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(12) The procedure outlined in ref 11 utilizes an open beaker for the peroxy acid preparation. We found this procedure to be unsatisfactory on humid days, yielding peroxy acid samples with low active oxygen content.
(13) The temperature of the reaction mixture should be carefully monitored. If the mixture is heated above 53 °C a steady rise in the internal reaction temperature may be seen. The exotherm may lead to frothing, spillage, and peroxy acid batches with low active oxygen content.

Cyanohydrin Synthesis of 2,3-Dihydroxy-2,3-dimethylbutanoic Acid

Jack E. Powell,* Carl Osuch, Harvey R. Burkholder, Santi Kulprathipanja, James H. Miller, Leon G. Stadtherr, and Russell G. Baughman

Ames Laboratory, DOE and Department of Chemistry, Iowa State University, Ames, Iowa 50011

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From 3-hydroxy-3-methyl-2-butanone (**1**) via cyanohydrin synthesis and subsequent hydrolysis, the intermediates 2,3-dihydroxy-2,3-dimethylbutanonitrile (**2**), 3-chloro-1,2-dihydroxy-2,3-dimethylbutanimine hydrochloride (**3**), 3-chloro-1,2-dihydroxy-2,3-dimethylbutanamide (**4**), and 2,3-dihydroxy-2,3-dimethylbutanamide (**5**) have been isolated en route to 2,3-dihydroxy-2,3-dimethylbutanoic acid (**6**). Compound **2** reverted to **1** in the presence of base. In aqueous NaOH or NaOMe in Et_2O, compounds **3** and **4** gave (by HCl abstraction) 2,3-epoxy-1-hydroxy-2,3-dimethylbutanimine (**7**), tautomeric with 2,3-epoxy-2,3-dimethylbutanamide (**8**). Acid hydrolysis of **2** (at 40–50 °C) led principally to **5**, but at higher temperatures to 3-methyl-2-butanone (**9**) via a pinacol–pinacolone type rearrangement involving the intermediates 2,2-dimethyl-3-oxobutanamide (**10**) and 2,2-dimethyl-3-oxobutanoic acid (**11**), which decarboxylates spontaneously to **9**. In the acid hydrolysis of **2** to obtain **5** and **6** directly, substantial amounts of the byproduct 2-hydroxy-2,3-dimethyl-3-butenoic acid (**12**) were encountered; better yields of the desired products were obtained when the dihydroxynitrile (**2**) was first treated with 2 mol of acetic anhydride per mole to form its diacetate and somewhat diluted hydrochloric acid was used in lieu of saturated aqueous HCl.

Interest in the effects of adding a second methyl group to the β-carbon atom of 2,3-dihydroxy-2-methylbutanoic acid on the acid ionization constant and the chelating properties of the ligand moiety prompted an attempt to synthesize 2,3-dihydroxy-2,3-dimethylbutanoic acid from 3-hydroxy-3-methyl-2-butanone (via a route used in preparing 2,3-dihydroxy-2-methylpropanoic acid and 2,3-dihydroxy-2-methylbutanoic acid from acetol and acetoin precursors, respectively[1,2]). After several failures to obtain the expected amide and acid from unisolated cyanohydrin, using standard procedures, it was decided to perform a step-by-step isolation (by ion-exclusion chromatography and anion exchange when appropriate) of the various intermediates, in order to ascertain at what point the process failed.

While no one (to date) has reported the synthesis of either 2,3-dihydroxy-2,3-dimethylbutanamide (DHDMB amide) or DHDMB acid, Cantacuzène and Ricard[3] prepared the corresponding DHDMB nitrile by acid hydrolysis (dilute H_2SO_4) of 2,3-epoxy-2,3-dimethylbutanonitrile and reported its ^1H NMR spectra in $CDCl_3$, benzene, and DMF and its IR spectrum in CCl_4. Since they failed to obtain the nitrile in crystalline form, only its boiling point [130 °C (15 Torr)] was given.

It was immediately ascertained that DHDMB nitrile could be prepared in good yield from the KCN-catalyzed combination of 3-hydroxy-3-methyl-2-butanone and excess liquid HCN (the reaction temperature being controlled at ~30 °C by refluxing of the HCN). The cyanohydrin (DHDMB nitrile) was readily obtained as a white crystalline solid (mp 67–69 °C) from ethyl acetate, whose ^1H NMR spectrum in $CDCl_3$ coincided with the liquid prepared by Cantacuzène and Ricard[3] from 2,3-epoxy-2,3-dimethylbutanonitrile. Typical of cyanohydrins, our DHDMB nitrile yielded the original ketone and NaCN (instantaneously and quantitatively) when treated with excess aqueous NaOH.

Acid hydrolysis of the DHDMB nitrile posed a problem in that undesired dark byproducts were obtained copiously at elevated temperatures, and conversion of the nitrile was inordinately slow in concentrated hydrochloric acid or dilute acid at room temperature. When the nitrile was dissolved in hydrochloric acid and saturated with HCl gas below 35 °C (a standard procedure), the tertiary 3-hydroxyl was replaced by chloride, and 3-chloro-1,2-dihydroxy-2,3-dimethylbutanimine hydrochloride (rather than the expected DHDMB amide) resulted. This hydrochloride (upon recovery and washing with ether) decomposed spontaneously at room temperature (over a 24-h period) to the corresponding 3-chloro-2-hydroxy-2,3-dimethylbutanamide by evolving HCl.

Dilute (~1 M) aqueous solutions of 3-chloro-2-hydroxy-2,3-dimethylbutanamide (CHDMB amide) slowly generate H_3O^+ via hydrolysis (replacement of the tertiary –Cl by –OH). The resulting DHDMB amide then presumably undergoes very slow hydrolytic conversion to DHDMB acid. At temperatures as low as 80 °C, when either CHDMB amide or DHDMB amide is hydrolyzed in dilute HCl, CO_2 is evolved at an appreciable rate and the major product isolated (and positively identified by its ^1H NMR spectrum) is 3-methyl-2-butanone. The characteristic odor of this ketone could be detected after a day even in conversions carried out at 45 °C. Isolation of 2,2-dimethyl-3-oxobutanamide (mp 120–122 °C),[4] whose oxime melts at 162–164 °C,[5] in the acid hydrolysis of both CHDMB amide and DHDMB amide indicates that DHDMB amide readily undergoes a pinacol–pinacolone type of rearrangement. As the resulting 2,2-dimethyl-3-oxobutanamide hydrolyzes to 2,2-dimethyl-3-oxobutanoic acid, decarboxylation of this unstable substance occurs. The chief product obtained is 3-methyl-2-butanone.

If 3-chloro-2-hydroxy-2,3-dimethylbutanamide in aqueous solution is treated with excess base, abstraction of HCl, rather than replacement of –Cl by –OH, occurs. An epoxyhydroxy-

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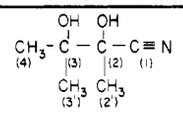
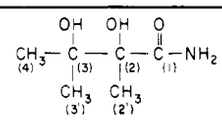
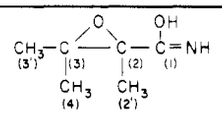
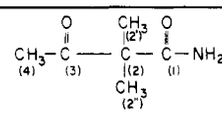
Table I. Melting Points and ^1H NMR Chemical Shifts of 2,3-Dihydroxy-2,3-dimethylbutanoic Acid and Derivatives

compd	registry no.	mp, °C	^1H NMR[a]	
2	26429-38-9	67–69	$CDCl_3$:	1.30 (s, 3 H), 1.44 (s, 3 H), 1.51 (s, 3 H), 2.87 (b, 1 H), 4.03 (b, 1 H)
			D_2O:	1.31 (s, 6 H), 1.57 (s, 3 H)
4	66483-60-1	139–141	$CDCl_3$:	1.57 (s, 3 H), 1.68 (s, 3 H), 1.74 (s, 3 H)
			D_2O:	1.51 (s, 3 H), 1.68 (s, 6 H)
5	66483-61-2	111–113	$CDCl_3$:	1.29 (s, 6 H), 1.46 (s, 3 H)
			D_2O:	1.22 (s, 6 H), 1.38 (s, 3 H)
6	66483-62-3	102–104	$CDCl_3$:	1.34 (s, 6 H), 1.48 (s, 3 H)
			D_2O:	1.27 (s, 6 H), 1.42 (s, 3 H)
7	66483-63-4	157–158	$CDCl_3$:[b]	1.379 (s, 6 H), 1.545 (s, 3 H), 6.299 (s, 2 H)
		[lit.[6] 157–158][c]	CCl_4:	1.34 (s, 6 H), 1.44 (s, 3 H)
10	66483-64-5	122–124	$CDCl_3$:[b]	1.408 (s, 6 H), 2.333 (s, 3 H), 6.136 (s, 2 H)
		[lit.[4] 121–122]	D_2O:	1.39 (s, 6 H), 2.25 (s, 3 H)
12	17891-10-0	85–57	$CDCl_3$:	1.61 (s, 3 H), 1.83 (d-d, 3 H), 5.02 (d-d, 1 H) 5.21 (t, 1 H), 7.41 (b, 2 H)
		[lit.[7] 87–88]		[lit.:[7] 1.59 (s), 1.82 (d-d), 5.03 (q), 5.22 (q), 7.52 (s)]

[a] Chemical shift in ppm relative to Me_4Si ($CHCL_3$, CCl_4) and DSS (D_2O). [b] Recorded at 90 MHz on Brucker HX-90 spectrophotometer. [c] Previously identified as a different compound; see text.

Table II. ^{13}C Chemical Shifts[a]

COMPOUND	STRUCTURE	C-1	C-2	C-3	C-4	C-3'	C-2'	C-2''
2		121.406	74.495	75.210	22.517 & 22.387		25.051	—
5		180.855	78.458	74.430	26.220 & 24.401		22.127	—
7		174.359	65.464	63.710	20.308		15.565	—
10		175.008	55.978	208.859	26.090	—	22.452	

[a] In ppm from internal Me_4Si.

imine, less soluble than the CHDMB amide, is obtained. The identity of this substance was at first puzzling, as the ^1H NMR (in CCl_4, $CDCl_3$) showed only two peaks with a ratio of 2:1. All of the NMR data are summarized in Tables I and II. The compound was eventually determined by X-ray diffraction to be 2,3-epoxy-1-hydroxy-2,3-dimethylbutanimine.

This epoxyhydroxyimine appears to be identical with the substance (mp 157–158 °C) obtained by Delbaère[6] on heating 2,3-epoxy-2,3-dimethylbutanonitrile a few minutes with dilute aqueous NaOH (mistakenly identified by Delbaère as 2,3-dihydroxy-2,3-dimethylbutanonitrile). It is now apparent that rapid hydrolysis of the epoxynitrile to the imine occurred in Delbaère's reaction, rather than opening of the ether linkage.

Another substance isolated in substantial amounts in attempts to convert 3-chloro-2-hydroxy-2,3-dimethylbutanamide to 2,3-dihydroxy-2,3-dimethylbutanoic acid (which helps to confirm the position of the Cl in CHDMB amide) was the known compound 2-hydroxy-2,3-dimethyl-3-butenoic acid.

Formation of CHDMB amide, 2-hydroxy-2,3-dimethyl-3-butenoic acid, 2,2-dimethyl-3-oxobutanamide, 3-methyl-2-butanone, and unidentified dark-colored substances is repressed by converting the isolated nitrile to its diacetate ester and utilizing somewhat diluted (~8 M) hydrochloric acid at 50 °C in converting DHDMB nitrile to DHDMB amide. Conversion under these conditions requires about 40 h.

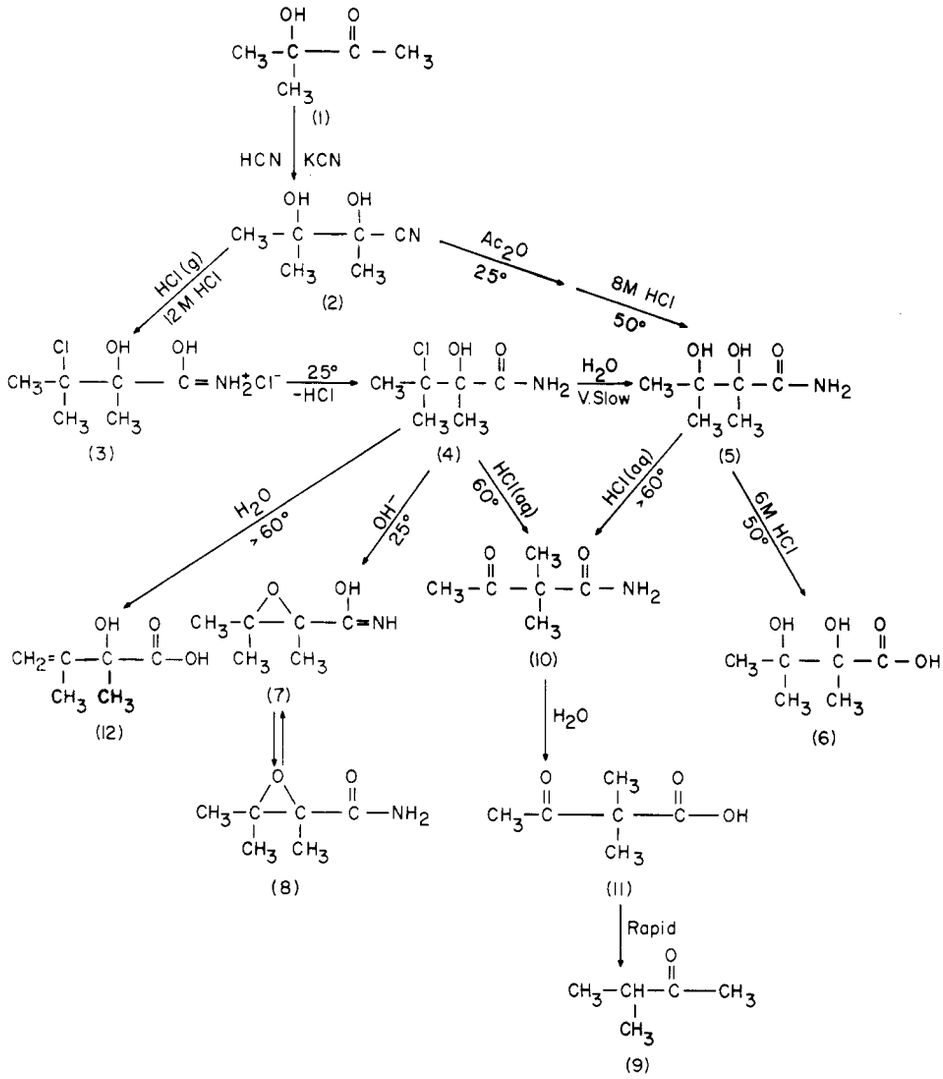
Although DHDMB acid was obtained by basic hydrolysis of DHDMB amide, better yields were obtained via slow hydrolysis in 6 M HCl at 50 °C (2 weeks).

Experimental Section

The melting points were taken on an electrothermal melting point apparatus and are uncorrected. The proton magnetic resonance spectra were measured on a Varian A-60 or Hitachi Perkin-Elmer R-20 B spectrometer. The ^{13}C NMR spectra were recorded on a Brucker HX-90 spectrometer. Some ^1H spectra were also obtained using pulsed Fourier transform at 90 MHz on the Brucker HX-90. All chemical shifts were measured in parts per million relative to internal Me_4Si (or DSS in D_2O).

2,3-Dihydroxy-2,3-dimethylbutanonitrile (2, see Scheme I). 3-Hydroxy-3-methyl-2-butanone (1; 1 mol, 120 g of 85%) at 0 °C was mixed with 5 mol (135 g) of freshly prepared, chilled, anhydrous, liquid HCN (in a 3-L, three-neck, round-bottom flask equipped with an ice-water-cooled reflux condenser, a thermometer, and a magnetic stirrer and immersed in an ice bath *in an efficient hood*). Solid KCN catalyst (0.1 g) was added, and the assembly was raised just out of the bath to initiate the reaction. When the temperature began to rise rapidly (at about 15 °C) the assembly was lowered to touch the ice bath, after which the reaction continued at the reflux temperature (~30 °C). When (upon seeding) solid cyanohydrin (2) separated copiously (in the course of about 30 min), an additional 3.2 mol of chilled 1 was added at a rate sufficient to maintain the reaction at 30–35 °C. Nitrile 2 accumulated eventually to the extent that the magnetic stirrer could no longer handle the load. At this point the reversible reaction was stopped by adding 8 mL of glacial acetic acid in 500 mL

Scheme I. Relationships between Intermediate, Products, and Byproducts Observed.



of ethyl acetate. The flow of ice water in the reflux condenser was then stopped, and the flask was gently warmed in a heating mantle to distill off excess HCN and bring the ethyl acetate to a state of reflux.

After recooling to about 18 °C, the mixture was transferred to an open beaker *in the hood*, seeded with a few crystals of **2**, and then chilled to near 0 °C. The snow-white crystalline product was recovered by filtration, and the filtrate was evaporated first to 300 mL and then to 150 mL to obtain two additional crops of nitrile. The pale-yellow residual liquid was principally **1**, containing a small amount of **2**.

The crystalline cyanohydrin weighed 413 g (3.2 mol = 76% yield) and melted (after recrystallization from ethyl acetate) at 67–69 °C. Anal. Calcd for $C_6H_{11}NO_2$: C, 55.79; H, 8.58; N, 10.84; O, 24.77. Found: C, 55.8; H, 8.5; N, 11.1; O, 24.6. The equivalent weight computed from base-liberated CN^- was 129 (theoretical 129.16). Note: In the open **2** reverts slowly to **1** and HCN.

3-Chloro-1,2-dihydroxy-2,3-dimethylbutanimine Hydrochloride (3). **2** (0.5 mol, 64.5 g) in 63 mL of 12 N HCl was saturated with HCl gas at such a rate that the temperature reached about 35 °C. After 4 h, and a weight gain of 19.9 g, considerable **3** separated. The mixture was allowed to stand overnight and was then resaturated with HCl, giving an additional gain of 11.2 g. It was then allowed to stand for 48 h, at which time a qualitative test revealed no **2** remaining. At this point, the crystalline **3** was filtered off and rinsed with 30 mL of anhydrous ether (5 mL at a time). After a 1-h exposure to the air, the produce weighed 52.5 g and appeared dry (although reeking of HCl).

3-Chloro-2-hydroxy-2,3-dimethylbutanamide (4). In two separate experiments, 25.5 g and 17.0 g of **3** lost 5.3 g and 3.5 g, respectively, on standing 24 h at 23 °C in the open. Recrystallized from ethyl acetate, the residual material melted at 139–141 °C and exhibited an equivalent weight of 166.3, based on an independent analysis for Cl ($C_6H_{12}ClNO_2$ = 165.62). Anal. Calcd for $C_6H_{12}ClNO_2$: C, 43.51; H, 7.30; Cl, 21.40; N, 8.45; O, 19.32. Found: C, 43.4; H, 7.4; Cl, 21.1; N, 8.5; O, 19.6.

2,3-Epoxy-1-hydroxy-2,3-dimethylbutanimine (7). A 3.31-g sample of **4** (0.02 mol), dissolved in 30 mL of diethyl ether, was combined with 0.02 mol of NaOMe in ether. NaCl separated immediately, and after 0.5 h the salt was filtered off and washed with ether. When a vacuum was pulled on the filtrate to reduce the volume, silky-white needles separated from the cold solution. The yield was nearly quantitative, and when recrystallized from ether, ethyl acetate, or water, the product (**7**) melted at 157–158 °C (with sublimation). Anal. Calcd for $C_6H_{11}NO_2$: C, 55.79; H, 8.58; N, 10.84; O, 24.77. Found: C, 55.7; H, 8.6; N, 11.0; O, 24.7.

2,3-Dihydroxy-2,3-dimethylbutanamide (5). When 20.7 g (0.125 mol) of **4** and 26.0 g (0.128 mol) of **3** were dissolved, each in 100 g of H_2O, they slowly generated H_3O^+. In dilute solution the replacement of –Cl by –OH appears to be first order with respect to **4** and independent of the H_3O^+ concentration. The rate constants at 23 and 45 °C are 0.004 and 0.16 h^{-1}, respectively.

After 72 h at room temperature followed by 48 h at 45 °C, the products of the parallel experiments above were combined, diluted to 500 mL, and pumped into a 1-in. system (three, 4-ft beds in series) of −40 + 50 mesh, H^+-form, Dowex 50W-X8, cation-exchange resin (at a rate of 1.25 mL/min). The mixture was then subjected to ion-exclusion separation by eluting the system with deionized water at the same rate. The effluent solution was collected in a series of 30 95-mL fractions. Samples 5–14 contained primarily the expected $^3/_8$ mol of HCl. A weak acid and **5** eluted principally in fractions 14–21. Fractions 22–26 yielded a second nitrogen-containing substance subsequently identified as 2,2-dimethyl-3-oxobutanamide[4] (by its empirical formula and melting point, 121–122 °C). Upon evaporation of fractions 16–20 (under vacuum at 35 °C) to a thick syrup and dissolving this in 15 mL of hot ethyl acetate, 6.0 g of **5** was obtained as

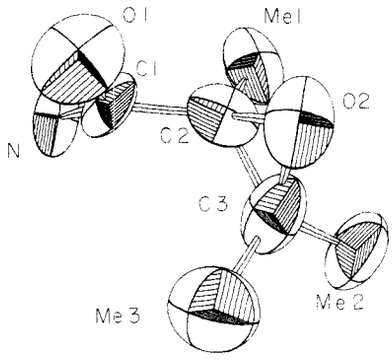


Figure 1. The molecular structure of 2,3-epoxy-1-hydroxy-2,3-dimethylbutanimine, with thermal ellipsoids drawn at the 50% probability level.

a first crop on cooling. Recrystallized from $CHCl_3$, **5** melted at 111–113 °C. The mother liquor from fractions 16–20, after heating with base and removal of the Na^+ by cation exchange, yielded (instead of the expected DHDMB acid (**6**)) 3.2 g of 2-hydroxy-2,3-dimethylbutenoic acid (**12**) melting at 85–87 °C (lit.[7] 87–88 °C).

Subsequently, as one of a series of experiments intended to optimize conversion of **2** to **5**, 0.3 mol of **2** was treated with 0.6 mol of acetic anhydride and a drop of H_2SO_4 to form the diacetate ester of the dihydroxynitrile. Hydrolysis of the CN functional group was then carried out by adding 83 mL of 12 N HCl and 36 mL of H_2O and heating at 40 °C for 15 h. The yield of **5** (isolated by ion exclusion as described above) was 81.6%. The formula weight by base-evolved NH_3 was 147 ($C_6H_{13}NO_3$ = 147.18). Anal. Calcd for $C_6H_{13}NO_3$: C, 48.96; H, 8.90; N, 9.52; O, 32.61. Found: C, 48.4; H, 8.8; N, 9.5; O, 33.3

2,3-Dihydroxy-2,3-dimethylbutanoic acid (6). A 14-g sample of **5** (0.095 mol) was dissolved in 150 mL of H_2O, treated with 40 g (1 mol) of NaOH pellets, and refluxed for 2 h. The mixture was then diluted to 250 mL and passed through a bed containing a 50% excess of −40 + 50 mesh, H^+-form, Dowex 50W-X8, cation-exchange resin. The acidic effluent was collected and evaporated at 40 °C. The residue was recrystallized from $CHCl_3$ to a white monobasic acid (mp 102–104 °C) which exhibited a formula weight of 151.7 ($C_6H_{12}O_4$ = 148.16). Anal. Calcd for $C_6H_{12}O_4$: C, 48.64; H, 8.16; O, 43.20. Found: C, 48.4; H, 8.3; O, 43.3. The ^1H NMR spectrum of the acid (**6**) was indistinguishable from that of the amide (**5**) in D_2O (Table I).

DHDMB acid (**6**) was also obtained via consecutive prolonged acid hydrolyses of the diacetate ester of DHDMB nitrile and DHDMB amide. For example, 300 g of the nitrile (**2**) was first converted to the diacetate ester by adding a slight excess of acetic anhydride and a few drops of H_2SO_4. After 1 h, the reaction mixture was treated with 325 mL of H_2O and 650 mL of concentrated HCl. At 50 °C, the conversion of nitrile to amide was found to be nearly complete in 40 h (conclusion based on qualitative test for cyanohydrin). The mixture was diluted to 4 L, passed onto a system consisting of a series of six 4-in. × 4-ft, H^+-cycle, −40 + 50 mesh, Dowex 50W-X8, cation-exchange beds at a rate of about 1 L/h, and then ion-exclusion eluted with deionized water at the same rate. The fractions free of HCl and containing DHDMB amide (and some **6**) were combined and vacuum evaporated at 50 °C to a solid mass. This mixture was then dissolved in 4 L of 6 N HCl and maintained at 50 °C for 2 weeks. The DHDMB acid was finally isolated along with some unconverted amide by a repetition of the ion-exclusion process above. The amide was next eliminated by adsorbing the acid on an acetate-cycle anion-exchange column, and the acid was recovered by displacing it with 0.25 M HCl. When the residue, obtained by vacuum evaporation of the effluent, was recrystallized from $CHCl_3$, the combined crops of **6** totaled 151 g.

Table III. Selected Interatomic Distances (Å) for 2,3-Epoxy-1-hydroxy-2,3-dimethylbutanimine

C(1)–O(1)	1.41 (2)	C(2)–O(2)	1.463 (27)
C(1)–N	1.17 (3)	C(3)–O(2)	1.416 (34)
C(1)–C(2)	1.53 (3)	C(3)–Me(2)	1.55 (3)
C(2)–Me(1)	1.45 (3)	C(3)–Me(3)	1.54 (4)
C(2)–C(3)	1.485 (33)		

Table IV. Bond Angles (deg) for 2,3-Epoxy-1-hydroxy-2,3-dimethylbutanimine

O(1)–C(1)–N	126.4 (2.5)	O(2)–C(2)–C(3)	57.4 (1.8)
O(1)–C(1)–C(2)	110.8 (2.2)	C(2)–O(2)–C(3)	62.1 (1.4)
N–C(1)–C(2)	122.3 (2.0)	C(2)–C(3)–O(2)	60.5 (1.8)
C(1)–C(2)–Me(1)	114.9 (2.4)	C(2)–C(3)–Me(2)	120.6 (2.7)
C(1)–C(2)–O(2)	117.3 (2.0)	C(2)–C(3)–Me(3)	121.8 (2.4)
C(1)–C(2)–C(3)	115.7 (2.5)	O(2)–C(3)–Me(2)	115.9 (2.5)
Me(1)–C(2)–O(2)	118.4 (2.3)	O(2)–C(3)–Me(3)	114.3 (2.3)
Me(1)–C(2)–C(3)	121.2 (2.7)	Me(2)–C(3)–Me(3)	112.8 (2.8)

X-Ray Crystallographic Data. A single crystal of the compound $C_6H_{11}NO_2$, mp 157–158 °C, d = 1.14, was mounted on a glass fiber and placed on a four-circle diffractometer and submitted to a routine crystal and molecular structure determination. The data fit obtained indicated that the compound should be considered the epoxyhydroxyimine (**7**) rather than the epoxyamide (**8**). The molecular structure is depicted in Figure 1, and pertinent bond angles and distances are listed in Tables III and IV.

Acknowledgments. We would like to thank Dr. R. A. Jacobson and the members of his group for the use of the X-ray diffractometer. This work was supported by U.S. Department of Energy, Division of Basic Energy Sciences.

Registry No.—**1**, 115-22-0; **3**, 66483-65-6.

Supplementary Material Available: Complete details of the structural determination of **7**, including procedure, F tables, and final positional and thermal parameters (3 pages). Ordering information is given on any current masthead page.

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