concentrated HCl (8 ml) and heated under reflux with stirring for 4 hr. Filtration of the product followed by treatment with dilute sodium bicarbonate solution and ethanol gave yellow crystals of **6b**: yield 1.3 g (88%); mp >305°; ir 1702 and 1612 cm^{-1}; uv max 222 nm (log ϵ 4.26), 260 (4.35), and 335 (4.05).

Anal. Calcd for $C_{14}H_7N_3O_3S$: C, 56.57; H, 2.37; N, 14.14. Found: C, 56.86; H, 2.67; N, 13.90.

Registry No.—**1a**, 24848-32-6; **1b**, 29669-34-9; **2a**, 24848-33-7; **2b**, 29669-36-1; **2c**, 29913-47-1; **3a**, 29669-37-2; **3b**, 29669-38-3; **3c**, 29669-39-4; **3d**, 29669-40-7; **4b**, 29669-41-8; **6a**, 29669-42-9; **6b**, 29669-43-0.

Acknowledgment.—Thanks are expressed to Dr. T. R. Govindachari for his interest in the above work and to Dr. S. Selvavinayakam for analytical and spectral data.

Synthesis of Pyrido[1,2-*a*]pyrimido[4,5-*b*]pyridine and Related Tricyclic Systems

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Contribution No. 217 from the CIBA Research Centre, Goregaon, Bombay-63, India

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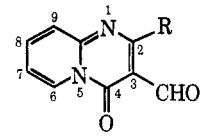
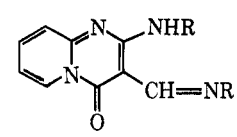
It has been reported earlier[1] that the pyrido[1,2-*a*]-pyrimidine nucleus can be functionalized by the Vilsmeier–Haack reaction to obtain 2-chloro-3-formyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (**1a**). We are describing below a facile synthesis of some tricyclic systems starting from **1a**.

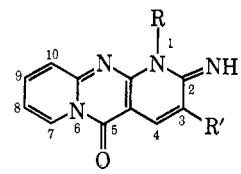
Reaction of **1a** with methylamine and benzylamine took place exothermically to give the aldimines **2a** and **2b**. The nmr spectrum of **2a** showed the presence of the $CH{=}NCH_3$ moiety, the methyl as a doublet at δ 3.42, and the methine proton as a quartet at δ 8.78 ($J_{CH,NCH_3} = -1.6$ Hz);[2] **2b** showed the presence of the $CH{=}NCH_2C_6H_5$ moiety, the methylene group as a doublet at δ 4.72, and the methine proton as a triplet at δ 9.03 ($J_{CH,NCH_2} = -1.3$ Hz). Acid hydrolysis of **2a** gave the aldehyde **1b**. Treatment of **2a** with malononitrile gave in excellent yield 3-cyano-2-imino-1-methyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimido[4,5-*b*]pyridine (**3a**) which showed in the ir spectrum the imino group at 3300 cm^{-1} and the conjugated cyano group at 2210 cm^{-1}. In general, the formation of the above tricyclic system was very facile using compounds containing active methylene groups adjacent to a cyano group. Thus, ethyl cyanoacetate, cyanoacetamide, and benzoyl acetonitrile[3] reacted with **2a** to give compounds **3b–d** and benzoyl acetonitrile with **2b** to give **3e**. The course of the reaction can be envisaged to proceed through the addition of the anions of the above reagents followed by elimination of methylamine or benzylamine to give compounds **3a–e**. Aminoacetonitrile and cyanamide



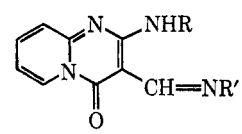
1a, R = Cl
b, R = $NHCH_3$



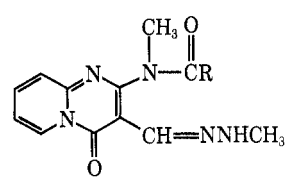
2a, R = CH_3
b, R = $CH_2C_6H_5$



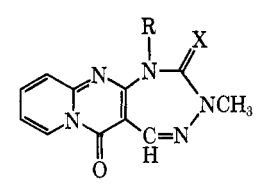
3a, R = CH_3; R′ = CN
b, R = CH_3; R′ = $COOC_2H_5$
c, R = CH_3; R′ = $CONH_2$
d, R = CH_3; R′ = COC_6H_5
e, R = $CH_2C_6H_5$; R′ = COC_6H_5



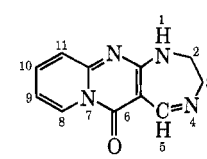
4a, R = CH_3; R′ = $NHCH_3$
b, R = $CH_2C_6H_5$; R′ = $NHCH_3$
c, R = CH_3; R′ = $-N(CH_3)-CO-Cl$
d, R = CH_3; R′ = $-N(CH_3)-CO-N$⟨morpholine⟩
e, R = $CH_2C_6H_5$; R′ = $-N(CH_3)-CS-Cl$
f, R = $CH_2C_6H_5$; R′ = $-N(CH_3)-CS-N$⟨morpholine⟩
g, R = CH_3; R′ = $-N(CH_3)-CO-COOC_2H_5$



4c′, R = Cl
d′, R = N⟨morpholine⟩



5a, X = O; R = CH_3
b, X = S; R = $CH_2C_6H_5$



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failed to react with **2a** and **2b**. Active methylene compounds such as acetylacetone, phenacyl chloride, or chloroacetone did not give tricyclic systems from **2a**, the only product which could be isolated and characterized being **1b**, the aldehyde corresponding to **2a**.

Methylhydrazine reacted with **2a** and **2b** to give the corresponding *N*-methylhydrazones **4a** and **4b**. On reaction with phosgene in toluene, **4a** gave a product

(1) E. A. Ingalls and F. D. Popp, *J. Heterocycl. Chem.*, **4**, 525 (1967).

(2) (a) K. Tori, M. Ohtsuru, and T. Kubota, *Bull. Chem. Soc. Jap.*, **39**, 1089 (1966); (b) W. Brugel, "Nuclear Magnetic Resonance Spectra and Chemical Structure," Academic Press, New York,-London, 1967, p 191.

(3) J. B. Dorsch and S. M. McElvain, *J. Amer. Chem. Soc.*, **54**, 2960 (1932).

which showed in the ir spectrum bands at 1740 and 1690 cm^{-1} and an identical uv spectrum in conformity with structure **4c**. Thiophosgene in toluene reacted with **4b** to give **4e** in an analogous way. **4c** and **4e** were surprisingly stable to aqueous alkali at ambient temperature but reacted with morpholine in refluxing dioxane to afford the morpholinoformyl derivative **4d** and the morpholinothioformyl derivative **4f**, respectively. The nmr spectrum of **4d** showed a doublet corresponding to the $NHCH_3$ group at δ 3.18 which coalesced to a singlet on addition of D_2O. Therefore, it is possible to rule out the alternate structures **4d'** for the morpholinoformyl derivative and **4c'** for its precursor, the chloroformyl derivative. **4d** underwent acid hydrolysis to give the aldehyde **1b**, providing confirmation of the structures **4d** and **4c** for the above derivatives. Attempts to cyclize **4c** and **4e** to obtain the pyrido[1,2-*a*]-pyrimido[4,5-*e*]-1,2,4-triazepine system **5a** and **5b** were unsuccessful. On treatment of **4a** with ethyl chloroformate, **4g** was obtained which resisted attempts at cyclization to give **5a**.

The β-chloroaldehyde **1a** reacted with ethylenediamine to give in good yield 1,2-dihydro-3*H*,6*H*-6-oxopyrimido[4,5-*e*]-1,4-diazepine **6**, whereas 2-chloro-5-nitrobenzaldehyde[4] on reaction with ethylenediamine failed to give any concrete results.[5]

Experimental Section

Melting points are uncorrected. The ir spectra were examined as Nujol mulls on a Perkin-Elmer Model 421 spectrophotometer. The uv spectra in 95% ethanol were recorded on a Beckman DK-2A Model spectrophotometer and the nmr spectra were recorded on a Varian A-60 spectrometer with TMS as internal standard. Thin layer chromatography (tlc) was performed on rilica gel G plates.

2-Chloro-3-formyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (1a) was prepared from 2,3-dihydro-2,4-dioxopyrido[1,2-*a*]pyrimidine by the Vilsmeier–Haack reaction in 76% yield, mp 226–227° (lit.[1] yield 39%, mp 224–226°).

2-Methylamino-3-(*N*-methyl)formimidoyl-4-oxo-4*H*-pyrido-[1,2-*a*]pyrimidine (2a).—To a stirred suspension of **1a** (4.0 g) in ethanol cooled to 10°, was added a saturated solution of methylamine in ethanol dropwise until a clear homogeneous solution was obtained. The reaction mixture which became exothermic was stirred for 2 hr. The yellow precipitate obtained was filtered and washed with water and 2-propanol. Recrystallization from ethanol gave 3.5 g of **2a** (85%): mp 154°; ir 3350, 1680, 1620, and 770 cm^{-1}; uv λ max 260 mμ (log ϵ 4.45), 352 (4.05); nmr ($CDCl_3$) δ 3.07 (d, 3 H, J = 6 Hz, $NHCH_3$), 3.42 (d, 3 H, J = −1.6 Hz, $CH{=}NCH_3$), 6.70–7.75 (m, 3 H, C-7, C-8, C-9 protons), 8.78 (q, 1 H, J = −1.6 Hz, $CH{=}NCH_3$), 8.80 (d, 1 H, C-6 H), 10.4 (s, broad, 1 H, NH). On addition of D_2O, the doublet at δ 3.07 coalesced to a singlet and the broad singlet at δ 10.4 disappeared.

Anal. Calcd for $C_{11}H_{12}N_4O$: C, 61.09; H, 5.59; N, 25.58. Found: C, 60.81; H, 5.66; N, 25.86.

2-Benzylamino-3-(*N*-benzyl)formimidoyl-4-oxo-4*H*-pyrido-[1,2-*a*]pyrimidine (2b) was obtained in an analogous way from **1a** and 3 equiv of benzylamine in 81% yield: mp 127° on recrystallization from ethanol; ir 3350, 1680, 1625, and 770 cm^{-1}; uv λ max 265 mμ (log ϵ 4.54), 358 (4.11); nmr ($CDCl_3$) δ 4.72 (d, 2 H, J = −1.3 Hz, $CH{=}NCH_2C_6H_5$), 4.80 (d, 2 H, J = 6 Hz, $NHCH_2C_6H_5$), 6.7–7.6 (m, 13 H, C-7, C-8, C-9, and phenyl protons), 8.86 (d, 1 H, C-6 H), 9.01 (t, 1 H, J = −1.3 Hz, $CH{=}NCH_2C_6H_5$), 11.25 (s, broad, 1 H, NH). On addition of D_2O, the doublet at δ 4.80 coalesced to a singlet and the singlet at δ 11.25 disappeared.

Anal. Calcd for $C_{23}H_{20}N_4O$: C, 74.98; H, 5.47; N, 15.21. Found: C, 74.93; H, 5.62; N, 14.94.

2-Methylamino-3-formyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (1b).—A solution of **2a** (1.0 g) in ethanol (20 ml) containing 6 *N* HCl (0.5 ml) was refluxed for 0.5 hr. The product obtained on cooling was filtered, washed with water, and recrystallized from 2-propanol to give 0.3 g of **1b**: mp 220°; ir 3300, 1695, 1615, and 1590 cm^{-1}; uv λ max 252 mμ (log ϵ 4.28), 350 (3.99); λ infl 276 mμ (log ϵ 4.08); nmr (CF_3COOH) δ 3.45 (d, 3 H, $NHCH_3$) 7.75–8.20 (m, 2 H, C-7, C-8 protons), 8.62 (m, 1 H, C-9 proton), 9.46 (d, 1 H, C-6 H), 10.1 (s, 1 H, CHO), 10.59 (s, broad, 1 H, NH).

Anal. Calcd for $C_{10}H_9N_3O_2$: C, 59.10; H, 4.46; N, 20.68. Found: C, 59.01; H, 4.56; N, 20.57.

2-Imino-1-methyl-3-(substituted)-4-oxo-4*H*-1,2-dihydropyrido-[1,2-*a*]pyrimido[4,5-*b*]pyrimidines (3a–d). 3-Cyano Derivative 3a.—To a solution of **2a** (2.03 g, 0.01 mol) in chloroform (15 ml) was added a solution of malononitrile (0.72 g, 0.011 mol) in chloroform and heated under reflux for 2 hr. The yellow crystalline product obtained was filtered and recrystallized from methanol–chloroform: yield 1.4 g (60%); mp 292°; ir 3300, 2210, and 1680 cm^{-1}; uv λ max 222 mμ (log ϵ 4.63), 272 (4.85), 4.03 (4.57); nmr (CF_3COOH) 4.35 (s, 3 H, NCH_3), 7.6–8.58 (m, 4 H, C-4, C-8, C-9, and C-10 protons), 9.28 (2 H, s, broad, C-7 and NH protons).

Anal. Calcd for $C_{13}H_9N_5O$: C, 62.14; H, 3.61; N, 27.88. Found: C, 61.68; H, 3.82; N, 27.58.

3-Carbethoxy derivative 3b was obtained from **2a** and ethyl cyanoacetate in 70% yield, mp 233° on recrystallization from chloroform–methanol.

Anal. Calcd for $C_{15}H_{14}N_4O_3$: C, 60.39; H, 4.73; N, 18.78. Found: C, 60.70; H, 4.81; N, 19.09.

3-Carboxamido derivative 3c was obtained from **2a** and cyanoacetamide in 63% yield, mp 320° on recrystallization from chloroform–methanol.

Anal. Calcd for $C_{13}H_{11}N_5O_2$: C, 57.98; H, 4.12; N, 26.01. Found: C, 58.27; H, 4.40; N, 25.80.

3-Benzoyl derivative 3d was obtained from **2a** and benzoyl acetonitrile in 67% yield: mp 262–263° from 2-propanol–methylene chloride; ir 3305, 1700, 1660, and 1580 cm^{-1}; uv λ max 252 mμ (log ϵ 4.43), 307 (4.14).

Anal. Calcd for $C_{19}H_{14}N_4O_2$: C, 69.08; H, 4.27; N, 16.96. Found: C, 68.77; H, 4.60; N, 16.81.

1-Benzyl-2-imino-3-benzoyl-4-oxo-4*H*-1,2-dihydropyrido[1,2-*a*]-pyrimido[4,5-*b*]pyrimidine (3e) was obtained from **2b** and phenacyl cyanide in 58% yield, mp 215° from chloroform–methanol.

Anal. Calcd for $C_{25}H_{18}N_4O_2$: C, 73.87; H, 4.46; N, 13.79. Found: C, 73.59; H, 4.65; N, 14.02.

2-Methylamino-3-(*N*-methyl)aminoformimidoyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (4a).—Methylhydrazine (1.01 g, 0.022 mol) in ethanol (10 ml) was added to **2a** (4.32 g, 0.02 mol) and stirred under reflux for 2 hr. The precipitate obtained was filtered and recrystallized from methanol to afford 4.1 g (94%) of **4a**: mp 210°; ir 3260, 1655, and 1605 cm^{-1}; uv λ max 264 mμ (log ϵ 4.47); nmr ($CDCl_3$ + CD_3SOCD_3) δ 2.90 (s, 3 H, $=NNHCH_3$), 3.15 (d, 3 H, $NHCH_3$), 6.9–7.6 (m, 3 H, C-7, C-8, and C-9 protons), 8.22 (s, 1 H, CH=N), 8.86 (d, 1 H, C-6 proton).

Anal. Calcd for $C_{11}H_{13}N_5O$: C, 57.13; H, 5.67; N, 30.29. Found: C, 57.42; H, 5.87; N, 30.18.

2-Benzylamino-3-(*N*-methyl)aminoformimidoyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (4b) was obtained in 61% yield from **2b** under conditions described for **4a**, mp 156° on recrystallization from methanol.

Anal. Calcd for $C_{17}H_{17}N_5O$: C, 66.43; H, 5.58; N, 22.79. Found: C, 66.80; H, 5.66; N, 23.00.

***N*-Chloroformyl derivative 4c** was obtained in 74% yield by refluxing **4a** with excess of phosgene in toluene: mp 226° on recrystallization from CH_2Cl_2–hexane; ir 1720, 1695, and 1615 cm^{-1}; uv λ max 262 mμ (log ϵ 3.92).

Anal. Calcd for $C_{12}H_{12}ClN_5O_2$: C, 49.07; H, 4.12; N, 23.84. Found: C, 49.18; H, 4.18; N, 23.80.

***N*-(Morpholinoformyl) derivative 4d** was obtained by treatment of **4c** with morpholine in dioxane in 66% yield: mp 193° on recrystallization from methylene chloride; ir 3290, 1680, and 1630 cm^{-1}; uv λ max 262 mμ (log ϵ 4.51); nmr ($CDCl_3$) δ 3.18 (d, 3 H, $NHCH_3$), 3.35 (s, 3 H, $=NNCH_3$), 3.55 (m, 4 H, NCH_2), 3.72 (m, 4 H, OCH_2), 6.85–7.75 (m, 3 H, C-7, C-8, and C-9 protons), 8.26 (s, 1 H, CH=N), 8.86 (d, 1 H, C-6 proton), and 8.95 (s, 1 H, NH).

Anal. Calcd for $C_{16}H_{20}N_6O_3$: C, 55.80; H, 5.85; N, 24.41. Found: C, 55.61; H, 5.71; N, 24.09.

Acid Hydrolysis of 4d to 1b.—A solution of **4d** (0.5 g) in ethanol (15 ml) containing 2 *N* HCl (5 ml) was refluxed for 1 hr.

(4) H. Erdmann, *Justus Liebigs Ann. Chem.*, **272**, 148 (1892).

(5) Unpublished observations by authors.

The solvent was removed under reduced pressure to give a residue which was treated with water to give 0.2 g of a product which melted at 219° on recrystallization from 2-propanol and was identical with **1b** described above by mixture melting point, spectral comparison, and tlc behavior.

N-**Thioformyl derivative 4e** was obtained in 71% yield by refluxing **4b** with thiophosgene in toluene: mp 236° on recrystallization from methylene chloride–ether; ir 3250, 1660, and 1610 cm^{-1}.

Anal. Calcd for $C_{18}H_{16}ClN_5OS$: C, 56.03; H, 4.18; N, 18.15. Found: C, 56.21; H, 4.36; N, 17.86.

N-**Morpholinothioformyl derivative 4f** was obtained in 56% yield from **4e** and morpholine, mp 166° on recrystallization from methylene chloride–ether.

Anal. Calcd for $C_{22}H_{24}N_6O_2S$: C, 60.54; H, 5.54; N, 19.26. Found: C, 60.76; H, 5.52; N, 19.22.

2-Methylamino-3-(*N*-carbethoxy-*N*-methyl)aminoformimidoyl-4-oxo-4*H*-pyrido[1,2-*a*]pyrimidine (4g).—To a suspension of **4a** (2.31 g, 0.01 mol) in dioxane (80 ml) containing pyridine (0.8 g) was added ethyl chloroformate (1.08 g, 0.01 mol) and the mixture was heated with stirring under reflux for 4 hr. The precipitate obtained on cooling was filtered and recrystallized from methylene chloride to give 1.8 g of **4g**: mp 190°; ir 3510, 1680, and 1660 cm^{-1}; uv λ max 260 mμ (log ε 4.48).

Anal. Calcd for $C_{14}H_{17}N_5O_3$: C, 55.43; H, 5.65; N, 23.09. Found: C, 55.16; H, 6.17; N, 22.91.

6-Oxo-3*H*,6*H*-1,2-dihydropyrimido[4,5-*e*]-1,4-diazepine (6).—Ethylenediamine (3.6 g, 0.06 mol) was added to a stirred suspension of **1a** (4.16 g, 0.02 mol) in dioxane (50 ml) and heated under reflux for 4 hr. The precipitate obtained on cooling was filtered and washed with water and ethanol. On recrystallization from chloroform–dioxane 2.9 g (78%) of product was obtained: mp 310°; ir 1680, 1610, and 1460 cm^{-1}; uv[6] λ max 217 and 265 mμ; nmr (CF_3COOH) δ 4.25 (s, broad, 4 H, $NHCH_2$ and $=NCH_2$), 7.3–7.7 (m, 3 H, C-9, C-10, and C-11 protons), 8.32 (m, 1 H, CH=N), 9.03 (d, 1 H, C-8 proton).

Anal. Calcd for $C_{11}H_{10}N_4O$: C, 61.67; H, 4.71; N, 25.97. Found: C, 61.49; H, 4.75; N, 25.77.

Registry No.—**1b**, 29494-74-4; **2a**, 29494-75-5; **2b**, 29494-76-6; **3a**, 29494-77-7; **3b**, 29494-78-8; **3c**, 29494-79-9; **3d**, 29494-80-2; **3e**, 29494-81-3; **4a**, 29494-82-4; **4b**, 29494-83-5; **4c**, 29494-84-6; **4d**, 29494-85-7; **4e**, 29494-86-8; **4f**, 29494-87-9; **4g**, 29494-88-0; **6**, 29494-89-1.

Acknowledgment.—Thanks are expressed to Dr. T. R. Govindachari for his interest in the above work and Dr. S. Selvavinayakam for analytical and spectral data.

(6) Only qualitative assay could be performed owing to the high degree of insolubility of the compound in solvents.

Syntheses and Cis–Trans Isomerization of Light-Sensitive Benzenediazo Sulfides

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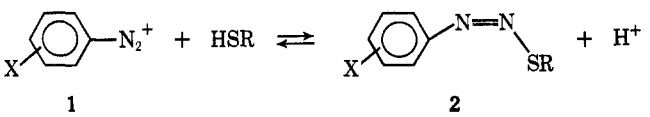
Although until recently benzenediazoalkyl sulfides were considered highly decomposable,[1] Van Zwet and Kooyman succeeded in preparing benzenediazo-*tert*-butyl sulfide and its 2,4,6-trimethyl derivative and in determining the cis–trans isomerization and other physical properties.[2] We were unable to prepare other derivatives by the same method but found a more general synthesis which is presented in the Experimental Section. In this way a number of new derivatives were synthesized and studied, especially in view of their applicability in photographic physical development systems, a subject extensively discussed elsewhere.[3] For photographic applications the very slow thermal cis-to-trans isomerization and the stability of the cis isomer are important properties which were also helpful in studying the synthesis. The reaction was found to depend on the equilibrium between diazonium ion **1** and benzene-*cis*-diazo sulfide **2**. The substituents X



1 **2**

appear to determine the quantity of cis isomer **2** formed when the reactants **1** and thiol are brought together. The solution of the reactants showed, *e.g.*, when X was 3,5-Cl_2-4-$N(CH_3)_2$, immediately the absorption spectrum of the cis isomer **2** which must mean according to empirical determinations that there was at most 4% diazonium left. A dilute solution of H_2SO_4 had to be added to move the equilibrium to the left which produced the diazonium spectrum. On the other hand, when, *e.g.*, X was 2,5-$(OCH_3)_2$-4-(4′-tolylmercapto), a compound photographically uninteresting and therefore not extensively studied, the diazonium spectrum was observed which changed to the cis spectrum if NaOH solution was added to move the equilibrium to the right. Generally, when the NaOH solution was added too rapidly, the diazonium salt decomposed with the formation of nitrogen. Optimum yields of pure cis isomers were obtained when the NaOH solution was slowly added to give a final pH of 6.

The final step in the synthesis is the thermal isomerization of the cis isomers to obtain the photographically applied trans isomers. We found that the thermal isomerization in benzene, the solvent chosen by Van Zwet and Kooyman,[2] even in yellow safe-light was accompanied by decomposition. Much purer products were obtained by heating in isooctane at 90° for 2 hr.

The data in Table I were calculated from 5 to 12 points by the method of least squares. The deviations are standard deviations. The correlation coefficients were all better than 0.99. Frequency factors *f*

TABLE I

THERMAL CIS-TO-TRANS ISOMERIZATION RATE CONSTANTS *k* IN ETHANOL AT 60° FOR BENZENEDIAZO SULFIDES ($C_6H_5N_2SR$) WITH SUBSTITUENTS X ATTACHED TO THE BENZENE RING[a]

X	R	10^4k, sec^{-1}	Log *f*	*A*, kcal/mol
4-NO_2	*tert*-Butyl	0.52 ± 0.01	12.4 ± 0.7	25.3 ± 1.0
2-Cl-4-NO_2	*tert*-Butyl	0.83 ± 0.01	11.7 ± 0.7	24.0 ± 1.1
4-CN	*tert*-Butyl	0.48 ± 0.01	18.3 ± 0.9	28.8 ± 1.3
4-Cl	*tert*-Butyl	0.39 ± 0.04	11.2 ± 1.0	23.8 ± 1.6
4-NO_2	*tert*-Octyl	1.40 ± 0.01	12.2 ± 0.4	24.4 ± 0.6
4-NO_2	$C(C_6H_5)_3$	35 ± 2	11.7 ± 0.3	21.5 ± 0.5

[a] Frequency factors *f* and activation energies *A*.

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