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By the coupling reactions of trimethylsilylacetylene and 2-chloro-3,6-diisobutylpyrazine, 3,6-diisobutyl-2-trimethylsilylethynylpyrazine or 1,2-bis(3,6-diisobutylpyrazin-2-yl)acetylene was obtained, depending on the solvent used. The former substance coupled with various aryl and heteroaryl halides to give 1-aryl-2-pyrazinylacetylenes.

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In order to introduce an ethynyl group into the aromatic rings, aryl halides were submitted to the coupling reaction with trimethylsilylacetylene in the presence of the palladium catalysts, and the products were hydrolysed subsequently in an alkaline medium [1]. The resulting ethynyl compounds are very useful as a building block for syntheses of indole, benzofuran and benzothiophene derivatives [2]. We have recently reported the coupling reactions of chloropyrazines with acetylenes using palladium catalysts [3]. In relation to this work, we carried out the coupling reaction of 2-chloro-3,6-diisobutylpyrazine (**1**) with trimethylsilylacetylene in the presence of tetrakis(triphenylphosphine)palladium. When *N,N*-dimethylformamide was used as a solvent, 1,2-bis(3,6-diisobutylpyrazin-2-yl)acetylene (**2**) was obtained. On the other hand, the reaction in triethylamine gave interestingly 3,6-diisobutyl-2-trimethylsilylethynylpyrazine (**3**). The latter product was further submitted to the coupling reaction with some aryl and heteroaryl halides to afford 1-aryl-2-pyrazinylacetylenes. Such a 1,2-differently substituted acetylene was recently produced by Arcadi, *et al.* as a by-product in the palladium-catalysed reductive addition reactions of 2-acetamidophenyltrimethylsilylacetylene with phenyl iodide [4]. In this report, the coupling reactions of aryl and heteroaryl

halides with **3** prepared from **1** and trimethylsilylacetylene will be described.

The reaction of **1** with trimethylsilylacetylene was carried out in the presence of tetrakis(triphenylphosphine)palladium by heating at 100° for 2 hours in *N,N*-dimethylformamide to give colorless crystals of **2**. In the infrared spectrum of **2**, no absorption band was observed at 2200 cm⁻¹, and ¹H-nmr spectrum of **2** showed only the presence of an isobutyl group and a pyrazine ring. On the basis of these spectral data, the structure of **2** was established as 1,2-bis(3,6-diisobutylpyrazin-2-yl)acetylene. When triethylamine was used as a solvent for this reaction in place of *N,N*-dimethylformamide, a colorless oil **3** was obtained. The ¹H-nmr spectrum of **3** showed a singlet at 0.28 ppm due to the methyl group of the trimethylsilyl moiety. The spectral and analytical data of **3** were consistent with 3,6-diisobutyl-2-trimethylsilylethynylpyrazine. By means of copper(I) iodide/bis(triphenylphosphine)palladium dichloride as a catalyst, the reaction of **1** with trimethylsilylacetylene in triethylamine did not proceed and the starting material was recovered.

Next, the reaction of **3** was studied. The reaction of **3** with 4-bromonitrobenzene in the presence of tetrakis(triphenylphosphine)palladium in *N,N*-dimethylformamide at

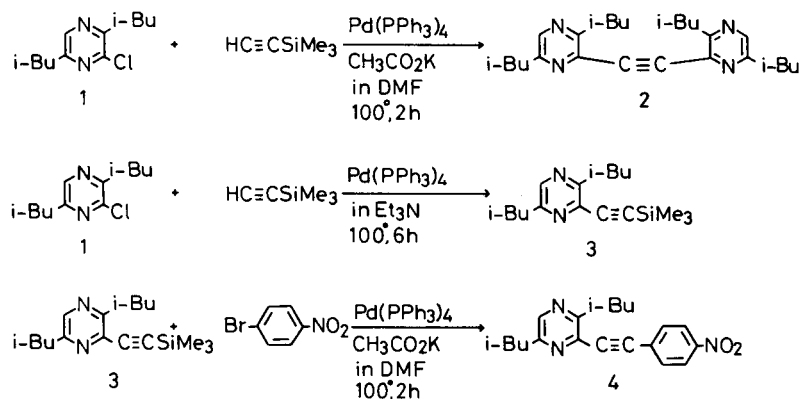
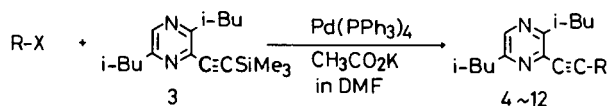


Chart 1

Table I

Reactions of Aryl and Heteroaryl Halides with
3,6-Diisobutyl-2-trimethylsilylethynylpyrazine



Starting material R-X	Product	Yield (%) [a]
4-Bromonitrobenzene	4	94 (68)
2-Bromonitrobenzene	5	64 (41)
4-Bromoacetophenone	6	88 (27)
4-Bromobenzoic acid methyl ester	7	96 (43)
2-Bromopyridine	8	88 (27)
2-Chloroquinoline	9	96 (47)
2-Chloropyrazine	10	96 (20)
2-Chloro-3,6-dimethylpyrazine	11	66 (37)
2-Chloro-5,6-diphenylpyrazine	12	66 (40)

[a] Yields in the one-pot reaction are in parentheses.

100° for 2 hours gave 1-(4-nitrophenyl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (**4**) in a 94% yield. Various aryl and heteroaryl halides were also submitted to the reaction with **3**. These results are summarized in Table I.

In order to simplify the procedure, we tried to develop a one-pot synthesis of **4**. After **1** was heated with trimethylsilylacetylene and tetrakis(triphenylphosphine)palladium in triethylamine in a sealed tube at 100° for 6 hours, the solvent was evaporated. To the resulting oil, 4-bromonitrobenzene, potassium acetate, tetrakis(triphenylphosphine)palladium and *N,N*-dimethylformamide were added, and then the reaction mixture was heated at 100° for 2 hours to give **4** in a 68% yield. The yields in some cases are also shown in Table I.

Consequently, unsymmetrical 1-aryl-2-pyrazinylacetylenes were easily obtained in satisfactory yields from a trimethylsilylethynylpyrazine by coupling reactions with aryl halides.

EXPERIMENTAL

All melting and boiling points are uncorrected. Melting points were recorded on a Yanagimoto micro-melting point apparatus. All uv spectra were taken in 95% ethanol using a Hitachi 557 spectrophotometer, ir spectra on a Shimadzu IR-400 spectrometer, and ¹H-nmr spectra in deuteriochloroform using a Varian EM-390 instrument with tetramethylsilane as an internal standard. Mass spectra were obtained with a Hitachi M-80 spectrometer. All products were purified by medium pressure column chromatography with a UVILOG ALPC-100 as a pump, UVILOG 5 IIIa (Oyo-Bunko Kiki Co., Ltd., Tokyo) as a uv detector and Kieselgel 60 (Merck AG, Darmstadt) as a packing material. All experiments were carried out in an argon atmosphere.

1,2-Bis(3,6-diisobutylpyrazin-2-yl)acetylene (**2**).

A mixture of 2-chloro-3,6-diisobutylpyrazine (**1**) (452 mg, 2 mmoles), trimethylsilylacetylene (196 mg, 2 mmoles), potassium acetate (294 mg, 3 mmoles) and tetrakis(triphenylphosphine)palladium (116 mg, 0.1 mmole) in *N,N*-dimethylformamide (5 ml) was heated at 100° for 2 hours. After the solvent was evaporated *in vacuo*, the residue was triturated with water (20 ml) and extracted with ether (10 ml x 3). The ethereal extract was dried over sodium sulfate and condensed to give a crude product, which was purified by medium pressure column chromatography and eluted with a mixture of hexane-ethyl acetate (20-1) to yield **2** (341 mg, 84%). Colorless prisms, mp 78.5-79.5° (methanol-water); uv: λ max 228 (log ε = 4.15, shoulder), 260.5 (4.00), 322 (4.39), 330 (4.38) nm; ¹H-nmr: δ 0.97 (12H, d, J = 7 Hz, 2 x CH₂CH(CH₃)₂), 1.00 (12H, d, J = 7 Hz, 2 x CH₂CH(CH₃)₂), 2.27 (4H, m, J = 7 Hz, 4 x CH₂CH(CH₃)₂), 2.72 (4H, d, J = 7 Hz, 2 x CH₂CH(CH₃)₂), 3.03 (4H, d, J = 7 Hz, 2 x CH₂CH(CH₃)₂), 8.40 (2H, s, pyrazine H) ppm; ms: m/z 406 (M⁺).

Anal. Calcd. for C₂₆H₃₆N₄: C, 76.80; H, 9.42; N, 13.78. Found: C, 76.89; H, 9.57; N, 13.80.

3,6-Diisobutyl-2-trimethylsilylethynylpyrazine (**3**).

After a mixture of **1** (2.26 g, 10 mmoles), trimethylsilylacetylene (1.08 g, 11 mmoles) and tetrakis(triphenylphosphine)palladium (0.58 g, 0.5 mmole) in triethylamine (30 ml) in a sealed tube was heated at 100° for 6 hours, the solvent was removed by distillation *in vacuo*. The residue was triturated with water (50 ml) and extracted with methylene chloride (20 ml x 3) to give **3** (2.69 g, 93%). Colorless oil, bp 153-156°/torr; uv: λ max 219 (log ε = 4.01), 237.5 (4.17), 246 (4.20), 299.5 (4.28), 320 (3.94, shoulder) nm; ir (film): 2160 (C≡C) cm⁻¹; ¹H-nmr: δ 0.28 (9H, s, Si(CH₃)₃), 0.95 (6H, d, J = 7 Hz, CH₂CH(CH₃)₂), 0.97 (6H, d, J = 7 Hz, CH₂CH(CH₃)₂), 2.22 (2H, m, 2 x CH₂CH(CH₃)₂), 2.63 (2H, d, J = 7 Hz, CH₂CH(CH₃)₂), 2.87 (2H, d, J = 7 Hz, CH₂CH(CH₃)₂), 8.28 (1H, s, pyrazine H) ppm; ms: m/z 288 (M⁺).

Anal. Calcd. for C₁₇H₂₈N₂Si: C, 70.78; H, 9.78; N, 9.71. Found: C, 70.48; H, 9.76; N, 9.61.

General Procedure for the Reaction of Aryl and Heteroaryl Halides with 3,6-Diisobutyl-2-trimethylsilylethynylpyrazine (**3**).

After a mixture of an aryl or heteroaryl halide (1.1 mmoles), **3** (288 mg, 1 mmole), potassium acetate (147 mg, 1.5 mmoles) and tetrakis(triphenylphosphine)palladium (58 mg, 0.05 mmole) in *N,N*-dimethylformamide (5 ml) was heated at 100° for 2 hours, the solvent was evaporated *in vacuo*. The residue was triturated with water (15 ml) and extracted with methylene chloride (10 ml x 3). The methylene chloride extract was usually worked up to give crude products, which were purified by medium pressure column chromatography and eluted with a mixture of hexane-ethyl acetate.

1-(4-Nitrophenyl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (**4**).

This compound had the following physical properties: pale yellow prisms, mp 82-83.5° (hexane); uv: λ max 327.5 (log ε = 4.11), 334 (4.10, shoulder) nm; ir (potassium bromide): 1350, 1525 (NO₂) cm⁻¹; ¹H-nmr: δ 0.97 (6H, d, J = 7 Hz, CH₂CH(CH₃)₂), 1.00 (6H, d, J = 7 Hz, CH₂CH(CH₃)₂), 2.20 (2H, m, J = 7 Hz, 2 x CH₂CH(CH₃)₂), 2.67 (2H, d, J = 7 Hz, CH₂CH(CH₃)₂), 2.90 (2H, d, J = 7 Hz, CH₂CH(CH₃)₂), 7.72 (2H, d, J = 10 Hz, benzene H), 8.22 (2H, d, J = 10 Hz, benzene H), 8.28 (1H, s, pyrazine H) ppm; ms: m/z 337 (M⁺).

Anal. Calcd. for C₂₀H₂₈N₂O₂: C, 71.19; H, 6.87; N, 12.45. Found: C, 71.46; H, 7.04; N, 12.31.

1-(2-Nitrophenyl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (**5**).

This compound had the following physical properties: pale yellow prisms, mp 56-57.5° (methanol-water); uv: λ max 227 (log ε = 4.08), 254.5 (4.17), 270 (3.91, shoulder), 306 (4.04), 332 (4.04) nm; ir (potassium bromide): 1350, 1530 (NO₂) cm⁻¹; ¹H-nmr: δ 0.93 (6H, d, J = 6.5 Hz, CH₂CH(CH₃)₂), 0.97 (6H, d, J = 6.5 Hz, CH₂CH(CH₃)₂), 2.17 (2H, m, 2 x

$\text{CH}_2\text{CH}(\text{CH}_3)_2$, 2.65 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.97 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 7.33-7.73 (2H, m, benzene H), 7.82 (1H, m, benzene H), 8.08 (1H, m, benzene H), 8.25 (1H, s, pyrazine H) ppm; ms: m/z 337 (M^+).

Anal. Calcd. for $\text{C}_{20}\text{H}_{23}\text{N}_3\text{O}_2$; C, 71.19; H, 6.87; N, 12.45. Found: C, 71.47; H, 6.90; N, 12.52.

1-(4-Acetylphenyl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (6).

This compound had the following physical properties: pale yellow prisms, mp 58-60° (methanol-water); uv: λ max 220 (log $\epsilon = 4.11$, shoulder), 237.5 (3.88, shoulder), 284 (4.16), 320.5 (4.33), 328.5 (4.33) nm; ir (potassium bromide): 1700 (COCH_3) cm^{-1} ; $^1\text{H-nmr}$: δ 0.90 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 0.97 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.17 (2H, m, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.56 (3H, s, COCH_3), 2.63 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.88 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 7.67 (2H, d, $J = 9$ Hz, benzene H), 7.97 (2H, d, $J = 9$ Hz, benzene H), 8.30 (1H, s, pyrazine H) ppm; ms: m/z 334 (M^+).

Anal. Calcd. for $\text{C}_{22}\text{H}_{26}\text{N}_2\text{O}$; C, 79.00; H, 7.84; N, 8.38. Found: C, 78.84; H, 7.59; N, 8.28.

1-(4-Methoxycarbonylphenyl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (7).

This compound had the following physical properties: colorless needles, mp 80-81° (hexane); uv: λ max 234.5 (log $\epsilon = 3.95$, shoulder), 265 (3.99, shoulder), 282 (4.09), 322 (4.31) nm; ir (potassium bromide): 1730 (CO_2CH_3) cm^{-1} ; $^1\text{H-nmr}$: δ 0.85 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 0.92 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.18 (2H, m, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.60 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.87 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 3.87 (3H, s, CO_2CH_3), 7.62 (2H, d, $J = 9$ Hz, benzene H), 8.03 (2H, d, $J = 9$ Hz, benzene H), 8.27 (1H, s, pyrazine H) ppm; ms: m/z 350 (M^+).

Anal. Calcd. for $\text{C}_{25}\text{H}_{26}\text{N}_2\text{O}_2$; C, 75.40; H, 7.48; N, 7.99. Found: C, 75.61; H, 7.48; N, 7.94.

1-(Pyridin-2-yl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (8).

This compound had the following physical properties: colorless prisms, mp 57-58° (methanol-water); uv: λ max 258.5 (log $\epsilon = 4.05$), 278 (4.02), 286 (4.08), 312.5 (4.34) nm; $^1\text{H-nmr}$: δ 0.97 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 1.03 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.23 (2H, m, $J = 7$ Hz, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.70 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.98 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 7.28 (1H, m, pyridine H), 7.65 (2H, m, pyridine H), 8.30 (1H, s, pyrazine H), 8.67 (1H, m, pyridine H) ppm; ms: m/z 293 (M^+).

Anal. Calcd. for $\text{C}_{19}\text{H}_{23}\text{N}_5$; C, 77.77; H, 7.90; N, 14.32. Found: C, 77.67; H, 7.94; N, 14.14.

1-(Quinolin-2-yl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (9).

This compound had the following physical properties: colorless prisms, mp 62-62.5° (methanol-water); uv: λ max 241 (log $\epsilon = 4.38$), 247 (4.39), 263.5 (4.44), 298 (4.23, shoulder), 324 (4.40), 334.5 (4.45), 347 (4.41) nm; $^1\text{H-nmr}$: δ 0.88 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 0.95 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.18 (2H, m, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.62 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.95 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 7.43-7.87 (4H, m, quinoline H), 8.13 (1H, d, $J = 9$ Hz, quinoline H), 8.15 (1H, d, $J = 9$ Hz, quinoline H), 8.30 (1H, s, pyrazine H) ppm; ms: m/z 343 (M^+).

Anal. Calcd. for $\text{C}_{23}\text{H}_{25}\text{N}_5$; C, 80.43; H, 7.34; N, 12.24. Found: C, 80.53; H, 7.31; N, 12.27.

1-(Pyrazin-2-yl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (10).

This compound had the following physical properties: colorless prisms, mp 43.5-44.5° (methanol-water); uv: λ max 225 (log $\epsilon = 4.10$, shoulder), 257.5 (4.01), 315 (4.35), 322.5 (4.34, shoulder) nm; $^1\text{H-nmr}$: δ 0.97 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 1.00 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.22 (2H, m, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.68 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.95 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 8.30 (1H, s, pyrazine H), 8.53 (1H, d, $J = 2$ Hz, pyrazine H), 8.60 (1H, dd, $J = 2$ Hz and 1 Hz, pyrazine H), 8.80 (1H, d, $J = 1$ Hz, pyrazine

H) ppm; ms: m/z 294 (M^+).

Anal. Calcd. for $\text{C}_{18}\text{H}_{22}\text{N}_4$; C, 73.43; H, 7.53; N, 19.03. Found: C, 73.28; H, 7.58; N, 18.79.

1-(3,6-Dimethylpyrazin-2-yl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (11).

This compound had the following physical properties: colorless prisms, mp 84-85.5° (methanol-water); uv: λ max 227 (log $\epsilon = 4.23$, shoulder), 259 (4.08), 322 (4.50), 330 (4.50) nm; $^1\text{H-nmr}$: δ 0.93 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 0.98 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.17 (2H, m, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.53 (3H, s, CH_3), 2.65 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.73 (3H, s, CH_3), 2.93 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 8.27 (2H, s, pyrazine H) ppm; ms: m/z 322 (M^+).

Anal. Calcd. for $\text{C}_{20}\text{H}_{26}\text{N}_4$; C, 74.49; H, 8.13; N, 17.38. Found: C, 74.25; H, 8.09; N, 17.33.

1-(5,6-Diphenylpyrazin-2-yl)-2-(3,6-diisobutylpyrazin-2-yl)acetylene (12).

This compound had the following physical properties: colorless prisms, mp 110.5-111.5° (hexane); uv: λ max 235 (log $\epsilon = 4.21$, shoulder), 308-311 (4.20), 348.5 (4.41) nm; $^1\text{H-nmr}$: δ 0.92 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 0.98 (6H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.18 (2H, m, $J = 7$ Hz, 2 x $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.63 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 2.93 (2H, d, $J = 7$ Hz, $\text{CH}_2\text{CH}(\text{CH}_3)_2$), 7.17-7.53 (10H, m, benzene H), 8.27 (1H, s, pyrazine H), 8.77 (1H, s, pyrazine H) ppm; ms: m/z 446 (M^+).

Anal. Calcd. for $\text{C}_{30}\text{H}_{30}\text{N}_4$; C, 80.68; H, 6.77; N, 12.55. Found: C, 80.66; H, 6.72; N, 12.57.

One-Pot Procedure for Preparation of 1-Aryl-2-(3,6-diisobutylpyrazin-2-yl)acetylenes.

A solution of 2-chloro-3,6-diisobutylpyrazine (**1**) (226 mg, 1 mmole), trimethylsilylacetylene (108 mg, 1.1 mmoles), and tetrakis(triphenylphosphine)palladium (58 mg, 0.05 mmole) in triethylamine (2 ml) was heated at 100° for 6 hours in a sealed tube. The solvent was removed under reduced pressure. To the resulting oil, an aryl or heteroaryl halide (1 mmole), potassium acetate (147 mg, 1.5 mmoles), tetrakis(triphenylphosphine)palladium (58 mg, 0.05 mmoles), and *N,N*-dimethylformamide (5 ml) were added and then the reaction mixture was heated at 100° for 2 hours. The solvent was evaporated *in vacuo* and the residue was triturated with water (15 ml) and extracted with ether (10 ml x 3). The ethereal extract was usually worked up to give crude products, which were purified by medium pressure column chromatography and eluted with a mixture of hexane-ethyl acetate to yield 1-aryl-2-(3,6-diisobutylpyrazin-2-yl)acetylenes.

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