 7.63-7.65 (m, 2H, ArH), 7.71-7.72 (m, 2H, ArH), 7.89-7.90 (m, 2H, ArH), 7.97-7.98 (m, 2H, ArH), 8.27-8.29 (m, 2H, ArH). IR (KBr, ν_{max}, cm^{-1}): 1637 (CO) cm^{-1}. Anal. Calcd. for $C_{28}H_{23}N_3O_2$: C 77.58, H 5.35, N 9.69; found: C 77.43, H 5.29, N 9.54.

3-Methyl-*N*-(3-methylbenzoyl)-*N*-(4-(phenyldiazenyl)-phenyl)benzamide (5c): Starting with 4-(phenyldiazenyl)-aniline (**2**) (197 mg, 1 mmol), 3-methylbenzoyl chloride (339 mg, 2.2 mmol), according to the general procedure A, **5c** was isolated as a redish solid (342 mg, 79 %). Reaction temperature 45 °C for 4 h. m.p. = 154 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.36 (s, 3H), 7.23-7.24 (m, 1H, ArH), 7.39-7.41 (m, 2H, ArH), 7.49-7.52 (m, 3H, ArH), 7.63-7.65 (m, 2H), 7.74 (m, 3H, ArH), 7.83-7.85 (m, 2H, ArH), 7.98-8.00 (m, 2H, ArH), 8.27-8.29 (m, 2H, ArH). IR (KBr, ν_{max}, cm^{-1}): 1640 (CO). Anal. Calcd. for $C_{28}H_{23}N_3O_2$: C 77.58, H 5.35, N 9.69; found: C 77.49, H 5.31, N 9.61.

4-Methyl-*N*-(4-methylbenzoyl)-N-(4-(phenyldiazenyl)-phenyl)benzamide (5d): Starting with 4-(phenyldiazenyl)-aniline (**2**) (197 mg, 1 mmol), 4-methylbenzoyl chloride (339

mg, 2.2 mmol), according to the general procedure A, **5d** was isolated as a redish solid (355 mg, 82 %). Reaction temperature 45 °C for 4 h. m.p. = 157 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 2.33 (s, 6H, 2 × CH_3), 7.21-7.23 (m, 1H, ArH), 7.37-7.40 (m, 4H, ArH), 7.62-7.64 (m, 2H, ArH), 7.71-7.73 (m, 2H, ArH), 7.86-7.89 (m, 4H, ArH), 8.02-8.04 (m, 2H, ArH), 8.30-8.32 (m, 2H, ArH). IR (KBr, ν_{max}, cm^{-1}): 1638 (CO). Anal. Calcd. for $C_{28}H_{23}N_3O_2$: C 77.58, H 5.35, N 9.69; found: C 77.49, H 5.28, N 9.54.

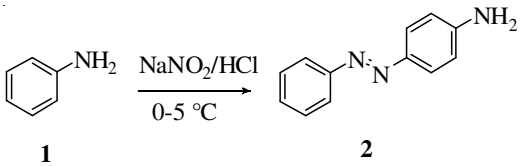
2-Chloro-*N*-(2-chlorobenzoyl)-*N*-(4-(phenyldiazenyl)-phenyl)benzamide (5e): Starting with 4-(phenyldiazenyl)-aniline (**2**) (197 mg, 1 mmol), 2-chlorobenzoyl chloride (380 mg, 2.2 mmol), according to the General procedure A, **5e** was isolated as a redish solid (431 mg, 91 %). Reaction temperature 45 °C for 4 h. m.p. = 183 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 7.25-7.26 (m, 1H, ArH), 7.41-7.43 (m, 2H, ArH), 7.49-7.51 (m, 2H, ArH), 7.63-7.66 (m, 4H, ArH), 7.71-7.73 (m, 2H, ArH), 7.75-7.77 (m, 2H, ArH), 8.01-8.03 (m, 2H, ArH), 8.31-8.33 (m, 1H, ArH). IR (KBr, ν_{max}, cm^{-1}): 1653 (CO). Anal. Calcd. for $C_{26}H_{17}C_{l2}N_3O_2$: C 65.83, H 3.61, N 8.86; found: C 65.78, H 3.54, N 8.79.

***N*-Cinnamoyl-*N*-(4-((*E*)-phenyldiazenyl)phenyl)-cinnamamide (5f):** Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), cinnamoylbenzoyl chloride (334 mg, 2.2 mmol), according to the general procedure A, **5f** was isolated as a redish solid (370 mg, 81 %). Reaction temperature 45 °C for 4 h. m.p. = 169 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 6.72 (d, *J* = 17.5 Hz, 2H), 7.21-7.23 (m, 1H, ArH), 7.34-7.37 (m, 4H, CHAr), 7.41 (d, *J* = 17.6 Hz, 1H), 7.57-7.59 (m, 4H, ArH), 7.63-7.65 (m, 2H, ArH), 7.75-7.77 (m, 2H, CHAr), 8.01-8.03 (m, 2H, CHAr), 8.29-8.31 (m, 2H, CHAr). IR (KBr, ν_{max}, cm^{-1}): 1640 (CO). Anal. Calcd. for $C_{30}H_{23}N_3O_2$: C 78.75, H 5.07, N 9.18; found: C 78.70, H 4.98, N 9.05.

(E)-*N*-Octanoyl-*N*-(4-(phenyldiazenyl)phenyl)octan-amide (5g): Starting with 4-(phenyldiazenyl)aniline (**2**) (197 mg, 1 mmol), octanoyl-chloride (356 mg, 2.2 mmol), according to the general procedure A, **5g** was isolated as a redish solid (332 mg, 74 %). Reaction temperature 45 °C for 4 h. m.p. = 175 °C. ^{1}H NMR (250 MHz, $CDCl_3$): δ = 0.78 (t, *J* = 7.7 Hz, 4H, $CH_3CH_2(CH_2)_6$), 1.23-1.30 (m, 16H, $CH_3(CH_2)_4(CH_2)_3$), 1.53-1.55 (m, 4H, $CH_3CH_2(CH_2)_6$), 2.35 (t, *J* = 7.4 Hz, 4H, $CH_3CH_2(CH_2)_6$), 7.13-7.15 (m, 2H, ArH), 7.59-7.62 (m, 4H, ArH), 7.71 (d, *J* = 7.4 Hz, 2H, ArH), 7.95-7.97 (m, 2H, ArH), 8.23 (d, *J* = 7.7 Hz, ArH). IR (KBr, ν_{max}, cm^{-1}): 1638 (CO). Anal. calc. for $C_{28}H_{39}N_3O_2$: C 74.80, H 8.74, N 9.35; found: C 74.73, H 8.68, N 9.23.

RESULTS AND DISCUSSION

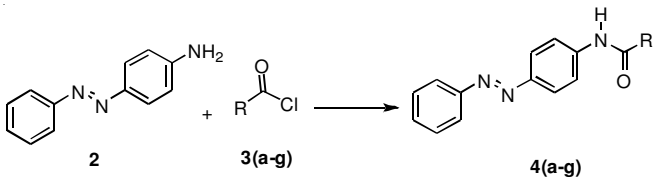
4-(Phenyldiazenyl)aniline (**2**) was prepared in 80 % yield by the reaction of commercially available aniline (**1**) with $NaNO_2$ under acedic media (**Scheme-I**).



1 2

Scheme-I: Synthesis of 4-(phenyldiazenyl)aniline (**2**). Reagents and conditions: i, Aniline (**1**) (2.0 equiv), -0-5 °C, distilled water $NaNO_2$/HCl

Different mono substituted *N*-(4-(phenyldiazenyl)phenyl)-benzamide (**4a-g**) were prepared in 79-89 % yields by the reaction of 4-(phenyldiazenyl)aniline (**2**) with 1.1 equiv. of different substituted benzoyl chlorides **3a-g** (**Scheme-II**, Table-1). The best yields were obtained when the reactions were carried out using NEt_3 and DIPA as the base as well as solvent, while employment of other base, such as NaOH, KOH resulted in a decrease of the yield. The use of potassium phosphate (K_3PO_4) as the base and 1,4-dioxane as a solvent gave optimal yields. The best yield was obtained for the reaction of simple benzoyl chlorides. The lowest yield was obtained for 4-methyl chlorides which might be attributed to its high nucleophilicity (due to the electron-donating methyl group).



2 3(a-g) 4(a-g)

Scheme-II: Synthesis of **4a-g**. Reagents and conditions: i) **2** (1 equiv.), **3a-g** (1.1 equiv.) at room temperature for 10 min (**R** is representing aryl and alkyl groups)

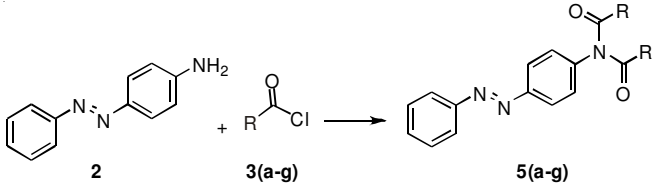
TABLE-1
SYNTHESIS OF MONO-ARYL AND ALKYL DIAZO-BENZAMIDE (**4a-g**)

4	R	% (4)[a]
a	C_6H_5	84
b	2-MeC_6H_4	79
c	3-MeC_6H_4	81
d	4-MeC_6H_4	80
e	2-ClC_6H_4	89
f	C_8H_7	83
g	C_7H_{15}	78

[a]Yield of isolated products

The amidation of 4-(phenyldiazenyl)aniline (**2**) with an equimolar ratio of different substituted benzoyl chlorides **3a-g** (2.2 equiv.) afforded the disubstituted *N*-benzoyl-*N*-(4-(phenyl-diazenyl)phenyl)benzamide **5a-g** in 76-91 % yield (Table-2, **Scheme-III**). The yields of the products derived from chloro derivative **5e** were generally higher than those of others derivatives which might be explained by the high stability of the chloro group. No clear trend was observed for the dependence of the yields from the type of benzoyl chloride employed.

The one-pot reaction of **5a-g** were also carried out by the addition of different substituted aryl or alkyl benzoyl chlorides, which were afforded two fold di-substituted arylated/alylated aiazobenzamides **5a-g**, at 90 °C for 4 h to achieve good yield. These reactions were successful for both electron-rich and



2 3(a-g) 5(a-g)

Scheme-III: Synthesis of **5a-g**; *Reagents and conditions:* i, **2** (1.0 equiv.), **3a-g** (2.2 equiv.), 4 h (R is representing aryl and alkyl groups)

TABLE-2
SYNTHESIS OF BIS-ARYL AND
ALKYL DIAZOBENZAMIDE (**5a-g**)

4	R	% (**5**)[a]
a	C_6H_5	83
b	2-MeC_6H_4	76
c	3-MeC_6H_4	79
d	4-MeC_6H_4	82
e	2-ClC_6H_4	91
f	C_8H_7	81
g	C_7H_{15}	74

[a]Yield of isolated products

electron-poor benzoyl chlorides as shown in **Scheme-III** and Table-2. During the optimization, it proved to be important that the first step was carried out at 45 °C for 10 min to achieve a good selectivity **4a-g**. All reactions proceeded in excellent yield.

Conclusion

In conclusion, we reported an efficient method for the synthesis different azo amidation of 4-(phenyldiazenyl)aniline with different substituted aromatic benzoyl chlorides and aliphatic carbonyl chlorides to get different mono- and *N*-protected di- azo-containg amides which provide a convenient and sequential azo-amidation.

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