A Planar Cyclobutane Ring. Structure of *trans*-1,3-Cyclobutanedicarboxylic Acid

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Abstract: Crystals of *trans*-1,3-cyclobutanedicarboxylic acid are monoclinic, space group $P2_1/c$, with $a = 5.452$, $b = 7.813$, $c = 8.131$ A; $\beta = 107.22°$ (two molecules/cell). The crystal structure, determined by X-ray diffraction, contains chains of hydrogen-bonded molecules. The cyclobutane ring is planar with C–C single bond lengths of 1.567 ± 0.006 and 1.552 ± 0.006 A.

The structure of simple cyclobutane derivatives is of interest because two different conformations have been observed. Planar rings have been found in compounds such as tetraphenylcyclobutane,[1,2] the photodimer of cyclopentenone,[3] and tetracyanocyclobutane.[4] Puckered rings have been found in octachlorocyclobutane,[1,5] anemomin,[6] and the *cis* and *trans* isomers of 1,2-dibromo-1,2-dicarbomethoxycyclobutane.[7] Studies in the gas phase and in liquid crystals indicate that cyclobutane,[8,9] bromocyclobutane,[10] and octafluorocyclobutane[11] are also puckered. Another structural problem concerns the length of the C–C single bond in cyclobutane rings. Values as high as 1.60 A have been reported, but in most cases the results have not been of the highest precision. We have determined the crystal structure of *trans*-1,3-cyclobutanedicarboxylic acid in order to provide accurate structural parameters.

Experimental Section

The synthesis of *trans*-1,3-cyclobutanedicarboxylic acid was reported by Deutch and Buchman[12] who showed that the synthesis of Markownikoff and Krestownikoff[13] was incorrect; the latter had prepared 1-methyl-1,2-cyclopropanedicarboxylic acid. An authentic sample of *trans*-1,3-cyclobutanedicarboxylic acid (mp 190°) prepared by Buchman was obtained through the courtesy of Professor K. N. Trueblood of U.C.L.A. A set of X-ray diffraction photographs was collected by the equiinclination Weissenberg technique and used to solve the structure. The final refinement was carried out with data from a Picker manual diffractometer equipped with a full circle goniometer, scintillation counter, and pulse-height analyzer. The crystal used was a tiny block with no dimension greater than 0.1 mm. Intensities were gathered by the stationary-crystal–stationary-counter method using Cu Kα ($\lambda = 1.5418$ A) radiation. The intensities were converted to structure factors with no correction for absorption.

Crystal Data

Crystals of *trans*-1,3-cyclobutanedicarboxylic acid are monoclinic with $a = 5.452 \pm 0.005$, $b = 7.813 \pm 0.005$, $c = 8.131 \pm 0.008$ A; $\beta = 107.22 \pm 0.15°$. The space group based on systematically absent reflections is $P2_1/c$. Assuming two molecules/unit cell the calculated density is 1.45 g/cm^3.

Determination of the Structure

In space group $P2_1/c$ with two molecules per cell, each molecule must be centrosymmetric such that the center of symmetry of the molecule coincides with a center of symmetry of the unit cell. We applied the symbolic addition procedure,[14] using the film data, and obtained eight sets of possible signs. Eight electron-density maps were calculated, none of which gave a reasonable structure. However, the y and z coordinates of the large peaks in these maps were similar, and the resulting two-dimensional structure was refined by least squares[15] to a value of $R = \Sigma||F_o| - |F_c||/\Sigma|F_o|$ of 0.24. The x coordinates were easily obtained from the expected geometry of the molecule resulting in a structure which refined to $R = 0.15$.

We were later shown by an anonymous referee that the symbolic addition procedure could be applied with immediate success. Using reflections 150, $11\bar{1}$, and 413 to define the origin and assigning letters to reflections $36\bar{3}$, 013, and $21\bar{7}$, the signs of 57 reflections with E larger than 1.5 could be determined. The five largest peaks in an E map, calculated with the unknown letters all +, corresponded to the five heavy atoms.

The final refinement was accomplished by full-matrix, least-squares calculations using 362 nonzero independent structure factors measured with the diffractometer. The function minimized was $\Sigma(F_o - F_c)^2$. Atomic scattering factors for neutral atoms were taken from the "International Tables for X-ray Crystallography."[16] After five cycles of refinement of carbon and oxygen coordinates and anisotropic temperature factors in the form $\exp[-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + \beta_{12}hk + \beta_{23}kl + \beta_{13}hl)]$, R was 0.09. The three largest peaks in a difference map[15] calculated at this stage could be attributed to the ring hydrogen atoms. The next three peaks could not be hydrogen since they

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(1) T. N. Margulis, *Acta Cryst.*, **19**, 857 (1965).

(2) J. D. Dunitz, *ibid.*, **2**, 1 (1949).

(3) T. N. Margulis, *ibid.*, **18**, 742 (1965).

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(7) I. L. Karle, J. Karle, and K. Britts, *J. Am. Chem. Soc.*, **88**, 2918 (1966).

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(9) L. C. Snyder and S. Meiboom, *Chem. Eng. News*, **44**, 51 (April 25, 1966).

(10) W. G. Rothchild and B. P. Dailey, *J. Chem. Phys.*, **36**, 2931 (1962).

(11) H. P. Lemaire and R. L. Livingston, *J. Am. Chem. Soc.*, **74**, 5732 (1952).

(12) D. H. Deutsch and E. R. Buchman, *Experientia*, **6**, 462 (1950).

(13) W. Markownikoff and A. Krestownikoff, *Ann.*, **208**, 333 (1881).

(14) See, *e.g.*, I. L. Karle and J. Karle, *Acta Cryst.*, **19**, 92 (1965).

(15) A modified version of the program of Gantzel, Sparks, and Trueblood was used for all least-squares calculations. Fourier series were evaluated with a program by A. Zalkin.

(16) "International Tables for X-ray Crystallography," Vol. III, Kynoch Press, Birmingham, 1962, p 202.

Table I. Carbon and Oxygen Parameters[a]

Atom	x	y	z	β_{11}	β_{22}	β_{33}	β_{12}	β_{23}	β_{13}
C(1)	0.1901	−0.0489	0.0707	0.030	0.0149	0.0090	0.001	0.003	−0.001
C(2)	−0.0853	−0.1232	0.0212	0.039	0.0149	0.0106	−0.007	0.004	0.002
C(3)	0.3228	−0.0299	0.2589	0.032	0.0141	0.0107	0.003	0.003	0.000
O(1)	0.2282	−0.0693	0.3713	0.040	0.0277	0.0095	−0.022	0.005	0.001
O(2)	0.5529	0.0383	0.2915	0.036	0.0357	0.0103	−0.025	0.002	−0.000
				Estimated Standard Deviations × 10^4					
C(1)	7	5	4	20	8	6	20	10	10
C(2)	8	5	5	20	9	8	20	20	10
C(3)	7	5	5	20	8	7	20	20	10
O(1)	5	4	3	10	8	5	20	10	10
O(2)	6	5	3	10	9	5	20	10	10

[a] Anisotropic temperature factors are in the form $\exp[-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + \beta_{12}hk + \beta_{23}kl + \beta_{13}hl)]$.

occurred between bonded carbon atoms. The seventh strongest peak corresponded to the acid hydrogen. With the parameters of all the atoms including hydrogen adjusted by least squares, a final value for R of 0.050 was obtained. Initially, isotropic temperature factors were allowed to vary for each hydrogen, but this procedure resulted in the acid hydrogen oscillating perpendicular to the O–H bond and its B increasing without limit. In the final refinement this hydrogen atom was assigned a B value of 4.0 A^2, while the B's of the ring hydrogens were allowed to vary. The final structure parameters are given in Tables I and II. Observed and calculated structure factors are listed in Table III.

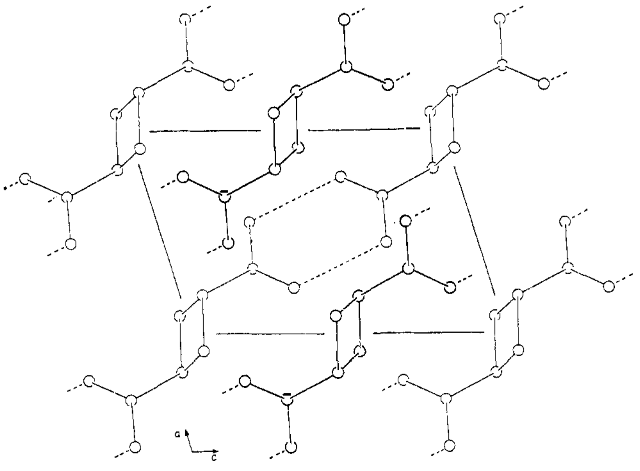

Figure 1. The crystal structure viewed along [010].

Discussion

Each molecule of *trans*-1,3-cyclobutanedicarboxylic acid is hydrogen-bonded to two other molecules, one at each end, to give infinite chains running parallel to [101]. The crystal structure is shown in Figure 1; one molecule is shown in Figure 2. The hydrogen-bonded atoms C(1), C(3), O(1), O(2), O(1′), O(2′), C(3′), and C(1′) are coplanar, no atom deviating by more than 0.009 A from a least-squares plane. The conformation of the molecule is such that atoms C(1), C(2), C(3), O(1), and O(2) are coplanar to within 0.01 A. The cyclobutane ring is planar.

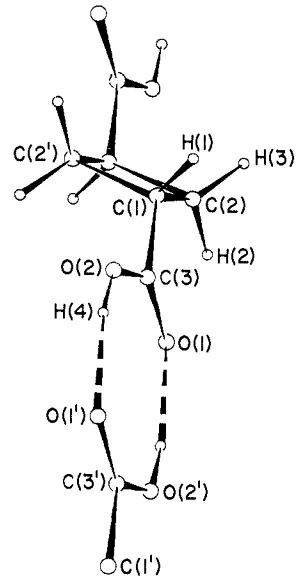

Figure 2. One molecule viewed perpendicular to [100].

Table II. Hydrogen Parameters[a]

Atom	x	y	z	B, A^2
H(1)	0.313	−0.123	0.021	1.8
H(2)	−0.153	−0.152	0.121	2.0
H(3)	−0.110	−0.229	−0.057	1.9
H(4)	0.623	0.061	0.392	4.0[b]

[a] Standard deviations are 0.007, 0.006, and 0.006 for x, y, and z coordinates, respectively. [b] Assigned.

Bond distances and angles are given in Table IV. The standard deviations estimated from the least-squares results are 0.006 A for C–C bonds, 0.005 A for C–O bonds, 0.04 A for C–H bonds, and 0.3° for angles not involving hydrogen. These standard deviations assume random errors in the intensity data and should be considered lower limits. Bond lengths, except those involving hydrogen,[17] were corrected for thermal motion assuming one atom to ride on another.[18] The dimensions of the carbonyl group after correction are in good agreement with values found in other satu-

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(17) The temperature factors for hydrogen have little physical meaning since the atomic scattering factors used are for the free atom; see L. H. Jensen and M. Sundaralingam, *Science*, **145**, 1185 (1964).

(18) W. R. Busing and H. A. Levy, *Acta Cryst.*, **17**, 142 (1964).

Table III. Observed (FOBS) and Calculated (FCAL) Structure Factors × 10^2

K	L	FOBS	FCAL	K	L	FOBS	FCAL	K	L	FOBS	FCAL	K	L	FOBS	FCAL	K	L	FOBS	FCAL
		H = 0		2	-6	847	804	3	-3	631	598	4	-1	139	163	2	-3	931	945
2	0	6294	6622	3	-6	584	580	4	-3	212	210	0	-2	692	678	3	-3	791	728
4	0	401	368	5	-6	170	202	5	-3	347	387	1	-2	537	562	4	-3	589	703
6	0	349	328	1	-7	1520	1525	6	-3	312	323	2	-2	253	199	5	-3	274	264
1	1	802	796	2	-7	988	945	0	-4	2653	2614	3	-2	400	389	6	-3	204	241
2	1	1672	1599	3	-7	873	875	1	-4	295	286	4	-2	288	307	0	-4	677	666
3	1	1202	1199	4	-7	580	591	2	-4	1546	1428	6	-2	273	289	2	-4	223	213
4	1	1007	912	0	-8	181	220	3	-4	202	154	7	-2	139	187	3	-4	353	362
5	1	273	227	0	0	602	630	4	-4	180	147	1	-3	1064	994	4	-4	314	342
6	1	352	352	1	0	1324	1315	5	-4	228	315	4	-3	822	835	5	-4	446	449
7	1	199	260	2	0	1956	1977	6	-4	328	353	5	-3	450	463	6	-4	196	224
8	1	182	218	3	0	3038	3021	1	-5	1258	1179	6	-3	958	923	1	-5	297	284
0	2	691	762	4	0	1330	1269	2	-5	570	517	7	-3	243	255	2	-5	337	378
2	2	2185	2222	5	0	2015	1940	4	-5	201	219	0	-4	358	411	3	-5	442	436
4	2	1692	1643	6	0	203	193	6	-5	619	638	2	-4	132	202	4	-5	476	493
6	2	356	323	7	0	773	764	0	-6	505	474	3	-4	452	474	5	-5	264	259
1	3	4177	4159	1	1	619	628	1	-6	141	122	4	-4	264	251	0	-6	972	1033
2	3	2159	2056	3	1	1342	1274	2	-6	455	428	5	-4	624	586	1	-6	247	276
3	3	610	571	4	1	637	569	4	-6	355	359	7	-4	226	198	2	-6	636	638
4	3	1290	1185	5	1	265	215	5	-6	271	311	1	-5	314	245	3	-6	348	353
5	3	462	450	6	1	494	512	1	-7	1249	1186	2	-5	923	936	1	-7	397	409
0	4	719	693	7	1	319	342	2	-7	973	967	4	-5	536	488	2	-7	251	282
1	4	379	365	8	1	143	165	3	-7	671	660	0	-6	464	440	4	-7	291	356
2	4	341	313	0	2	4866	4906	4	-7	707	713	3	-6	421	452	0	-8	348	403
3	4	606	577	1	2	604	593	0	-8	702	732	4	-6	289	343	2	-8	327	386
4	4	215	197	2	2	2073	2019	2	-8	350	345	5	-6	647	703	0	0	319	308
6	4	439	446	3	2	588	611	3	-8	214	240	1	-7	369	376	1	0	466	517
1	5	597	564	4	2	446	400	0	0	1657	1618	2	-7	254	258	2	0	293	265
2	5	192	188	5	2	382	392	1	0	1164	1161	3	-7	414	407	3	0	726	771
3	5	1008	997	6	2	189	172	2	0	1299	1171	4	-7	457	510	5	0	298	311
4	5	159	162	7	2	149	123	3	0	627	584	1	-8	351	369	6	0	203	238
5	5	602	557	1	3	1583	1518	4	0	343	229	2	-8	192	168	1	1	1066	1036
0	6	452	550	2	3	1160	1116	5	0	183	244	3	-8	495	596	2	1	278	259
1	6	824	805	3	3	1178	1143	6	0	244	278	0	0	1149	1113	3	1	345	313
2	6	724	748	4	3	1012	953	8	0	224	293	1	0	1061	1003	1	2	452	441
3	6	1336	1297	6	3	448	458	1	1	448	421	2	0	614	584	2	2	333	343
4	6	704	692	7	3	490	478	2	1	2467	2383	3	0	1018	966	3	2	556	602
5	6	438	433	0	4	1295	1216	3	1	964	909	6	0	351	344	4	2	294	339
4	7	401	424	2	4	1344	1269	4	1	1212	1127	7	0	150	165	1	3	1360	1391
0	8	347	390	4	4	924	908	5	1	358	352	1	1	1638	1592	2	3	205	223
		H = 1		5	4	265	245	6	1	159	133	2	1	229	200	3	3	721	734
1	-1	5075	5192	7	4	295	350	7	1	226	225	3	1	605	552	0	4	396	433
2	-1	2508	2505	1	5	1013	976	0	2	117	164	4	1	319	324	2	4	274	287
3	-1	820	791	2	5	202	232	1	2	313	342	6	1	328	358			H = 5	
4	-1	957	951	3	5	269	243	2	2	265	238	0	2	1413	1412	1	-1	483	517
5	-1	318	336	5	5	430	474	3	2	1288	1277	1	2	717	690	3	-1	197	181
0	-2	3403	3500	6	5	236	258	4	2	348	358	2	2	1064	1043	4	-1	259	317
1	-2	653	609	0	6	569	600	5	2	1127	1134	3	2	821	814	2	-2	246	265
2	-2	1831	1795	1	6	149	182	6	2	146	181	4	2	485	452	4	-2	401	425
3	-2	432	321	2	6	466	521	7	2	410	490	5	2	180	172	1	-3	582	573
4	-2	892	754	3	6	468	470	1	3	712	705	1	3	276	268	3	-3	575	560
5	-2	226	230	4	6	218	248	2	3	810	787	2	3	1406	1374	4	-3	156	130
6	-2	280	303	5	6	530	560	3	3	762	721	3	3	493	501	5	-3	215	213
1	-3	989	872	2	7	189	213	4	3	248	245	4	3	999	1011	0	-4	1154	1170
2	-3	580	578	4	7	244	279	5	3	155	116	5	3	260	269	1	-4	222	249
3	-3	554	536	0	8	371	465	7	3	278	345	6	3	261	346	2	-4	719	683
4	-3	808	824	1	8	232	276	0	4	682	648	0	4	1524	1528	3	-4	375	364
5	-3	694	682			H = 2		1	4	801	793	2	4	1028	1048	1	-5	614	634
6	-3	449	462	1	-1	1076	1074	3	4	1395	1364	3	4	194	187	3	-5	139	092
7	-3	261	263	2	-1	1193	1183	4	4	777	774	4	4	276	314	0	-6	248	264
1	-4	1343	1265	4	-1	1986	1981	5	4	863	865	5	4	256	276	2	-6	308	316
2	-4	528	547	5	-1	367	388	6	4	248	256	1	5	361	398	1	-7	401	480
3	-4	1881	1853	6	-1	1194	1233	1	5	153	176	2	5	407	408	3	0	178	194
4	-4	578	528	8	-1	289	316	3	5	272	257	0	6	196	219	4	0	247	272
5	-4	418	366	0	-2	1426	1439	0	6	654	720	1	6	254	271	1	1	304	338
6	-4	151	191	1	-2	1228	1180	1	6	208	212			H = 4		2	1	210	227
7	-4	197	208	2	-2	1118	1082	2	6	315	363	1	-1	147	115	0	2	226	258
1	-5	1473	1400	3	-2	744	729	3	6	301	308	2	-1	428	425	1	2	156	169
2	-5	429	406	4	-2	223	178	1	7	673	752	3	-1	630	615	2	2	251	261
3	-5	849	808	5	-2	644	631	2	7	140	151	5	-1	520	563	3	1	456	529
4	-5	165	163	6	-2	140	093			H = 3		0	-2	847	786			H = 6	
5	-5	436	444	7	-2	666	687	1	-1	2406	2300	2	-2	350	300	0	-2	264	275
0	-6	1543	1530	1	-3	694	592	2	-1	745	671	4	-2	180	188	0	-4	365	405
1	-6	310	333	2	-3	397	406	3	-1	965	865	1	-3	1293	1283				

rated carboxylic acids.[19] The O–H distance of 0.81 A has little meaning since the hydrogen atom was not well resolved in the difference map. Both the corrected (1.552 and 1.567 A) and uncorrected (1.548 and 1.562 A) C–C bond lengths in the cyclobutane ring are larger

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(19) M. A. Higgs and R. L. Sass, *Acta Cryst.*, **16**, 657 (1963).

Table IV. Bond Lengths and Angles

	Bond length, A			
Atoms	Obsd	Temp corrected	Atoms	Angle, deg
C(1)–C(2)	1.562	1.567	C(2)–C(1)–C(2′)	90.5
C(1)–C(2′)	1.548	1.552	C(2)–C(1)–C(3)	113.7
C(1)–C(3)	1.494	1.494	C(2′)–C(1)–C(3)	116.2
C(3)–O(1)	1.315	1.345	C(1)–C(3)–O(1)	124.1
C(3)–O(2)	1.214	1.238	C(1)–C(3)–O(2)	113.0
O(1)–O(2′)	2.658	2.662[a]	O(1)–C(3)–O(2)	122.9
C(1)–H(1)	1.05		H(1)–C(1)–C(2)	118
C(2)–H(2)	1.01		H(1)–C(1)–C(2′)	112
C(2)–H(3)	1.03		H(1)–C(1)–C(3)	106
O(2)–H(4)	0.81		H(2)–C(2)–C(1)	114
			H(2)–C(2)–C(1′)	116
			H(2)–C(2)–H(3)	108
			H(3)–C(2)–C(1)	116
			H(3)–C(2)–C(1′)	114
			C(3)–O(2)–H(4)	115

[a] With the correction applied assuming the two atoms to move independently, this distance becomes 2.717 A.

than 1.537 A, the average value of a C–C single bond given by Sutton.[20]

The present work represents the third structure determination of a cyclobutane derivative in which the ring is planar, not part of a condensed polycyclic system, and not involved with endo- or exocyclic unsaturation. The other two, tetracyanocyclobutane[4] and tetraphenylcyclobutane,[1,2] crystallize in the same space group ($P2_1/c$) with the ring lying on a center of symmetry. The C–C bond lengths in the rings are comparable to ours: 1.566 ± 0.015 and 1.573 ± 0.015 A for tetraphenylcyclobutane; 1.547 ± 0.002 and 1.561 ± 0.002 A for tetracyanocyclobutane. Our work thus confirms the existence of longer-than-normal C–C single bonds in simple, planar cyclobutane derivatives.

Acknowledgment. This work was supported by the National Science Foundation under Grant GP-4550. We are indebted to Professor K. N. Trueblood for bringing this problem to our attention.

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(20) L. E. Sutton, "Tables of Interatomic Distances and Configuration in Molecules and Ions, Supplement 1956–1959," The Chemical Society, London, 1965.

γ Radiolysis of Cystine in Aqueous Solution. Dose-Rate Effects and a Proposed Mechanism[1]

John W. Purdie

Contribution from the Defence Chemical, Biological and Radiation Laboratories, Defence Research Board, Ottawa, Canada. *Received June 1, 1966*

Abstract: Solutions of L-cystine (CySSCy) in water (3×10^{-4} *M*) were exposed to 10,000 rads of Co^{60} γ rays. *G* values were determined for the following products: $CySO_2H$, $CySO_3H$, $CySO_2SH$, $CySSO_3H$, CySH, and CySSSCy. The effect of OH and e_{aq}^- scavengers on the yields was also investigated. The yields of $CySO_2H$, $CySO_3H$, and CySH were dose-rate dependent in the range 1 to 800 rads/min. A mechanism for the radiolysis is presented and discussed: CySOH appears to be the main precursor of both $CySO_2H$ and $CySO_3H$ with O_2^- participating in formation of the latter. CySSSCy, the yield of which was independent of dose rate, is probably produced from cystine by reaction with CyS radicals.

The disulfide bond of cystine, which is essential to the tertiary structure of many enzymes and proteins, is particularly sensitive to ionizing radiation. Eldjarn and Pihl[2] have postulated that the cysteine and cystine residues of proteins form mixed disulfides with radioprotective compounds such as cysteamine. During radiolysis the protective residue reacts readily with free radicals, thereby protecting the protein.

Radiolysis of cysteine, which is also a radioprotective compound, has been subjected to detailed study recently.[3] Aqueous solutions of cystine have been investigated by several workers,[4] but many of the early studies involved high doses. The following products were identified by Markakis and Tappel:[4d] cysteine, H_2S, sulfur, sulfate, ammonia, and alanine. A detailed analysis of the initial products was first achieved by Grant, *et al.*,[5] using paper chromatography and electrophoresis. Similar studies were conducted by Forbes and co-workers[6] who examined both ultraviolet photolysis and radiolysis of cystine solutions. Brdička, *et al.*,[7] have also studied the radiolysis of cystine solutions using similar methods. Approximate *G* values were estimated by Grant, *et al.*,[5] by comparing the spots of products on paper chromatograms with those from standard solutions. The present study was undertaken to determine the yields more accurately and elucidate the

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(1) Issued as DCBRL Report No. 508.

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