

## Photochemical Electrocyclization of 1,4,6-Trisubstituted Pyrimidin-2-ones to 2-Oxo-1,3-diazabicyclo[2.2.0]hex-5-enes<sup>1</sup>

By Takehiko Nishio,\* Akira Kato, Choji Kashima, and Yoshimori Omote, Department of Chemistry, University of Tsukuba, Sakura-mura, Niihari-gun, Ibaraki, 300-31, Japan

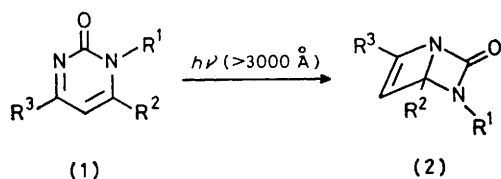
Photochemical reactions of 1,4,6-trisubstituted pyrimidin-2-ones have been examined. Irradiation of 1,4,6-trisubstituted pyrimidin-2-ones (1a—k) in benzene yielded the photochemical electrocyclic products, 3,4,6-trisubstituted 2-oxo-1,3-diazabicyclo[2.2.0]hex-5-enes (2a—k) in 14—85% yield as the sole product, while 1,6-dimethyl-4-dimethylaminopyrimidin-2-one (11) was photochemically inactive. The photoisomers (2a—k) were stable at room temperature.

THE photochemistry of nucleoside bases and their derivatives is an area of significant importance for understanding the photo-reactivity of nucleic acids and this area has been extensively studied.<sup>2</sup> It is also of interest to study the photochemical reactions of pyrimidin-2-ones related to cytosine, which is one of the nucleoside bases, and its derivatives. However, little attention has been paid to the photochemistry of pyrimidin-2-ones. Pfoertner<sup>3</sup> recently reported the photochemical alcohol addition and dimerization of 4,6-dimethylpyrimidin-2-ol, and Shetlar *et al.*<sup>4</sup> also reported the photochemical alcohol addition of 1-methyl-4-methylaminopyrimidin-2-one. In this paper we report the photochemical reactions of 1,4,6-trisubstituted pyrimidin-2-ones (1a—l) and the photochemical synthesis of 3,4,6-trisubstituted 2-oxo-1,3-diazabicyclo[2.2.0]hex-5-enes (2a—k).

### RESULTS AND DISCUSSION

When 1-phenyl-4,6-dimethylpyrimidin-2-one [(1a);  $\lambda_{\text{max}}$ (EtOH) 210 ( $\epsilon$   $2.23 \times 10^4$ ) and 304 nm ( $9.5 \times 10^3$ )] in benzene was irradiated in a Pyrex vessel with a high-pressure mercury lamp under an argon atmosphere for 15 h at room temperature, 3-phenyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2a), an isomer of (1a), was obtained in 67% isolated yield. The structure of

2,4-dimethyl-1-azacyclobutadiene, and 2,4-dimethyl-1-azacyclobutadienyl cation fragments, respectively. (ii) The photo-product (2a) showed i.r. absorption at 1760  $\text{cm}^{-1}$  characteristic of the fused urea carbonyl and 1640  $\text{cm}^{-1}$  due to the fused cyclobutene double bond, respectively. (iii) The n.m.r. spectrum of (2a) showed a singlet at  $\delta$  1.83 (3 H), a doublet at  $\delta$  2.08 (3 H,  $J$  1.5 Hz), and a quartet at  $\delta$  6.00 (1 H,  $J$  1.5 Hz), assignable to 4-methyl, 6-methyl, and olefinic protons, respectively, in addition to aromatic proton peaks, while that of (1a) showed a singlet at  $\delta$  2.40 (3 H), a doublet at  $\delta$  1.98 (3 H,  $J$  0.7 Hz), and a quartet at  $\delta$  6.20 (1 H,  $J$  0.7 Hz) assignable to 4-methyl, 6-methyl, and olefinic proton, respectively, in addition to aromatic proton peaks. The chemical shifts easily differentiate (2a) from (1a). As expected on the basis of its formulation, hydrogenation over palladium-charcoal of the photo-isomer (2a) gave 3-phenyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hexane (3) in 59.5% yield. The photo-isomer (2a) was very stable at room temperature. However, thermolysis of molten (2a) in a sealed tube at 200 °C, or irradiation of (2a) in methanol through quartz with 2537-Å radiation causes reversion to the starting pyrimidin-2-one (1a) in almost quantitative yield. Furthermore, the photo-isomer (2a) was treated with potassium methoxide in methanol to give methyl *N*-phenylcarbamate (4) and unidentified products from 2,4-dimethyl-1-azacyclobutadiene. Treatment of (1a) with potassium methoxide in methanol did not give any products, and (1a) was quantitatively recovered. The starting pyrimidin-2-one (1a) was obtained when (2a) was treated with hydrochloric acid, followed by sodium hydrogencarbonate. Similarly, irradiation of other 1,4,6-trisubstituted pyrimidin-2-ones (1b—k) under the same conditions gave the corresponding 3,4,6-trisubstituted 2-oxo-1,3-diazabicyclo[2.2.0]hex-5-enes (2b—k) in 14—85% yields (Table 1). These photo-isomers (2b—k) were also stable at room temperature and could be stored indefinitely. In the case of (2b) and (2j), they were converted back to the starting pyrimidin-2-ones (1b) and (1j) during purification by distillation (125 °C at 5 mmHg) or recrystallization (benzene-hexane). The spectral properties of the photoproducts (2a—k) are shown in Table 2. On the other hand, irradiation of 1,6-dimethyl-4-dimethylaminopyrimidin-2-one (11), analogous to cytosine, in benzene-methanol



(1)			(2)		
R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>
a; Ph	Me	Me	g; <i>o</i> -ClC <sub>6</sub> H <sub>4</sub>	Me	Me
b; Me	Me	Me	h; <i>p</i> -MeC <sub>6</sub> H <sub>4</sub>	Me	Me
c; <i>o</i> -MeC <sub>6</sub> H <sub>4</sub>	Me	Me	i; <i>p</i> -MeOC <sub>6</sub> H <sub>4</sub>	Me	Me
d; <i>o</i> -MeOC <sub>6</sub> H <sub>4</sub>	Me	Me	j; $\beta$ -naphthyl	Me	Me
e; <i>o</i> -EtC <sub>6</sub> H <sub>4</sub>	Me	Me	k; Me	Ph	Me
f; <i>o</i> -EtOC <sub>6</sub> H <sub>4</sub>	Me	Me	l; Me	Me	NMe <sub>2</sub>

(2a) was determined on the basis of physical data and elemental analysis. The photo-product (2a) was also distinguished from the starting pyrimidin-2-one (1a) as follows. (i) The mass spectrum of the photo-product (2a) revealed a base peak at  $m/e$  200 and major peaks at  $m/e$  119, 81, and 80, corresponding to phenyl isocyanate,

under the same conditions did not give any photo-products and (11) was quantitatively recovered. The formation of 3,4,6-trisubstituted 2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2) can be explained in terms of a photochemical

and 0.79 in benzene, respectively, and that of (1a) was 0.04 in methanol. The photo-reaction of (1a) was quenched slightly by penta-1,3-diene, and was not affected by triplet sensitizers such as benzophenone and

TABLE 2

The i.r. and n.m.r. spectra of the photo-products (2a—k)

Photoproduct	I.r. (cm <sup>-1</sup> ) *	N.m.r. (δ in CDCl <sub>3</sub> )
(2a)	1 760, 1 640, 1 600, 1 500. 1 380, 750	1.83 (s, 3 H), 2.08 (d, 3 H, <i>J</i> 1.5 Hz), 6.00 (q, 1 H, <i>J</i> 1.5 Hz), 6.75—7.4 (m, 5 H)
(2b)	1 760, 1 635, 1 380	1.58 (s, 3 H), 2.00 (d, 3 H, <i>J</i> 1.7 Hz), 2.75 (s, 3 H), 5.80 (q, 1 H, <i>J</i> 1.7 Hz)
(2c)	1 770, 1 640, 1 605, 1 500, 1 380, 750	1.58 (s, 3 H), 2.04 (d, 3 H, <i>J</i> 1.5 Hz), 2.31 (s, 3 H), 5.98 (q, 1 H, <i>J</i> 1.5 Hz), 6.9—7.1 (m, 4 H)
(2d)	1 775, 1 640, 1 590, 1 505, 1 360, 750	1.74 (s, 3 H), 2.11 (d, 3 H, <i>J</i> 1.5 Hz), 3.87 (s, 3 H), 6.16 (q, 1 H, <i>J</i> 1.5 Hz), 6.8—7.3 (m, 2 H), 7.3—7.5 (m, 1 H), 7.6—7.9 (m, 1 H)
(2e)	1 780, 1 640, 1 600, 1 495, 1 365, 755	1.20 (t, 3 H), 1.61 (s, 3 H), 2.07 (d, 1 H, <i>J</i> 1.7 Hz), 2.70 (q, 2 H), 6.03 (q, 1 H, <i>J</i> 1.7 Hz), 6.8—7.2 (m, 4 H)
(2f)	1 770, 1 640, 1 595, 1 505, 1 380, 745	1.41 (t, 3 H), 1.70 (s, 3 H), 2.04 (d, 3 H, <i>J</i> 1.5 Hz), 3.94 (q, 2 H), 5.87 (q, 1 H), <i>J</i> 1.5 Hz), 6.5—7.1 (m, 3 H), 7.25—7.5 (m, 1 H)
(2g)	1 775, 1 640, 1 585, 1 485, 1 365, 755	1.72 (s, 3 H), 2.09 (d, 3 H, <i>J</i> 1.5 Hz), 6.04 (q, 1 H, <i>J</i> 1.5 Hz), 6.9—7.6 (m, 4 H)
(2h)	1 765, 1 638, 1 610, 1 510, 1 385, 810, 765	1.83 (s, 3 H), 2.10 (d, 3 H, <i>J</i> 1.5 Hz), 2.32 (s, 3 H), 6.15 (q, 1 H, <i>J</i> 1.5 Hz), 7.20 (br s, 4 H)
(2i)	1 770, 1 640, 1 580, 1 510, 1 395, 840, 760	1.74 (s, 3 H), 2.02 (d, 3 H, <i>J</i> 1.5 Hz), 3.64 (s, 3 H), 5.88 (q, 1 H, <i>J</i> 1.5 Hz), 6.64 (d, 2 H, <i>J</i> 6.4 Hz), 6.94 (d, 2 H, <i>J</i> 6.4 Hz)
(2j)	3 050, 1 780, 1 643, 1 515, 1 380, 1 360, 800, 780, 680	1.69 (s, 3 H), 2.15 (d, 3 H, <i>J</i> 1.5 Hz), 6.18 (q, 1 H, <i>J</i> 1.5 Hz), 7.0—8.2 (m, 7 H)
(2k)	1 775, 1 650, 1 390, 1 380, 765, 700	2.08 (d, 3 H, <i>J</i> 1.4 Hz), 2.75 (s, 3 H), 6.18 (q, 1 H, <i>J</i> 1.4 Hz), 7.26 (br s, 5 H)

\* The i.r. spectra of (2a, c, and h—k) were measured in KBr, and those of (2b, and d—f) were measured neat.

electrocyclization reaction. Similar reactions of pyrazinone,\* 2-pyrone,<sup>6</sup> and 2-pyridone derivatives<sup>7</sup> have been reported. The formation of (2a) is enhanced in aprotic solvents such as benzene, dioxan, and acetone. However, acetonitrile showed no influence compared with a

acetophenone. These results are summarized in Table 3. The above results indicate that the photochemical electrocyclization of (1a) to (2a) proceeds mainly *via* the singlet state, considering that this photo-reaction is an intramolecular one.

TABLE 1

Photochemical reaction of 1,4,6-trisubstituted pyrimidin-2-ones in benzene

Reactant	Time/h	Yield (%) *		Quantum yield
		(2)	(1; recovered)	
(1a)	15	67	10	0.52
(1a)	2	61 <sup>b</sup>	39 <sup>b</sup>	
(1b)	15	33	30	0.79
(1b)	2	93 <sup>b</sup>	7 <sup>b</sup>	
(1c)	15	85	13	
(1d)	12	55	13	
(1e)	12	50	17	
(1f)	10	43	15	
(1g)	15	66	25	
(1h)	15	57	30	
(1i)	15	33	25	
(1j)	45	23	35	
(1k)	45	14	63	
(1l)	50		<i>ca.</i> 100	

\* The isolated yield. <sup>b</sup> The yield determined by g.l.c. The isolated yield of (2b) is not so high since the photo-product (2b) converts gradually back to the starting pyrimidin-2-one (1b) during column chromatography. A similar phenomenon is observed for (1j) and (1k).

protic solvent such as methanol upon photo-reaction. The quantum yields for the photochemical electrocyclization of (1a) and (1b) [to (2a) and (2b)] were 0.52

\* In the photochemistry of pyrazinone, Furrer reported that 1,3,5,6-tetramethylpyrazin-2-one gave an unstable 1,2,4,6-tetramethyl-3-oxo-2,5-diazabicyclo[2,2,0]hex-5-ene, which could not be isolated, and trapped as dehydro-derivative by hydrogenation.<sup>5</sup>

TABLE 3

Photochemical reactions of 1-phenyl-4,6-dimethylpyrimidin-2-one (1a) in various solvents under argon for 15 h

Solvent	Additive	Yield of (2a) (%) *
Benzene		72
Benzene	Benzophenone	15
Benzene	Acetophenone	15
Benzene	Penta-1,3-diene	51
Dioxan		50
Acetonitrile		15.5
Propan-2-ol		33.5
Methanol		15
Water		Trace

\* Determined by g.l.c.

#### EXPERIMENTAL

The i.r., n.m.r., and mass spectra were obtained on Hitachi 260-30, R-20, and RMU-6MG spectrometers, respectively. A Ushio 450-W high-pressure mercury lamp was used as an irradiation source.

*Materials.*—The 1,4,6-trisubstituted pyrimidin-2-ones (1a—l) were prepared according to literature methods.<sup>8-10</sup>

*General Procedure for the Photoreaction of 1,4,6-Tri-substituted Pyrimidin-2-ones (1a—l).*—A solution of the pyrimidin-2-one (1) (1 mmol) in solvent (45 ml) was irradiated in a Pyrex vessel under an argon atmosphere with a high-pressure mercury lamp for 10—50 h at room temperature. After removal of the solvent, the residue was chromatographed on a silica gel column. Elution with benzene-ethyl acetate (4:1 or 2:1) afforded the photo-product (2) and then elution with ethyl acetate gave the un-

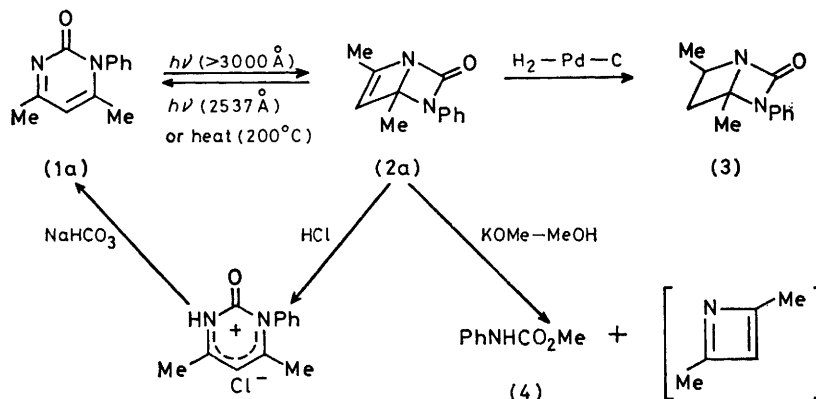
reacted pyrimidin-2-one (1). (i) 3-Phenyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2a), m.p. 64.5—66.0 °C (from benzene—hexane);  $\lambda_{\text{max}}$  (EtOH) 239.5 ( $\epsilon$  5.41  $\times$  10<sup>4</sup>), 270sh (1.59  $\times$  10<sup>4</sup>), and 280 nm sh (1.25  $\times$  10<sup>4</sup>);  $m/e$  200 ( $M^+$ ), 185, 172, 119, 81, 80, 77, and 76 (Found: C, 72.15; H, 6.1; N, 13.85. C<sub>12</sub>H<sub>12</sub>N<sub>2</sub>O requires C, 71.95; H, 6.05; N, 14.0%).

(ii) 3,4,6-Trimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2b), b.p. 125 °C at 5 mmHg (decomp.) (Kugelrohr temperature);  $m/e$  138 ( $M^+$ ), 123, 110, 97, 81, and 56 (Found:

[2.2.0]hex-5-ene (2j), m.p. 56.5—58.0 °C (Found: C, 76.65; H, 5.6; N, 11.0. C<sub>16</sub>H<sub>14</sub>N<sub>2</sub>O requires C, 76.75; H, 5.65; N, 11.2%).

(xi) 3,6-Dimethyl-4-phenyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2k), m.p. 133 °C (decomp.) (from benzene—hexane) (Found: C, 72.0; H, 5.85; N, 13.65. C<sub>12</sub>H<sub>12</sub>N<sub>2</sub>O requires C, 71.95; H, 6.05; N, 14.0%).

Hydrogenation of (2a).—A solution of (2a) (100 mg) in methanol (20 ml) was hydrogenated over palladium—charcoal (50 mg). The usual work-up gave 3-phenyl-4,6-



C, 60.6; H, 7.15; N, 20.55. C<sub>7</sub>H<sub>10</sub>N<sub>2</sub>O requires C, 60.85; H, 7.3; N, 20.25%).

(iii) 3-o-Tolyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2c), m.p. 74.5—75.5 °C (from benzene—hexane);  $\lambda_{\text{max}}$  (EtOH) 229 ( $\epsilon$  1.00  $\times$  10<sup>4</sup>) and 267sh (8.2  $\times$  10<sup>3</sup>);  $m/e$  214 ( $M^+$ ), 199, 186, 185, 90, 80, 77, and 76 (Found: C, 72.85; H, 6.6; N, 12.85. C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O requires C, 72.85; H, 6.6; N, 13.05%).

(iv) 3-o-Methoxyphenyl-4,6-dimethyl-1,3-diazabicyclo[2.2.0]hex-5-ene (2d), b.p. 120 °C at 5 mmHg (Kugelrohr temperature) (Found: C, 73.55; H, 6.95; N, 12.25. C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O<sub>2</sub> requires C, 73.65; H, 7.05; N, 12.25%).

(v) 3-o-Ethylphenyl-4,6-dimethyl-1,3-diazabicyclo[2.2.0]hex-5-ene (2e), b.p. 112 °C at 2 mmHg (Kugelrohr temperature);  $m/e$  228 ( $M^+$ ), 213, 200, 199, 198, and 144 (Found: C, 73.55; H, 6.95; N, 12.25. C<sub>14</sub>H<sub>16</sub>N<sub>2</sub>O requires C, 73.65; H, 7.05; N, 12.25%).

(vi) 3-o-Ethoxyphenyl-4,6-dimethyl-1,3-diazabicyclo[2.2.0]hex-5-ene (2f), b.p. 125 °C at 5 mmHg (Kugelrohr temperature);  $m/e$  244 ( $M^+$ ), 229, 216, 214, and 199 (Found: C, 68.7; H, 6.5; N, 11.55. C<sub>14</sub>H<sub>16</sub>N<sub>2</sub>O<sub>2</sub> requires C, 68.85; H, 6.6; N, 11.45%).

(vii) 3-o-Chlorophenyl-4,6-dimethyl-1,3-diazabicyclo[2.2.0]hex-5-ene (2g), b.p. 125 °C at 5 mmHg (Kugelrohr temperature) (Found: C, 61.25; H, 4.75; N, 12.2. C<sub>12</sub>H<sub>11</sub>ClN<sub>2</sub>O requires C, 61.4; H, 4.75; N, 11.95%).

(viii) 3-p-Tolyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2h), m.p. 62.0—63.0 °C (from benzene—hexane);  $\lambda_{\text{max}}$  (EtOH) 243 ( $\epsilon$  6.23  $\times$  10<sup>4</sup>), 275.5sh (1.47  $\times$  10<sup>4</sup>), and 285 nm sh (1.04  $\times$  10<sup>4</sup>);  $m/e$  214 ( $M^+$ ) 199, 186, 90, 80, 77, and 76 (Found: C, 73.0; H, 6.6; N, 13.05. C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O requires C, 72.85; H, 6.6; N, 13.05%).

(ix) 3-p-Methoxyphenyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hex-5-ene (2i), m.p. 66.0—67.0 °C (from benzene—hexane) (Found: C, 72.45; H, 6.6; N, 13.35. C<sub>13</sub>H<sub>14</sub>N<sub>2</sub>O<sub>2</sub> requires C, 72.85; H, 6.6; N, 13.05%).

(x) 3- $\beta$ -Naphthyl-4,6-dimethyl-2-oxo-1,3-diazabicyclo-

dimethyl-2-oxo-1,3-diazabicyclo[2.2.0]hexane (3) (60 mg), m.p. 115.0—116.0 °C (from benzene—hexane);  $\nu_{\text{max}}$  (KBr) 1765, 1600, 1500, 1380, and 690 cm<sup>-1</sup>;  $\delta$ (CDCl<sub>3</sub>) 1.45 (d, 3 H,  $J$  7.2 Hz), 1.73 (s, 3 H), 2.56 (dd, 1 H,  $J$  7.2, 12.8 Hz), 2.58 (dd, 1 H,  $J$  12.8, 27.8 Hz), 4.0—4.7 (m, 1 H), and 6.9—7.6 (m, 5 H) (Found: C, 71.05; H, 7.0; N, 13.55. C<sub>12</sub>H<sub>14</sub>N<sub>2</sub>O requires C, 71.25; H, 7.0; N, 13.6%).

Reaction of (2a) with Potassium Methoxide.—To a solution of potassium (20 mg) in methanol (5 ml) was added dropwise a solution of (2a) (100 mg) in methanol (10 ml) with stirring. The mixture was stirred for 3 h at room temperature, poured into ice-water, and then extracted with dichloromethane. The extract was washed with 10% hydrochloric acid and water, and dried over magnesium sulphate. After removal of the solvent, the residual oil was chromatographed (benzene) to give methyl *N*-phenylcarbamate (4) (70 mg), b.p. 150 °C at 4 mmHg (Kugelrohr temperature);  $\nu_{\text{max}}$  (film) 3300, 1735, 1700, 760, and 695 cm<sup>-1</sup>;  $\delta$ (CDCl<sub>3</sub>) 3.75 (s, 3 H), 6.8 (br s, 1 H, D<sub>2</sub>O exchangeable), and 7.0—7.5 (m, 5 H) (Found: C, 63.55; H, 5.95; N, 9.15. C<sub>8</sub>H<sub>9</sub>NO<sub>2</sub> requires C, 63.55; H, 6.0; N, 9.25%).

Treatment of (2a) with Hydrochloric Acid.—To a solution of (2a) (100 mg) in methanol (10 ml) was added a few drops of concentrated hydrochloric acid. The mixture was stirred for 20 h at room temperature, poured into water, and extracted with dichloromethane. The extract was washed with 10% sodium hydrogencarbonate solution and water, dried over magnesium sulphate, and evaporated *in vacuo* to give the pyrimidin-2-one (1a) quantitatively.

Quantum Yield Determinations.—Benzophenone—benzhydrol actinometry was used for quantum yield determination. The 300-nm line of a Rayonet photochemical reactor was used as an irradiation source. Samples (0.1M solutions) in Pyrex tubes were degassed to *ca.* 10<sup>-3</sup> mmHg in three freeze-thaw cycles and sealed. The samples were irradiated individually in succession. Photolyses were

carried out to 30—50% conversion. The degree of reaction was determined by g.l.c.

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