A Novel Synthesis of 2-Aminopyrroles Using a Three-Component Reaction

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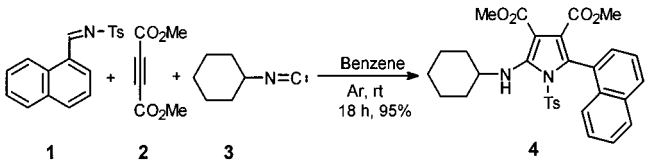
Multicomponent reactions (MCRs), by virtue of their convergence, productivity, facile execution, and generally high yields of products, have attracted much attention from the vantage point of combinatorial chemistry.[1,2] Of pivotal importance in this area are the isocyanide-based MCRs such as the versatile Ugi and Passerini reactions.[3,4]

In the context of our ongoing studies on heterocyclic construction mediated by 1,3 dipoles[5] and zwitterionic intermediates,[6] the possibility of trapping the 1:1 intermediate formed between dimethyl acetylenedicarboxylate (DMAD) and isocyanides with suitable dipolarophiles appeared attractive from the viewpoint of devising a novel MCR.[7]

Although the addition of isocyanides to DMAD itself has been studied in detail by a number of research groups,[8−14] attempts to trap the initially formed 1:1 intermediate with external dipolarophiles have failed.[11] Prompted by the recent success in developing a synthesis of aminofuran derivatives[15] by the reaction of aldehydes and the zwitterionic intermediate, we reasoned that the latter would react with *N*-tosylimines[16] to afford aminopyrrole derivatives. The results of our investigations attesting the rationale are presented here. It may be mentioned that there are only isolated reports available on 2-aminopyrrole synthesis.[17−20]

Initially, we carried out the reaction of *N*-tosyl naphthaldimine, DMAD, and cyclohexyl isocyanide [21] in dry benzene at room temperature; the condensation product **4** was obtained in excellent yield (Scheme 1).

Scheme 1



In the ^{1}H NMR spectrum, the two carbomethoxy groups were observed at δ 3.30 and 3.74 as two singlets and the NH proton resonated at δ 6.15, supporting the IR absorption at 3328 cm^{-1}. The ^{13}C signals for the two ester carbonyls were seen at δ 164.7 and 165.2.

The reaction was found to be general and efficient. Interestingly it was found that tosylimines from ortho substituted aldehydes gave stable, isolable products; the results are summarized in Table 1.

tert-Butyl isocyanide also elicited the same reactivity pattern, and the results are tabulated in Table 2.

Although we have tried the reaction with dibenzoyl acetylene, methyl propiolate, and tetracyanoethylene in place of DMAD, no stable product could be isolated from the reaction.

Mechanistically, the reaction may involve the initial generation of a zwitterionic intermediate from isocyanide and DMAD, which adds to the carbon−nitrogen double bond of the tosylimine to yield an intermediate iminolactam. Subsequently, it undergoes a [1,5] H shift to yield aminopyrroles, as illustrated in Scheme 2.

To explore the scope of this reaction further, we have extended it to quinoneimines **6**, in which case we could isolate corresponding iminolactams **7** (Scheme 3).

To confirm the structure of the adducts, we carried out detosylation of **4a** using TBAF in THF at room temperature to afford the aminopyrrole **8** in 71% yield (Scheme 4).

In conclusion, we have uncovered a novel and efficient multicomponent reaction of *N*-tosylimines, DMAD, and isocyanides for the synthesis of 2-aminopyrrole systems. It is interesting to note that 2-aminopyrroles have found use as synthetic precursors for acyclic nucleoside analogues of the pyrrolo[2,3-*d*]pyrimidine ring system.[22]

Experimental Section

General. Melting points were recorded on a Büchi melting point apparatus and are uncorrected. NMR spectra were recorded at 300 (^{1}H) and 75 (^{13}C) MHz on a Bruker dpx 300

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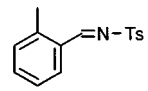
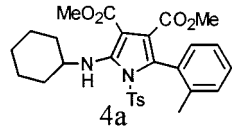
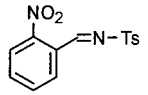
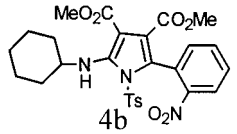
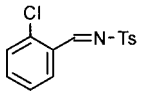
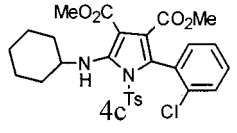
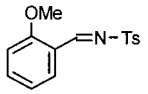
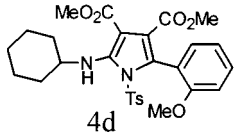
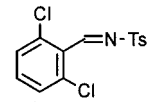
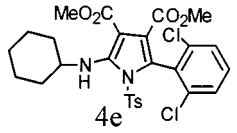
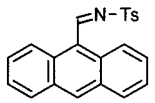
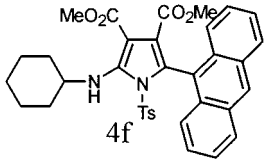
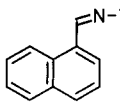
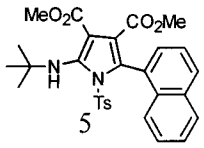
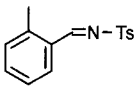
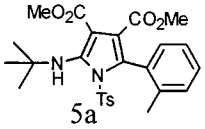
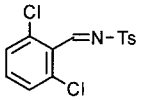
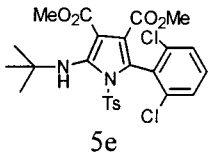
Table 1. Reaction of Tosylimines with DMAD and Cyclohexyl Isocyanide

Entry	Tosylimines	Conditions	Product	Yield(%)[a]
1	 1a	Benzene Ar, rt, 22 h	 4a	92%
2	 1b	Benzene Ar, rt, 20 h	 4b	87%
3	 1c	Benzene Ar, rt,16 h	 4c	82%
4	 1d	Benzene Ar, rt, 15 h	 4d	79%
5	 1e	Benzene Ar, rt, 21 h	 4e	94%
6	 1f	Benzene Ar, Reflux 2 h	 4f	93%

[a] Isolated yield.

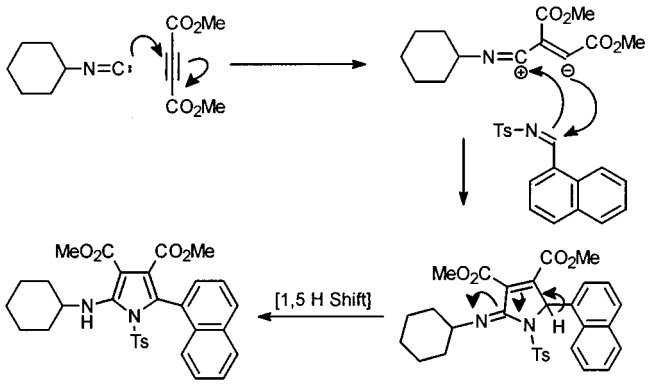
Table 2. Reaction of Tosylimines with DMAD and *tert*-Butyl Isocyanide

Entry	Tosylimines	Conditions	Product	Yield(%)[a]
1	 1	Benzene Ar, rt, 19 h	 5	72%
2	 1a	Benzene Ar, rt, 14 h	 5a	96%
3	 1e	Benzene Ar, rt,15 h	 5e	93%

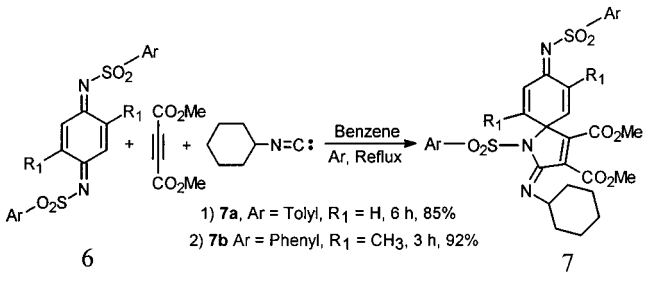
[a] Isolated yield.

spectrometer. The spectra were run in $CDCl_3-CCl_4$, v/v 3:1, and chemical shifts are reported (δ) relative to TMS (^1H) and $CDCl_3$ (^{13}C) as the internal standards. Mass spectra were recorded under EI/HRMS (at 5000) resolution using an Auto Spec. M mass spectrometer. IR spectra were recorded on a Nicolet Impact 400D FT-IR spectrophotometer. Elemental analyses were obtained on

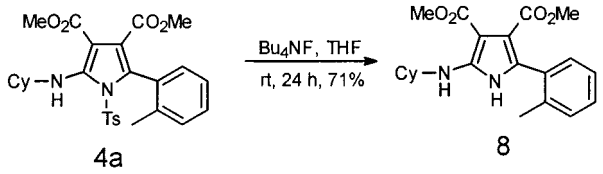
Scheme 2



Scheme 3



Scheme 4



a Perkin-Elmer-2400 elemental analyzer. Dimethyl acetylenedicarboxylate was purchased from Aldrich Chemical Co. and was used without further purification. Cyclohexyl isocyanide was prepared by a reported procedure.[21]

General Experimental Procedure for the Preparation of 2-Aminopyrroles. A mixture of *N*-tosylimine (1.0 mmol) and DMAD (1.1 mmol) in dry benzene (10 mL) was purged with argon for 5 min. To this mixture was added cyclohexyl isocyanide (120 mg, 1.1 mmol) via a syringe, and the reaction mixture was stirred at room temperature. After the reaction was complete as monitored by TLC, the solvent was removed under vacuum, and the product was purified by crystallization (CH_2Cl_2-hexane mixture) or by column chromatography wherever crystallization is not possible.

Aminopyrrole 4. Mp 149−150 °C; IR (KBr) ν_{max} 3328, 2927, 2850, 1734, 1681, 1575, 1449, 1377, 1222 cm^{-1}; ^1H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.25−2.16, (m, 10H), 2.32 (s, 3H), 3.30 (s, 3H), 3.74 (s, 3H), 3.80 (m, 1H), 6.15 (br s, 1H), 7.03 (d, J = 7.9, 2H), 7.25−7.52 (m, 7H), 7.79−7.85 (m, 2H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.6, 24.6, 25.1, 25.7, 34.0, 51.4, 51.8, 59.4, 99.8, 121.4, 124.4, 125.5, 126.1, 127.5, 128.1, 128.7, 129.3, 133.0, 133.1, 134.9, 145.2, 150.2, 164.7, 165.2. Anal. Calcd for $C_{31}H_{32}N_2O_6S$: C, 66.41; H, 5.75; N, 4.99; S, 5.71. Found: C, 66.03; H, 5.88; N, 5.18; S, 5.77.

Aminopyrrole 4a. Mp 135−136 °C; IR (KBr) ν_{max} 3324, 2930, 2859, 1732, 1655, 1596, 1495, 1442, 1360, 1295 cm^{-1}; ^1H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.15−1.93 (m, 10H), 2.21 (s, 3H) 2.40 (s, 3H), 3.48 (s, 3H), 3.72 (s, 3H), 3.76 (m, 1H), 5.99 (br s, 1H), 6.82 (d, J = 7.0, 1H), 7.04−7.28 (m, 5H), 7.44 (d, J = 8.3, 2H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 19.0, 21.5, 24.4, 24.5, 25.5, 34.8, 52.5, 52.9, 58.9, 107.8, 126.0, 127.5, 128.1, 128.2, 128.9, 129.4, 131.0, 131.6, 133.7, 136.0, 137.8, 143.4, 145.4, 161.0, 164.3. Anal. Calcd for $C_{28}H_{32}N_2O_6S$: C, 64.10; H, 6.14; N, 5.33; S, 6.11. Found: C, 64.21; H, 6.15; N, 5.66; S, 6.48.

Aminopyrrole 4b. Mp 76−77 °C; IR (KBr) ν_{max} 3386, 2930, 2856, 1736, 1689, 1563, 1530, 1450, 1384, 1344, 1230 cm^{-1}; ^1H

NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.12−2.16 (m, 10H), 2.41 (s, 3H), 3.52 (s, 3H), 3.73 (s, 4H), 5.82 (d, J = 9.2, 1H), 7.13−7.26 (m, 3H), 7.46−7.56 (m, 4H), 8.14−8.17 (m, 1H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.6, 24.9, 25.0, 25.6, 33.6, 51.3, 51.9, 59.6, 100.8, 124.7, 127.4, 129.3, 129.5, 132.0, 132.4, 134.8, 145.5, 148.6, 150.0, 164.4, 164.5; HRMS (EI) for $C_{27}H_{29}N_3O_8S$, calcd 555.1675, found 555.1655.

Aminopyrrole 4c. Mp 106−107 °C; IR (KBr) ν_{max} 3295, 2929, 2852, 1736, 1689, 1571, 1453, 1382, 1234, 1181 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.11−2.15 (m, 10H), 2.39 (s, 3H), 3.55 (s, 3H), 3.72 (s, 4H), 5.84 (d, J = 8.7, 1H), 7.20−7.55 (m, 8H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.7, 25.0, 25.1, 25.7, 33.9, 51.5, 52.0, 59.7, 100.6, 121.2, 125.8, 126.8, 127.6, 129.3, 129.5, 129.8, 130.6, 132.0, 134.8, 135.1, 145.5, 149.9, 164.6, 164.9. Anal. Calcd for $C_{27}H_{29}N_2O_6SCl$: C, 59.49; H, 5.36; N, 5.13; S, 5.88. Found: C, 59.76; H, 5.45; N, 5.18; S, 6.29.

Aminopyrrole 4d. Mp 73−74 °C; IR (KBr) ν_{max} 3321, 2932, 2852, 1747, 1662, 1610, 1506, 1453 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.16−2.15 (m, 10H), 2.37 (s, 3H), 3.54 (s, 3H), 3.70 (s, 3H), 3.73 (s, 3H), 3.85 (m, 1H), 5.92 (br s, 1H), 6.81−7.44 (m, 8H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.6, 25.1, 25.8, 33.8, 34.0, 51.2, 51.7, 55.2, 59.4, 100.1, 110.2, 119.6, 120.2, 120.4, 126.9, 127.5, 129.2, 130.0, 131.4, 135.1, 144.9, 150.3, 157.8, 164.7, 165.4.

Aminopyrrole 4e. Mp 170−171 °C; IR (KBr) ν_{max} 3289, 2927, 2853, 1738, 1689, 1564, 1446, 1377, 1228 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.06−2.03 (m, 10H), 2.41 (s, 3H), 3.58 (s, 3H), 3.65 (m, 1H), 3.74 (s, 3H) 5.38 (br s, 1H), 7.22−7.37 (m, 5H), 7.76 (d, J = 8.0, 2H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.7, 25.1, 25.7, 33.7, 51.5, 51.9, 59.1, 102.1, 120.0, 125.2, 127.6, 128.3, 129.4, 130.1, 130.5, 135.3, 136.6, 145.5, 147.6, 163.9, 164.7. Anal. Calcd for $C_{27}H_{28}N_2O_6SCl_2$: C, 55.96; H, 4.86; N, 4.83. Found: C, 56.10; H, 4.89; N, 4.58.

Aminopyrrole 4f. Mp 169−170 °C; IR (KBr) ν_{max} 3319, 3054, 2930, 2853, 1726, 1678, 1566, 1454, 1383, 1318, 1224 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.23−2.23 (m, 10H), 2.28 (s, 3H), 3.09 (s, 3H), 3.79 (s, 3H), 3.85 (m, 1H), 6.15 (br s, 1H), 6.84 (d, J = 7.9, 2H), 7.04 (d, J = 8.2, 2H), 7.20−7.50 (m, 6H), 7.95 (d, J = 8.3, 2H), 8.46 (s, 1H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.5, 25.2, 25.8, 34.0, 51.5, 52.1, 58.5, 99.8, 121.7, 124.3, 124.9, 125.3, 125.8, 126.0, 126.3, 127.7, 128.3, 128.8, 129.1, 130.9, 132.4, 134.9, 145.1, 148.4, 164.7, 164.8. Anal. Calcd for $C_{35}H_{34}N_2O_6S$: C, 68.83; H, 5.61; N, 4.58. Found: C, 68.84; H, 5.68; N, 4.55.

Aminopyrrole 5. Mp 129−130 °C; IR (KBr) ν_{max} 3344, 2952, 1739, 1695, 1558, 1446, 1384, 1210 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.41 (s, 9H), 2.30 (s, 3H), 3.28 (s, 3H), 3.77 (s, 3H), 5.06 (br s, 1H), 7.00 (d, J = 8.2, 2H), 7.24−7.48 (m, 7H), 7.79−7.86 (m, 2H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.5, 30.2, 51.5, 51.7, 58.1, 108.4, 120.9, 124.4, 125.2, 125.5, 126.1, 127.4, 128.1, 128.3, 129.2, 129.2, 129.4, 130.5, 132.9, 133.0, 135.3, 145.0, 146.8, 164.5, 164.8. Anal. Calcd for $C_{29}H_{30}N_2O_6S$: C, 65.15; H, 5.65; N, 5.23. Found: C, 65.18; H, 5.68; N, 4.99.

Aminopyrrole 5a. Mp 122−123 °C; IR (KBr) ν_{max} 3334, 2957, 1756, 1707, 1585, 1435, 1330, 1291 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.34 (s, 9H), 2.23 (s, 3H), 2.39 (s, 3H), 3.48 (s, 3H), 3.74 (s, 3H), 6.74 (d, J = 7.3, 1H), 7.03−7.39 (m, 7H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 20.3, 21.6, 30.1, 51.4, 51.7, 57.9, 108.0, 120.0, 124.6, 127.3, 128.8, 129.3, 129.6, 129.8, 130.7, 131.8, 135.6, 138.9, 145.0, 146.7, 164.3, 164.8. Anal. Calcd for $C_{26}H_{30}N_2O_6S$: C, 62.33; H, 6.06; N, 5.61; S, 6.43. Found: C, 62.11; H, 6.28; N, 5.67; S, 6.14.

Aminopyrrole 5e. Mp 172−173 °C; IR (KBr) ν_{max} 3344, 2959, 1732, 1707, 1539, 1452, 1371, 1178 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.26 (s, 9H), 2.40 (s, 3H), 3.58 (s, 3H), 3.76 (s, 3H), 7.20−7.38 (m, 5H), 7.73 (d, J = 8.1, 2H,); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.7, 30.0, 51.7, 51.8, 57.7, 111.2, 119.2, 127.7, 128.0, 128.4, 129.3, 130.0, 130.7, 136.1, 143.8, 145.2, 163.4, 164.5. Anal. Calcd for $C_{25}H_{26}N_2O_6SCl_2$: C, 54.25; H, 4.73; N, 5.06. Found: C, 54.14; H, 4.73; N, 4.76.

Iminolactam 7a. Mp 156−157 °C; IR (KBr) ν_{max} 2927, 2840, 1738, 1669, 1619, 1559 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.11−1.86 (m, 10H), 2.44 (s, 6H), 3.18 (m, 1H), 3.67 (s, 3H), 3.85 (s, 3H), 6.52 (d, J = 8.9, 1H), 6.64 (d, J = 9.7, 2H), 7.25−7.34 (m, 4H), 7.81 (d, J = 9.2, 1H), 7.88−7.93 (m, 4H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 21.5, 21.6, 24.4, 25.3, 34.5, 52.5, 53.1, 59.5, 66.4, 76.4, 124.9, 127.2, 128.7, 129.4, 129.5, 131.2, 135.0, 135.8, 141.9,142.5, 143.3, 143.6, 144.5, 159.8, 163.2, 163.5. Anal. Calcd for $C_{33}H_{35}N_3O_8S_2$: C, 59.53; H, 5.30; N, 6.31. Found: C, 59.28; H, 5.38; N, 5.98.

Iminolactam 7b. Mp 165−166 °C; IR (KBr) ν_{max} 2934, 2846, 1738, 1663, 1625, 1550, 1444 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.15−1.81 (m, 10H), 1.95 (s, 3H), 1.99 (s, 3H), 3.17 (m, 1H), 3.62 (s, 3H), 3.82 (s, 3H), 6.20 (s, 1H), 7.41−7.56 (m, 7H), 7.98−8.00 (m, 4H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 17.1, 18.7, 24.5, 25.3, 34.7, 34.7, 53.0, 53.1, 59.6, 69.2, 123.5, 126.9, 128.0, 128.6, 129.5, 132.4, 133.6, 134.6, 137.8, 138.8, 140.1, 141.8, 143.8, 153.2, 159.8, 163.5, 165.3.

Detosylation of 4a with Bu_4NF. **Procedure for the Preparation of *N*-Unsubstituted Aminopyrrole 8.** To a solution of **4a** (100 mg, 0.19 mmol) in THF (3 mL) was added a solution of Bu_4NF (1 M, 0.17 mL, 3 equiv) in THF. The mixture was then stirred at room temperature for 24 h. Methanol was added, and the mixture was concentrated under reduced pressure. The residue was dissolved in ethyl acetate (10 mL) and washed with saturated $NaHCO_3$, and the aqueous layer was extracted twice with ethyl acetate. The combined organic extract was washed with brine, dried over anhydrous Na_2SO_4, and concentrated. The residue was purified by column chromatography on silica gel to afford **8** as a white solid (51 mg, 71%): mp 165−166 °C; IR (KBr) ν_{max} 3355, 3218, 2930, 2850, 1657, 1595, 1557 cm^{-1}; ^{1}H NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 1.30−1.95 (m, 10H), 2.26 (s, 3H), 3.13 (m, 1H), 3.61 (s, 3H), 3.76 (s, 3H), 6.51 (br s, 1H), 7.20−7.27 (m, 4H), 7.50 (s, 1H); ^{13}C NMR ($CDCl_3-CCl_4$, v/v, 3:1) δ 19.5, 23.9, 24.9, 32.9, 49.6, 50.4, 50.6, 89.1, 111.0, 124.4, 126.6, 127.4, 129.1, 130.3, 131.6, 137.4, 147.3, 165.4, 165.5.

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Supporting Information Available: Copies of spectra (^{1}H, ^{13}C, and IR) for compounds **4**, **4a**−**c**, **4e**, **4f**, **5**, **5a**, **5e**, **7b,** and **8**. This material is available free of charge via the Internet at http://pubs.acs.org.

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