portions of ether. The combined ethereal extracts were washed with three 50-mL portions of water, separated, dried over anhydrous sodium sulfate, filtered, and evaporated in vacuo. The crude product thus obtained was refluxed for 10 h with 4 g of potassium hydroxide dissolved in a solution composed of 50 mL of water, 50 mL of THF, and 5 mL of ethanol. The solution was cooled, evaporated to a volume of 40 mL, and then diluted with 10 mL of water. The solution was neutralized with 50 mL of 6 N HCl and then extracted with two 50-mL portions of ether. The ether extracts were washed once with 10-mL of water and once with 10 mL of 5% aqueous sodium bicarbonate, dried over anhydrous magnesium sulfate, filtered, and evaporated in vacuo to give a viscous oil. Distillation of the oil afforded 4.2 g (78% from **2**) of the product: bp 175–180 °C (0.005 torr); $[\alpha]^{25}_D$ +53.2° (*c* 0.5, $CHCl_3$).

Idophinite (6). This compound was prepared by the same procedure used for the preparation of glucophinite except the diol **5** was used in place of the diol 1: mp 103–105 °C; $[\alpha]^{25}_D$ +78.4° (*c* 0.6, $CHCl_3$); ^1H NMR δ 0.9–1.2 (m, 10 H), 3.4–3.9 (m, 8 H), 6.2 (d, 1 H), 7.1–7.4 (m, 25 H); IR (CCl_4) 1020 (POC) cm^{-1}.

3-*O*-Benzyl-1,2-*O*-cyclohexylidene-α-L-idofuranose Ditosylate (7). This compound was prepared by the same procedure used for the preparation of ditosylate **2** except the diol **5** was used in place of the diol 1: $[\alpha]^{25}_D$ +89.2° (*c* 0.5, $CHCl_3$); ^1H NMR δ 0.9–1.2 (m, 10 H), 2.5 (s, 6 H), 3.6–4.2 (m, 8 H), 6.2 (d, 1 H), 7.1–7.4 (m, 13 H).

Glucophos (8). This compound was prepared by the same procedure used for the preparation of idophos except the ditosylate **7** was used in place of the ditosylate **2**: mp 162–165 °C; $[\alpha]^{25}_D$ +112° (*c* 0.4, $CHCl_3$); ^1H NMR δ 0.9–1.2 (m, 10 H), 2.3–2.9 (m, 3 H), 3.4–3.8 (m, 5 H), 5.2 (d, 1 H), 7.2–7.5 (m, 25 H).

General Procedure for the Reduction of the Olefinic Acids. To a 250-mL hydrogenation vessel were added 150 mL of THF, 0.1 mmol of $[Rh_2(COD)_2][BF_4]_2$,[11] and 0.3 mmol of phosphinite or phosphine. Then, 0.6 mmol of Et_3N and 10 mmol of the olefinic acid were added. The magnetically stirred solution was hydrogenated in a conventional apparatus at room temperature and 1 atm of hydrogen. All reactions were stopped and worked up after 1 h by conventional procedures.[4] Crude reaction yields were determined by ^1H NMR and were found to be quantitative. The results are given in Table I.

General Procedure for the Reduction of the Olefin Esters. The procedure was the same as above except that the reactions were run in the absence of Et_3N. The results are given in Table I.

(11) Prepared as previously described.[4]

Oxidation of Uric Acid. 1. Structural Revision of Uric Acid Glycols

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A reinvestigation of the chemistry of uric acid glycols (**2**) originally described by Biltz was made. On the basis of degradative evidence and infrared spectral characteristics, previously accepted structure **2** was revised, and the 4-hydroxy-2,5-dioxo-4-imidazolidinecarboxyureide (**4**) structure was assigned to these compounds. An unambiguous proof for the new formulation was obtained by the X-ray crystallographic study of the hemihydrate **4a**.

Summarizing the investigations on the oxidation of uric acid (**1a**),[2] Biltz in 1921 suggested that the first step is an oxidation at the double bond, leading to the intermediate glycol **2**.[3] In an effort to solve this intriguing problem, Biltz and co-workers investigated the reaction of alloxan (**3**) with ureas,[4] initially studied by Mulder,[5] and prepared identical products by oxidation of **1**,[6] assigning the structure **2** to these compounds.

A provocative hypothesis, however, that alloxan-like metabolites formed in vivo from **1a** may be involved in the etiology of diabetes mellitus gave an impetus for the reexamination of these compounds.[7]

We now wish to report the reinvestigation of the chemistry of alloxan-like compounds, revising previously accepted structure **2**. The reaction of alloxan (**3**) with urea under various experimental conditions[4,5,8] provided the necessary data for solving this structural problem. In accordance with Biltz's observations,[4c] product **4a**, mp 167–168 °C dec, is formed best by the reaction of an excess of urea with **3** in the presence of bromine and analyses for a hemihydrate $C_5H_6N_4O_5 \cdot {}^1/_2H_2O$. Its infrared spectrum in potassium bromide showed a sharp peak at 1808 cm^{-1}, along with three intense absorption bands in the 1780–1700-cm^{-1} region, characteristic of the hydantoin ring. The mass spectrum of **4a** showed no molecular ion at *m/e* 202, and only the fragment due to loss of water appeared at *m/e* 184. From the occurrence of significant metastable peaks at *m/e* 132.4, 108.1, and 90.6, it follows that the subsequent loss of CO and HNCO from *m/e* 184 gave fragments at *m/e* 156 and 141, respectively. The elimination of CO from *m/e* 141 gave ion at *m/e* 113.

The hemihydrate **4a** is smoothly dehydrated by treatment with acetic anhydride,[8] and subsequent heating with trifluoroacetic anhydride afforded spirodihydantoin **5**. The infrared spectra of **4a** and **5** showed similar features; a sharp absorption band at 1804 cm^{-1} characteristic of the hydantoin system was also present in the spectrum of **5**. The mass spectrum exhibited a fragmentation pattern similar to that of **4a**; *m/e* 184 ($M^{+\cdot}$) → *m/e* 156, and *m/e*

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(2) J. H. Lister in "The Chemistry of Heterocyclic Compounds", Vol. 24, Part II, D. J. Brown, Ed., Wiley, New York 1971, pp 239–250.

(3) H. Biltz and F. Max, *Chem. Ber.*, **54**, 2451 (1921).

(4) (a) H. Biltz and M. Heyn, *Chem. Ber.*, **45**, 1677 (1912); (b) H. Biltz, *ibid.*, **43**, 1511 (1910); (c) H. Biltz and M. Heyn, *ibid.*, **47**, 459 (1914).

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(6) (a) H. Biltz and E. Topp, *Chem. Ber.*, **44**, 1524 (1911); (b) H. Biltz and M. Heyn, *ibid.*, **52**, 774 (1919); (c) H. Biltz, *Justus Liebigs Ann. Chem.*, **368**, 170 (1909).

(7) (a) A. Lazarow, *Physiol. Rev.*, **29**, 48 (1949). (b) Reports have been published that a number of enzymes are capable of catalyzing the oxidation of uric acid to **3**: G. Soberon and P. P. Cohen, *Arch. Biochem. Biophys.*, **103**, 331 (1963), and references cited therein. (c) M. Griffiths, *J. Biol. Chem.*, **184**, 289 (1950), claimed that the administration of **1a** in glutathione depleted rabbits caused diabetes. (d) We found no diabetogenic activity for **4a**: M. Poje and B. Ročić, *Experientia*, submited for publication.

(8) R. Behrend and R. Zieger, *Justus Liebigs Ann. Chem.*, **410**, 337 (1915).

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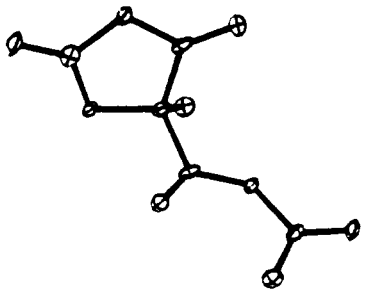


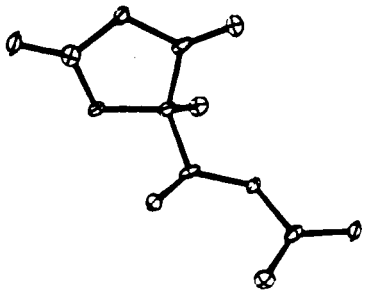
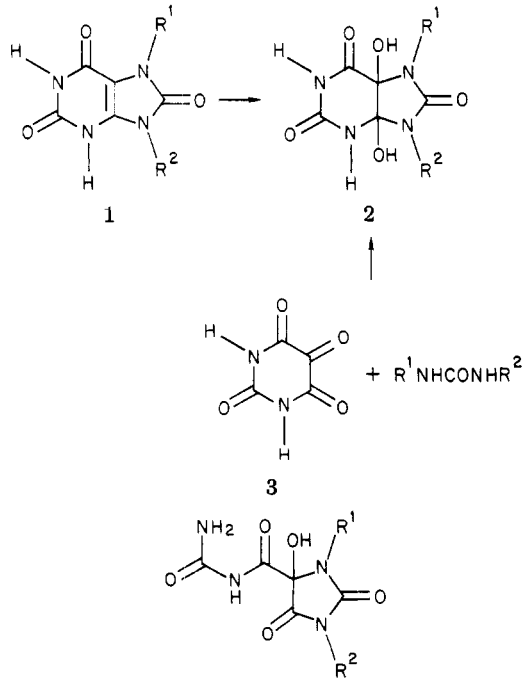


Figure 1. Stereoscopic view of the molecule **4a**.

Scheme I



4a, $R^1 = R^2 = H$
b, $R^1 = H$; $R^2 = Me$
c, $R^1 = H$; $R^2 = Et$
d, $R^1 = R^2 = Me$
e, $R^1 = R^2 = Et$

184 → *m/e* 141 → *m/e* 113.

Further evidence bearing on the structure 4 comes from the reaction with *o*-phenylenediamine or its hydrochloride which afforded the same product, 3-hydroxyquinoxaline-2-carboxyureide (**6**). The failure of **4a** to undergo condensation into an alloxazine under acid conditions ruled out the alloxan-like structure **2a**.[9]

The structure **4a** is also consistent with the facile acid hydrolysis which afforded urea and alloxanic acid (**7**). The infrared spectrum of **7** also exhibited the characteristic sharp peak at 1800 cm^{-1}. The interpretation of all the degradative evidence so far adduced, taken in conjunction with the spectral characteristics of products, led us to propose the structure 4, which certainly accounts for the hydantoin ring, as well as the relative positions of the tertiary hydroxyl group and the ureido function.

The structure **4a** was confirmed by direct single-crystal X-ray structure analysis of its hemihydrate. The asymmetric unit of the crystal contains two enantiomorphic molecules (1 and 2), linked by hydrogen bonding through a molecule of water of hydration. Atomic parameters defining the crystal structure, bond lengths and angles,

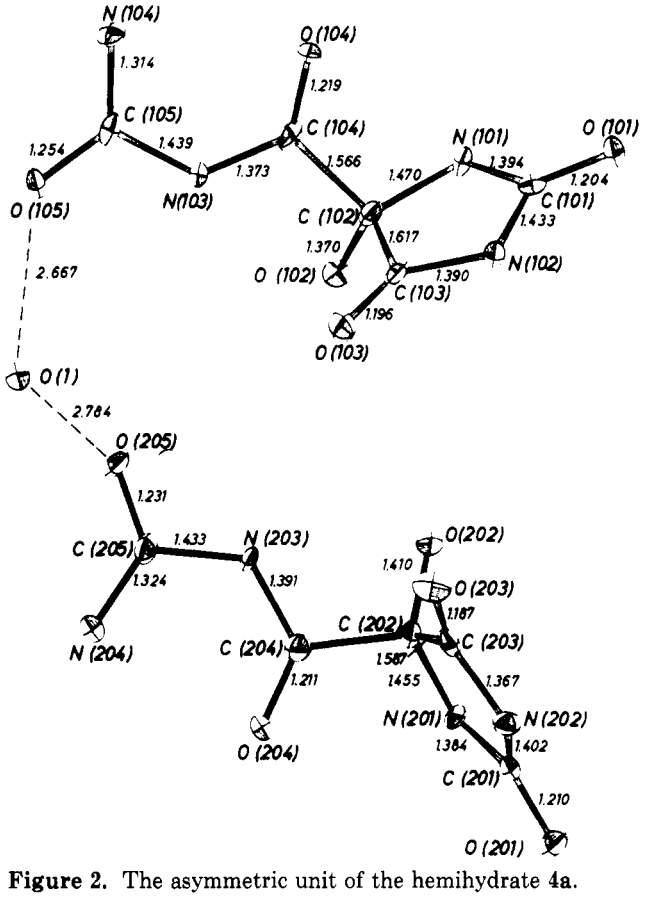


Figure 2. The asymmetric unit of the hemihydrate **4a**.

torsion angles, least-squares planes of interest, and intra- or intermolecular hydrogen bondings are given in the supplementary material. A stereoscopic view of one of the two molecules (1) is shown in Figure 1. The asymmetric unit of the hemihydrate **4a** is depicted in Figure 2, and packing in the unit cell is shown in Figure 3.

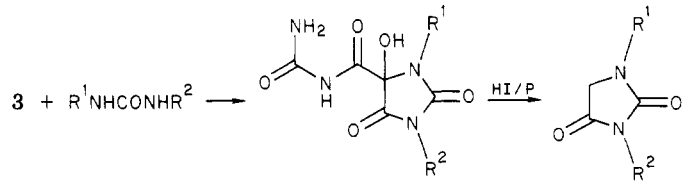
The constitution revealed by the X-ray crystallographic study of the parent compound **4a** leaves no doubt that the correct structures for at least four related compounds shown in Table I are **4b–e**. The presence of the hydantoin system is substantiated by the appearance of a sharp infrared absorption peak in the 1800-cm^{-1} region. The conversion into hydantoins **8a–e** by reaction with hydriodic acid and red phosphorus further strengthens the structural assignment (Table I).

(9) (a) O. Hinsberg, *Chem. Ber.*, 18, 1228 (1885); *Justus Liebigs Ann. Chem.*, **292**, 247 (1896); R. B. Barlow, H. R. Ing, and I. M. Lewis, *J. Chem. Soc.*, 3242 (1951); (b) O. Kühling, *Chem. Ber.*, **24**, 2363 (1891); *ibid.*, **27**, 2116 (1894); R. Kuhn and F. Weygand, *ibid.*, **67**, 1904 (1934).

(10) Tracer studies showed that the nitrogen shift takes place during rearrangement of alloxan (**3**): H. Kwart and I. Sarasolin, *J. Am. Chem. Soc.*, **83**, 1909 (1961); H. Kwart, R. W. Spayd, and C. J. Collins, *ibid.*, **83**, 2579 (1961).

(11) The structure 4 was originally assigned to poorly characterized products obtained by heating aqueous solutions of "uric acid glycols": H. Biltz and E. Topp, *Chem. Ber.*, **44**, 1511 (1911); *ibid.*, **46**, 1398 (1913); see also ref 4a and 4b. The structural revision was affected by extension of these compounds derived from **4**, and consequently interpretation of the chemistry in this series must be revised.

Table I. 4-Hydroxy-2,5-dioxo-4-imidazolidinecarboxyureides (4) and Corresponding Hydantoins 8



4 8

4[a,h]	yield,[b] %	mp, °C[c]	IR, cm^{-1}	8[a,h]	yield,[b] %	mp (bp), °C	recrystn solvent
a	75	166-168[d]	1808	a	44	220-221[d]	water
b	83	207-209[e]	1798	b	52	185-186[f]	ethanol
c	88	198-199[e]	1797	c	55	101-102[f]	CCl_4
d	83	176-178[e]	1800	d	55	44-45[f]	ether
e	93	112-114[e]	1793	e	58	(260-263)[g]	

[a] Satisfactory combustion analytical data (±0.3% for C, H, N) were obtained for all compounds listed in Table I. [b] Yields are based on recrystallized (distilled) products. [c] Decomposition accompanied melting. [d] Reference 4c. [e] Reference 4b. [f] Reference 13a. [g] Reference 13b. [h] a, $R^1 = R^2 = H$. b, $R^1 = H; R^2 = Me$. c, $R^1 = H; R^2 = Et$. d, $R^1 = R^2 = Me$. e, $R^1 = R^2 = Et$.

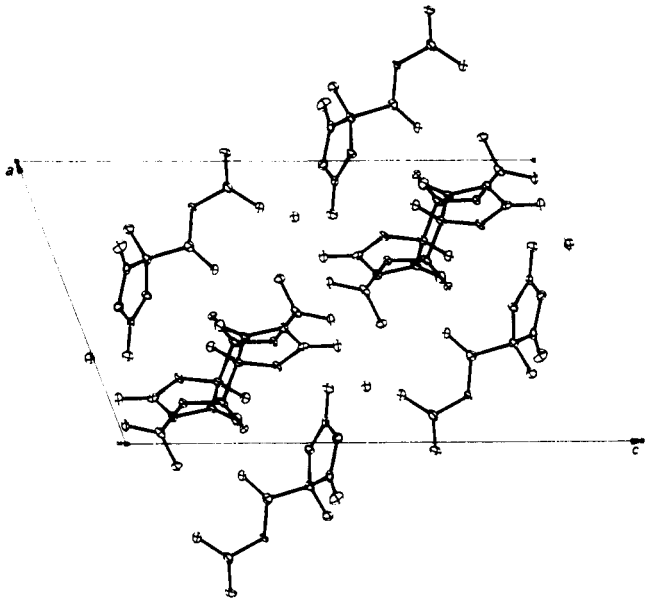
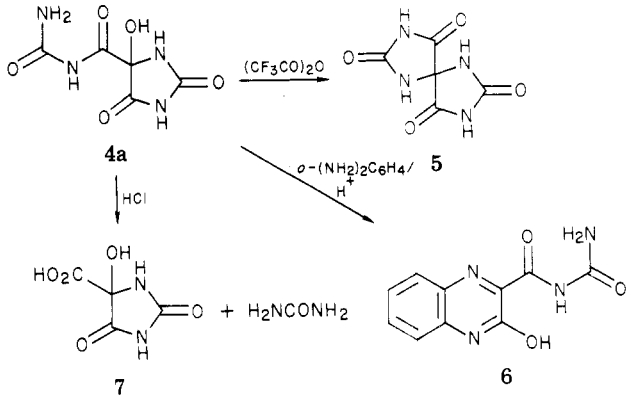


Figure 3. Packing diagram. The direction of projection is down *b*.

The formation of **4** from alloxan (**3**) and urea may be interpreted as a transamidation process which, obviously, does not involve an alloxanic acid-type rearrangement.[10] The reaction may be considered to follow the attack on the 5- and/or 4-position of **3** by urea, the subsequent transamidation results in pyrimidine ring fission and recyclization; the driving force is the stability of the hydantoin system in the outcome. The mechanism could operate with equal plausibility via an intermediate glycol **2**, but this cannot as yet be assessed on the strength of the available experimental material. The mechanistic analogy for this mode of alloxan ring opening exists in the chemistry of riboflavine.[9] The ureide of 3-hydroxyquinoxaline-2-carboxylic acid (**6**) is formed when alloxan (**3**) is treated with *o*-phenylenediamine, the latter being sufficiently basic to cleave the alloxan ring.[9a] If the reaction is carried out in the presence of an equivalent of mineral acid, the ring remains intact and the product is alloxazine.[9b] Similarly, **3** and urea failed to yield **4** in the presence of an acid.

In summary, the revision of the structure clarifies several previously anomalous features in the chemistry of these compounds.[11] It is obvious, for example, why they do not show properties of an alloxan-like compound and do not undergo hydrolysis to their original components or reduction to the parent uric acids (**1**), as might be expected from the uric acid glycol (**2**) formulation. Furthermore, this structural revision implies a more thorough reinvestigation of the oxidation of uric acid (**1**). Work is presently in progress in this laboratory to determine the structures of alloxan-like compounds derived from **1**, as well as to study their chemistry and in vivo activity.

Scheme II



Experimental Section

Melting points were determined, using a Kofler microscope, and are corrected. Infrared spectra were recorded on a Perkin-Elmer 257 spectrometer as KBr disks. Ultraviolet spectral data were obtained with a Perkin-Elmer 124 spectrophotometer. Mass spectra were determined on a Varian MAT CH-7 instrument at 70 eV and 100 μA.

Preparation of the Hemihydrate of 4-Hydroxy-2,5-dioxo-4-imidazolidinecarboxyureide (4a). Alloxan tetrahydrate (**3**, 42.8 g, 0.2 mol) was dissolved in water (90 mL) at 70 °C, and a solution of urea (17 g, 0.28 mol) in water (30 mL) was added with stirring. Bromine (3 g) was gradually added to the clear solution, and the reaction mixture was allowed to stand at room temperature, according to Biltz's procedure.[4c] The crystalline product (38 g, 90%), mp 165–168 °C dec, was collected. Recrystallization from water afforded **4a** as colorless prisms: mp 167–168 °C dec (lit.[4c] 165–168 °C dec); IR (KBr) 3450, 3390, 3350, 3330, 1808, 1777, 1745, 1710, 1585, 1425, 1385, 760 cm^{-1}; UV (water) 205 (ε 8300), 240 (sh) (ε 500) nm; MS (140 °C) *m/e* (rel intensity) 184 ($M^+ \cdot - H_2O$, 38.7), 156 (12.0), 141 (12.7), 129 (4.0), 114 (11.9), 113 (44.8), 100 (11.3), 85 (10.0), 70 (25.1), 69 (22.0), 60 (4.5), 55 (13.3), 54 (10.1), 44 (62.7), 43 (68.0), 42 (100.0).

Anal. Calcd for $C_5H_6N_4O_5 \cdot {}^1/_2H_2O$ (211.14): C, 28.44; H, 3.34; N, 26.54. Found: C, 28.35; H, 3.42; N, 26.33.

The product obtained in a 30% yield by slow evaporation of the aqueous solution of **3** and an excess of urea, according to a previously described procedure,[4a,5] was identical in all respects

with **4a**. The reaction of **3** with urea in the presence of sodium carbonate[8] also afforded **4a** in a 45% yield. The reaction failed, however, in the presence of an acid.

Preparation of Anhydrous 4a. A suspension of finely powdered **4a** hemihydrate (8.44 g, 0.04 mol) in acetic anhydride (40 mL) was refluxed for 1 min. The colorless powder was collected and then in turns covered with absolute ethanol (30 mL), filtered off, washed with an excess of dry ether, and dried in vacuo to yield anhydrous **4a** (6.86 g, 85%): mp 185–188 °C dec (lit.[8] mp 185–186 °C dec); IR (KBr) 1808 cm^{-1}.

Anal. Calcd for $C_5H_6N_4O_5$ (202.14): C, 29.71; H, 2.99; N, 27.72. Found: C, 29.60; H, 3.03; N, 27.66.

5,5′-Spirodihydantoin (5). A suspension of finely powdered **4a** (2.02 g, 0.01 mol) in trifluoroacetic anhydride (100 mL) was heated under reflux until it dissolved. The solvent was distilled off and the oily residue covered with methanol (20 mL). The product was recrystallized from water to yield **5** (1.0 g, 54%) as colorless prisms, which do not melt below 350 °C (lit.[12] mp > 330 °C): IR (KBr) 3495, 3470, 3370, 3220, 1804, 1763, 1738, 1670, 1565, 1440, 1400, 1288, 1263, 1198, 1078, 970, 728 cm^{-1}; UV (water) 205 (sh) (ϵ 6000) nm; MS (200 °C) *m/e* (rel intensity) 184 (M^+·, 19.3), 156 (9.7), 155 (9.3), 141 (9.0), 129 (70.0), 114 (5.7), 113 (24.3), 100 (12.0), 86 (10.7), 85 (6.0), 70 (19.3), 69 (11.7), 60 (4.6), 55 (6.0), 44 (100.0), 43 (95.0).

Anal. Calcd for $C_5H_4N_4O_4$ (184.12): C, 32.62; H, 2.19; N, 30.43. Found: C, 32.51; H, 2.20; N, 30.55.

Reaction of 4a with *o*-Phenylenediamine. 3-Hydroxyquinoxaline-2-carboxyureide (6). *o*-Phenylenediamine (0.6 g, 0.0055 mol) was added to a suspension of finely powdered **4a** (1.01 g, 0.005 mol) in glacial acetic acid (10 mL) and the reaction mixture refluxed for 3 h. Greenish-yellow crystals which separated on cooling were collected. Recrystallization from acetic acid afforded **6** (0.9 g, 77%): mp 242–244 °C dec (lit.[9a] mp 240–50 °C dec); IR (KBr) 3390, 3160, 1725, 1695, 1650, 1578, 1503, 1477, 1440, 1395, 770 cm^{-1}. Identical results were obtained when **4a** was treated with *o*-phenylenediamine dihydrochloride (1.0 g, 0.0055 mol) in glacial acetic acid (20 mL).

Anal. Calcd for $C_{10}H_8N_4O_3$ (232.21): C, 51.73; H, 3.47; N, 24.13. Found: C, 51.85; H, 3.51; N, 23.91.

Hydrolysis of 4a. Urea and Alloxanic Acid (7). A solution of **4a** (1.06 g, 0.005 mol) in concentrated hydrochloric acid (10 mL) was evaporated to dryness in vacuo. Finely powdered residue was dried in vacuo (10^{-3} Torr) over potassium hydroxide for 48 h and then in turns dissolved in water (10 mL), carefully neutralized with 5% barium hydroxide solution, and kept at 1 °C overnight. Crystalline precipitate (1.6 g) was filtered off, and an additional amount (0.2 g) was isolated by concentration of the filtrate. The clear filtrate was passed through a column of Dowex 50W (20–50 mesh, H^+ form, 3 g) and the resin washed with water (45 mL) and filtered through Amberlite MB 3 (20–50 mesh, H^+ and OH^- form, 1 g). The filtrate was evaporated to dryness in vacuo at a temperature not above 30 °C to yield urea (0.1 g, 33%), mp 131–133 °C.

The suspension of the barium salt of **7** in water (60 mL) was stirred with Amberlite IR 120 (28–35 mesh, H^+ form, 20 g) at 5 °C for 8 h. The resin was separated and washed with water (40 mL) and the aqueous solution evaporated to dryness in vacuo (30 °C). The residue was dissolved in absolute ethanol and filtered through charcoal. Crystalline alloxanic acid (**7**, 0.68 g, 85%) was deposited from concentrated ethanolic solution after several days at −5 °C. Repeated crystallizations from dry ether afforded colorless prisms of **7**: mp 162–163 °C dec (lit.[8] mp 162–163 °C dec); IR (KBr) 3410, 3380, 3235, 1800, 1760, 1740, 1723, 1290, 1267, 1167, 762, 740 cm^{-1}.

Anal. Calcd for $C_4H_4N_2O_5$ (160.09): C, 30.01; H, 2.52; N, 17.50. Found: C, 29.74; H, 2.81; N, 17.19.

General Procedure for the Preparation of 4a–e. The procedure of Biltz[4a] was used on a 0.05 mol scale with slight modifications. A finely powdered mixture of **3** (0.05 mol) and the appropriate urea derivative (0.05 mol) was dissolved in a minimal amount of water at 90 °C and the solution left overnight at room temperature. The crystalline product was collected and an additional quantity of substance isolated by concentration of the mother liquor in vacuo. Recrystallization from water afforded pure compounds **4a–e**. The relevant physical and spectral data are listed in Table I.

General Procedure for the Preparation of Hydantoins 8a–e. The modified procedure described by Biltz[13] was used on the 0.02 mol scale. Finely powdered **4** was added to a stirred mixture of constantly boiling hydriodic acid (15 g) and red phosphorus (0.9 g). The reaction mixture was stirred in a steam bath for 3 h, cooled, and filtered through a sintered-glass funnel. The clear filtrate was evaporated to dryness at a reduced pressure. The residue was crystallized from an appropriate solvent or distilled (Table I) to give hydantoins **8a–e**.

Crystallographic Data and X-Ray Structure Analysis. A parallelepiped-shaped crystal (0.50 × 0.20 × 0.01 mm) of 4-hydroxy-2,5-dioxo-4-imidazolidinecarboxyureide (**4a**) was used for the X-ray structure analysis. Lattice dimensions were determined by an automatic Siemens AED diffractometer, using Mo Kα (λ = 0.7107 Å) radiation. Crystal data: crystals of **4a**, $C_5H_6N_4O_5 \cdot {}^1/_2H_2O$, are monoclinic; space group $P2_1/n$; a = 10.558 (7), b = 9.072 (14), c = 17.905 (10) Å, β = 110.47 (5)°; V = 1606.6 $Å^3$; Z = 8; M = 211.14; ρ_c = 1.746 g cm^{-3} (two molecules of **4a** and a molecule of water per asymmetric unit); ρ_o = 1.72 g cm^{-3} (flotation in bromoform/dichloromethane).

Intensity data were measured, using Mo Kα (λ = 7107 Å, θ_{max} = 28°) radiation. A total of 3860 reflections were collected with 1730 considered observed, based on the criteria $I > \sigma(I)$. Absorption corrections were not necessary, because the absorption coefficient is very small. The structure was solved by direct methods[14] and refined by full-matrix least-squares procedure to R_1 = 0.108 and R_2 = 0.076 for 1730 independent observed reflections. Hydrogen atom locations could not be found in a difference Fourier map and were not considered further.

Registry No. **3**, 50-71-5; **4a**, 71886-22-1; **4b**, 71886-23-2; **4c**, 71886-24-3; **4d**, 71886-25-4; **4e**, 71886-26-5; **5**, 6541-63-5; **6**, 6275-81-6; **7**, 470-44-0; **7**-Ba, 71886-27-6; **8a**, 461-72-3; **8b**, 6843-45-4; **8c**, 2221-20-7; **8d**, 24039-08-5; **8e**, 71886-28-7; urea, 57-13-6; *o*-phenylenediamine, 95-54-5; *o*-phenylenediamine dihydrochloride, 541-69-5; methylurea, 598-50-5; ethylurea, 625-52-5; *N,N′*-dimethylurea, 96-31-1; *N,N′*-diethylurea, 623-76-7.

Supplementary Material Available: Tables II–VI listing positional and thermal parameters, bond lengths, and angles with their estimated standard deviations, torsion angles, intra- and intermolecular hydrogen bonding and information on least-squares mean planes of interest (4 pages). Ordering information is given on any current masthead page.

(12) H. Biltz, M. Heyn, and M. Bergius, *Justus Liebigs Ann. Chem.*, **413**, 77 (1916).

(13) (a) H. Biltz and M. Heyn, *Chem. Ber.*, **45**, 1666 (1912); (b) H. Biltz and F. Max, *Justus Liebigs Ann. Chem.*, **423**, 288 (1921).

(14) G. Germain, P. Main, and M. M. Woolfson, *Acta Crystallogr., Sect. B*, **26**, 274 (1970); P. Main, L. Lessinger, M. M. Woolfson, G. Germain, and J.-P. Declercq, "Multan 77. A System of Computer Programmes for the Automatic Solution of Crystal Structures from X-Ray Diffraction Data", University of York and Institute Lavoisier (Louvain-la-Neuce), 1977.