Short communication

Polyethylene glycol in water: A simple and environment friendly media for Strecker reaction

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Abstract

Polyethylene glycol (PEG) in water is found to be an inexpensive, non-toxic and eco-friendly reaction medium for the nucleophilic addition of trimethylsilyl cyanide (TMSCN) to imines (Strecker reaction) to afford α-aminonitriles in excellent yields. This protocol is effective to a wide variety of substrates with different functional groups and does not require the use of acid or base catalyst. PEG can be recovered and reused.
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Keywords: Strecker reaction; Imines; Trimethylsilyl cyanide (TMSCN); Polyethylene glycol (PEG); Water

1. Introduction

The conventional synthetic procedures invariably use organic solvents as media to provide homogeneous phase that allows molecular interactions effectively and bring the reaction to completion. But the organic solvents used are harmful and do not drive the reactions to total completion.

Development of a synthetic protocol that is nature-friendly, simple, efficient and cost effective remains an ever-challenging objective [1]. This report on the preparation of α-aminonitriles by the nucleophilic addition of trimethylsilyl cyanide to an imine in PEG and water at an ambient temperature is a successful endeavor that realised this goal.

Strecker reaction is an efficient method for the synthesis of α-aminonitriles [2] which are useful precursors for the synthesis of α-amino acids, various nitrogen containing heterocycles [3] and chiral building blocks [4]. Usually, it is carried out by the nucleophilic addition of a cyanide anion to the imines in presence of Lewis acid/base catalyst [5]. Pre- or post-activation of C=N bond of the imine is necessary to obtain satisfactory efficiencies. For pre-activation, the C=N moiety is substituted with activating carbonyl, sulfonyl, sulfinyl, phosphoryl and silyl groups [6]. They render imine carbon more electrophilic and facilitate cyanide nucleophilic addition on it. Presence of a Bronsted/Lewis acid or a metallic species is required for post-activation of the C=N system towards cyanide addition through the formation of iminium cations [7].

Several modifications of the Strecker reaction have been made and a variety of cyanating agents such as α-trimethylsiloxy nitriles and diethylphosphorocyanidates were used under various conditions [8]. One-pot procedures were also reported for this reaction using trimethylsilyl cyanide or tributyl tin cyanide as cyanating agent in the presence of different Lewis acid catalysts such as lithium perchlorate, polymeric scandium triflamide, vanadyl and ytterbium triflates [9]. Many of these methods required strong acidic conditions, expensive reagents, long reaction times, harsh experimental conditions and tedious work-up procedures that generates large amount of toxic waste. Apart from this, these protocols are limited to aldehydes only and are unsuccessful with ketones. Even the recent reports also used ionic liquids [10], water containing DMF [11], and $Se(OTf)_3$ in water [12] as solvent medium to carry out these reactions. But the serious drawback with them is liberation of hazardous PF_6, BF_4 and HF [13] in addition to the high cost of these reagents and disposability of the solvents [14].

Polyethylene glycol (PEG) and its monomethyl ethers are inexpensive, thermally stable, recoverable, and non-toxic media for phase-transfer catalysts. PEG, a biologically acceptable

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R = aryl, alkyl; R^1 = H, CH$_3$; R^2 = aryl

Scheme 1.
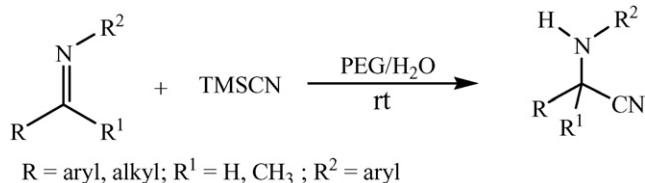
polymer used extensively in drug delivery and in bioconjugates as tool for diagnostics, has hitherto not been widely used as a solvent medium but has been used as a support for various transformations [15]. An efficient C–C bond formation by addition of cyanide anion to the imine without pre- or post-activation of its C=N bond in water in the absence of a catalyst is not yet reported. This prompted us to run Strecker reaction in an eco-friendly recyclable medium under mild conditions (Scheme 1).

2. Results and discussion

In the present procedure, trimethylsilyl cyanide, a safe and easy to handle reagent is employed as an effective cyanide ion source. No acid/base catalyst is used. PEG in water is opted as the reaction medium for three reasons: (i) it is eco-friendly, (ii) PEG can be precipitated in the work-up and recycled and (iii) PEG catalyses cyanide nucleophilic addition to the imine carbon by increasing its electrophilicity through hydrogen bonding by its hydroxyl group with the nitrogen of the imine and water plays the role as proton source. This proposition gains validity since no significant progress in this reaction is observed in the absence of PEG.

All the products were characterized by Mass, ^{1}H NMR and IR Spectroscopy and by comparison with known compounds [9].

Thus, this is the first practically feasible method for the Strecker reaction of various aldimines and ketoimines with TMSCN in PEG–water medium. The advantages of this method are: simple and mild experimental conditions, less reaction time, cost-effective due to recyclability of PEG, applicability to wide variety of substituted aldimines and ketomines, excellent yields and green chemistry avoiding hazardous solvents and toxic organic reagents.

3. Experimental

A mixture of imines (1 mmol), trimethylsilyl cyanide (1 mmol), PEG 400 (2 g) and water (1 ml) was placed in a 10 ml round-bottomed flask and the mixture was left stirring at room temperature. After completion of the reaction, the reaction mixture was cooled in dry ice–acetone bath to precipitate the PEG and extracted with ether. Ether layer was decanted, dried and concentrated under reduced pressure. The resulting product though seen as single compound by TLC, was further purified by passing over a column of silica gel. The recovered PEG can be reused.

Table 1
Strecker synthesis of α-amino nitriles from imines in the presence of PEG in water

Entry	R	R^1	R^2	Time (h)	Yield[a,b] (%)
1	C$_6$H$_5$	H	C$_6$H$_5$	3.0	94[c]
2	C$_6$H$_5$	H	4-F-C$_6$H$_4$	3.0	94
3	C$_6$H$_5$	H	C$_6$H$_5$-CH$_2$	5.0	88
4	4-Br-C$_6$H$_4$	H	C$_6$H$_5$	3.0	92
5	4-Cl-C$_6$H$_4$	H	4-F-C$_6$H$_4$	3.0	94
6	4-Me-C$_6$H$_4$	H	C$_6$H$_5$	3.0	90
7	4-Me-C$_6$H$_4$	H	Ts	3.0	90
8	2,4,6-Tri-Me-C$_6$H$_2$	H	C$_6$H$_5$	3.5	88
9	4-MeO-C$_6$H$_4$	H	C$_6$H$_5$	3.0	90
10	2,4-Di-MeO-C$_6$H$_3$	H	C$_6$H$_5$	3.5	86
11	4-Allyloxy-C$_6$H$_4$	H	C$_6$H$_5$	3.5	90
12	4-NO$_2$-C$_6$H$_4$	H	C$_6$H$_5$	5.0	84
13	C$_6$H$_5$-CH=CH	H	C$_6$H$_5$	5.0	82
14	2-Naphthyl	H	C$_6$H$_5$	3.0	90
15	C$_{10}$H$_{20}$	H	C$_6$H$_5$	5.0	85
16	C$_6$H$_5$	CH$_3$	C$_6$H$_5$	4.0	88
17	4-Me-C$_6$H$_4$	CH$_3$	C$_6$H$_5$	4.5	86
18	C$_6$H$_{10}$	–	C$_6$H$_5$	5.0	84

[a] All the products were characterised by ^{1}H NMR, IR, Mass Spectroscopy.
[b] Isolated yields after purification.
[c] PEG was recovered and reused for five consecutive runs.

3.1. 2-Anilino-2-phenylacetonitrile (Table 1; Entry 1)

White solid, mp 74–76 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.94 (d, 1H, J = 8.4 Hz), 5.40 (d, 1H, J = 8.4 Hz), 6.74 (d, 2H, J = 8.2 Hz), 6.87 (t, 1H, J = 7.5 Hz), 7.25 (t, 2H, J = 7.5 Hz), 7.40–7.50 (m, 3H), 7.57–7.65 (m, 2H); Mass m/z 208; IR (KBr): 3369, 2236 cm^{-1}.

3.2. 2-(4-Fluoroanilino)-2-phenylacetonitrile (Entry 2)

Yellow solid, mp 80–82 °C; ^{1}H NMR (CDCl$_3$, 300 MHz): δ 3.85 (d, 1H, J = 8.3 Hz), 5.31 (d, 1H, J = 8.3 Hz), 6.68–6.73 (m, 2H), 6.96 (t, 2H, J = 8.6 Hz), 7.14–7.48 (m, 3H), 7.59 (d, 2H, J = 7.5 Hz); Mass (EI) m/z 226 (M$^+$); IR (KBr): 3362, 2236 cm^{-1}; Anal. Calcd. for C$_{14}$H$_{11}$FN$_2$: C, 74.32; H, 4.90; N, 12.38. Found: C, 74.05; H, 4.84; N, 12.35.

3.3. 2-(Benzylamino)-2-phenylacetonitrile (Entry 3)

Colorless oil; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 1.80 (brs, NH), 3.95 (AB q, 2H, J = 13.5 Hz), 4.70 (s, 1H), 6.78 (d, 1H, J = 8.0 Hz), 7.15 (t, 1H, J = 7.8 Hz), 7.25–7.40 (m, 6H), 7.49–7.51 (m, 2H); Mass (EI) m/z 222 (M$^+$); IR (KBr): 3409, 2234 cm^{-1}.

3.4. 2-Anilino-2-(4-bromophenyl)acetonitrile (Entry 4)

Pale yellow solid, mp 86–88 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.96 (d, 1H, J = 7.6 Hz), 5.37 (d, 1H, J = 7.6 Hz), 6.72 (d, 2H, J = 8.4 Hz), 6.89 (t, 1H, J = 7.1 Hz), 7.26 (d,

2H, $J = 7.5$ Hz), 7.55 (dd, 4H, $J = 8.4$, 11.8 Hz); Mass (EI) m/z 287 (M$^+$); IR (KBr): 3326, 2233 cm^{-1}; Anal. Calcd. for C$_{14}$H$_{11}$BrN$_2$: C, 58.56; H, 3.86; N, 9.76. Found: C, 58.19; H, 3.80; N, 9.71.

3.5. 2-(4-Chlorophenyl)-2-(4-fluoroanilino)acetonitrile (Entry 5)

Pale yellow solid, mp 94–96 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.88 (d, 1H, $J = 8.2$ Hz), 5.32 (d, 1H, $J = 8.2$ Hz), 6.67–6.77 (m, 2H), 6.99 (t, 2H, $J = 8.5$ Hz), 7.52 (dd, 4H, $J = 8.6$, 14.7 Hz); Mass (EI) m/z 260 (M$^+$); IR (KBr): 3349, 2226 cm^{-1}; Anal. Calcd. for C$_{14}$H$_{10}$ClFN$_2$: C, 64.50; H, 3.87; N, 10.75. Found: C, 64.19; H, 3.74; N, 10.70.

3.6. 2-Anilino-2-(4-methylphenyl)acetonitrile (Entry 6)

White solid, mp 74–76 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 2.42 (s, 3H), 3.92 (d, 1H, $J = 7.8$ Hz), 5.35 (d, 1H, $J = 7.8$ Hz), 6.69–6.79 (m, 2H), 6.87 (t, 1H, $J = 7.3$ Hz), 7.21–7.30 (m, 4H), 7.49 (d, 2H, $J = 7.8$ Hz); Mass (EI) m/z 222 (M$^+$); IR (KBr): 3369, 2225 cm^{-1}.

3.7. N1-Cyano(4-methylphenyl)methyl-4-methyl-1-benzenesulfonamide (Entry 7)

White solid, mp 140–142 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 2.36 (s, 3H), 2.39 (s, 3H), 4.93 (d, 1H, $J = 7.8$ Hz), 5.42 (d, 1H, $J = 7.8$ Hz), 7.80 (d, 2H, $J = 8.6$ Hz), 7.23 (d, 3H, $J = 7.8$ Hz), 7.34 (d, 3H, $J = 7.8$ Hz); Mass (EI) m/z 300 (M$^+$); IR (KBr): 3269, 2236 cm^{-1}.

3.8. 2-Anilino-2-mesitylacetonitrile (Entry 8)

Colorless oil; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 2.31 (s, 3H), 2.47 (s, 6H), 3.72 (d, 1H, $J = 7.2$ Hz), 5.53 (d, 1H, $J = 7.2$ Hz), 6.76 (d, 3H, $J = 8.6$ Hz), 7.25 (t, 2H, $J = 7.6$ Hz), 6.91 (s, 2H); Mass (EI) m/z 250 (M$^+$); IR (KBr): 3332, 2226 cm^{-1}; Anal. Calcd. for C$_{17}$H$_{18}$N$_2$: C, 81.56; H, 7.25; N, 11.19. Found: C, 81.22; H, 7.18; N, 11.12.

3.9. 2-Anilino-2-(4-methoxyphenyl)acetonitrile (Entry 9)

White solid, mp 92–94 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.83 (s, 3H), 3.88 (d, 1H, $J = 7.6$ Hz), 5.32 (d, 1H, $J = 7.6$ Hz), 6.73 (d, 2H, $J = 8.4$ Hz), 6.93 (d, 2H, $J = 8.4$ Hz), 7.18–7.30 (m, 3H), 7.50 (d, 2H, $J = 8.4$ Hz); Mass (EI) m/z 238 (M$^+$); IR (KBr): 3337, 2225 cm^{-1}.

3.10. 2-Anilino-2-(2,4-dimethoxyphenyl)acetonitrile (Entry 10)

Pale yellow solid, mp 96–98 °C; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.83 (s, 3H), 3.88 (s, 3H), 4.20 (d, 1H, $J = 8.0$ Hz), 5.48 (d, 1H, $J = 8.0$ Hz), 6.73 (d, 2H, $J = 8.4$ Hz), 6.82–6.90 (m, 3H), 7.04 (s, 1H), 7.22 (t, 2H, $J = 8.0$ Hz); Mass (EI)

m/z 268 (M$^+$); IR (KBr): 3334, 2226 cm^{-1}; Anal. Calcd. for C$_{16}$H$_{16}$N$_2$O$_2$: C, 71.62; H, 6.01; N, 10.44. Found: C, 71.24; H, 5.92; N, 10.38.

3.11. 2-[4-(Allyloxy)phenyl]-2-anilinoacetonitrile (Entry 11)

Colorless oil; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.89 (d, 1H, $J = 7.6$ Hz), 4.53–4.64 (m, 2H), 5.26–5.39 (m, 2H), 5.45 (d, 1H, $J = 7.6$ Hz), 5.90–6.13 (m, 1H), 6.73 (d, 2H, $J = 8.4$ Hz), 6.89–7.01 (m, 3H), 7.18–7.30 (m, 2H), 7.49 (d, 2H, $J = 8.4$ Hz); Mass (EI) m/z 264 (M$^+$); IR (KBr): 3399, 2235 cm^{-1}; Anal. Calcd. for C$_{17}$H$_{16}$N$_2$O: C, 77.25; H, 6.10; N, 10.60. Found: C, 76.88; H, 6.02; N, 10.52.

3.12. 2-Anilino-2-(4-nitrophenyl)acetonitrile (Entry 12)

Viscous liquid; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 4.05 (d, 1H, $J = 8.0$ Hz), 5.35 (d, 1H, $J = 8.0$ Hz), 6.80 (d, 2H, $J = 8.0$ Hz), 6.90 (t, 1H, $J = 7.9$ Hz), 7.20 (t, 2H, $J = 7.9$ Hz), 7.67 (d, 2H, $J = 8.1$ Hz), 8.10 (d, 2H, $J = 8.1$ Hz); Mass (EI) m/z 253 (M$^+$); IR (KBr): 3421, 2235 cm^{-1}.

3.13. (E)-2-Anilino-4-phenyl-3-butenenitrile (Entry 13)

Pale yellow solid; ^{1}H NMR (CDCl$_3$, 200 MHz): δ 3.80 (d, 1H, $J = 8.1$ Hz), 5.05 (m, 1H), 6.30 (dd, 1H, $J = 6.9$, 17.3 Hz), 6.78 (d, 1H, $J = 8.0$ Hz), 6.90 (t, 1H, $J = 7.9$ Hz), 7.08 (dd, 1H, $J = 1.7$, 17.3 Hz), 7.25–7.45 (m, 8H); Mass (EI) m/z 234 (M$^+$); IR (KBr): 3334, 2226 cm^{-1}.

3.14. 2-Anilino-2-(2-naphthyl)acetonitrile (Entry 14)

Pale yellow solid, mp 94–96 °C; ^{1}H NMR (CDCl$_3$, 300 MHz): δ 3.89 (d, 1H, $J = 8.4$ Hz), 5.91 (d, 1H, $J = 8.4$ Hz), 6.73–6.84 (m, 2H), 7.02 (t, 2H, $J = 8.4$ Hz), 7.49–7.60 (m, 4H), 7.85–7.98 (m, 4H); Mass (EI) m/z 258 (M$^+$); IR (KBr): 3327, 2224 cm^{-1}; Anal. Calcd. for C$_{18}$H$_{14}$N$_2$: C, 83.69; H, 5.46; N, 10.84. Found: C, 83.26; H, 5.38; N, 10.78.

3.15. 2-Anilinoundecanenitrile (Entry 15)

Liquid; ^{1}H NMR (CDCl$_3$, 200 MHz): 0.90 (t, 3H, $J = 6.8$ Hz), 1.20–1.40 (m, 12H), 1.50–1.65 (m, 2H), 1.80–1.90 (m, 2H), 3.80 (brs, NH), 4.05–4.15 (m, 1H), 6.60 (d, 2H, $J = 8.0$ Hz), 6.80 (t, 1H, $J = 7.9$ Hz); 7.20 (t, 2H, $J = 7.9$ Hz); Mass (EI) m/z 258 (M$^+$); IR (KBr): 3405, 2235 cm^{-1}.

3.16. 2-Anilino-2-phenylpropanenitrile (Entry 16)

White solid, mp 136–138 °C; ^{1}H NMR (CDCl$_3$, 300 MHz): δ 1.93 (s, 3H), 4.18 (s, 1H), 6.48 (d, 2H, $J = 8.3$ Hz), 6.77 (t, 1H, $J = 7.9$ Hz), 7.08 (t, 2H, $J = 7.9$ Hz), 7.30–7.42 (m, 3H), 7.60 (d, 2H, $J = 7.2$ Hz); IR (KBr): 3385, 2225 cm^{-1}; Mass (EI) m/z 222 (M$^+$).

3.17. 2-Anilino-2-(4-methylphenyl)propanenitrile (Entry 17)

White solid, mp 126–128 °C; ^{1}H NMR (CDCl$_3$, 300 MHz): δ 1.93 (s, 3H), 2.36 (s, 3H), 4.25 (s, 1H), 6.43 (d, 2H, $J = 8.0$ Hz), 6.58 (t, 1H, $J = 7.4$ Hz), 7.02–7.10 (m, 4H), 7.49 (d, 2H, $J = 8.0$ Hz); IR (KBr): 3387, 2228 cm^{-1}; Mass: m/z 236 (M$^+$).

3.18. 1-Anilino-1-cyclohexanecarbonitrile (Entry 18)

White solid, mp 74–76 °C; IR (KBr): 3354, 2226 cm^{-1}; ^{1}H NMR (CDCl$_3$, 300 MHz): δ 1.56–1.84 (m, 8H), 2.32–2.36 (m, 2H), 3.65 (s, 1H), 6.62 (d, 2H, $J = 8.0$ Hz), 6.80 (t, 1H, $J = 7.9$ Hz); 7.20 (t, 2H, $J = 7.9$ Hz); IR (KBr): 3354, 2226 cm^{-1}; Mass: m/z 200 (M$^+$).

Acknowledgements

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