natural quartz crystals is similar to that predicted by the dynamical theory for perfect, absorbing crystals.

Using the results of the theoretical investigation, it has been possible to obtain experimental F values with an accuracy of $\pm 10\%$ from one crystal, without the necessity for making detailed assumptions about the degree of perfection of its texture. From the combination of observations from a series of crystals it may be possible to increase the accuracy of this method. Higher accuracy may be obtained if the absorbing effect of any projections on the crystal surface can be found from the experimental variation of $J(\beta)$ with β for a weak reflexion.

A disadvantage of the method is that the crystals used must be quite large, and show faces suitable for mounting. However, wherever applicable the method leads to far more accurate results than any previous investigation.

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The Structure of Urea. Interatomic Distances and Resonance in Urea and Related Compounds*

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The interatomic distances in urea have been redetermined by Fourier and least-squares analysis of complete data obtained with Cu $K\alpha$ radiation. The results are C=O = 1·26 Å, C–N = 1·34 Å, ∠ N–C–O = 121°, N–H · · · O = 2·99 Å and N–H · · · O′ = 3·04 Å. Evidence for the position of the hydrogen atoms is found in the electron-density sections. Improved agreement of calculated with observed structure factors was obtained by including the hydrogen scattering in the calculations, and by use of anisotropic temperature factors.

Revised values for carbon–oxygen and carbon–nitrogen double bonds are suggested, and these values are used to discuss the effect of resonance on interatomic distances in urea and related compounds.

Introduction

Because of its simplicity, the structure of urea was one of the first organic structures to be determined by X-ray crystallographic methods (Mark & Weissenberg, 1923; Hendricks, 1928; Wyckoff, 1930, 1932; Wyckoff & Corey, 1934). These early studies agreed on the space group, unit cell, and approximate parameters. The space group is D_{2d}^3–$P\bar{4}2_1m$; the unit cell contains two molecules of $CO(NH_2)_2$, with atoms in the following positions: 2 C in (*c*): $(0, \frac{1}{2}, z)$, $(\frac{1}{2}, 0, \bar{z})$; 2 O in a second set of positions (*c*); 4 N in (*e*):

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$(x, \frac{1}{2}+x, z)$, $(\bar{x}, \frac{1}{2}-x, z)$, $(\frac{1}{2}+x, \bar{x}, \bar{z})$, $(\frac{1}{2}-x, x, \bar{z})$; and 8 H in two additional sets of positions (*e*). This structure is illustrated in Fig. 1. The packing of the

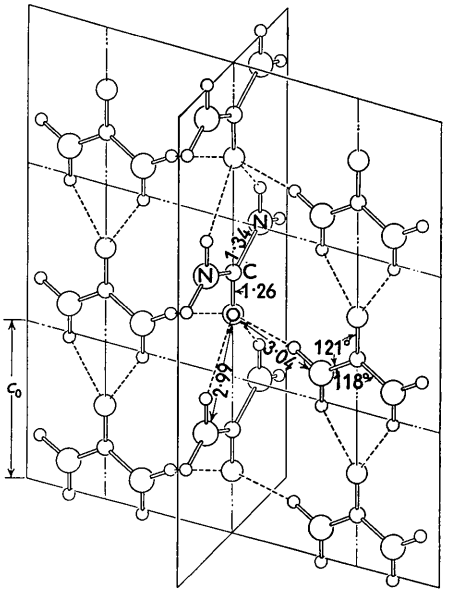


Fig. 1. View of the urea structure, showing final bond distances and angles. Hydrogen bonds are indicated by the broken lines.

urea molecules is determined largely by hydrogen bonds, N–H · · · O.

A part of the attack in this laboratory on the problem of the structure of proteins has included the precise determination of the molecular dimensions and hydrogen bonding in various amino acids and related compounds. This made it seem worthwhile to determine, with the advanced techniques now available, the interatomic distances in crystalline urea in order to obtain additional precise information regarding the dimension of molecules involving resonance associated with the –NH–CO– group. A comparison of our final results with those of the earlier studies will be given later.

Experimental

Essentially all of the reflections accessible to Cu $K\alpha$ radiation were collected by Weissenberg photography on two crystals. The first crystal, with c_0 the rotation axis, was reduced to a cylinder 0·22 mm. in diameter, and zero-, first-, second- and third-layer-line data were recorded. The second crystal, with the zone axis [110] vertical, was a cylinder 0·35 mm. in diameter; the data of the layer lines zero to five were recorded. The small size of the crystals minimized absorption effects. Intensities were estimated by means of calibrated intensity strips and the multiple-film technique. The intensities of the strong reflections were estimated from powder photographs in order to eliminate extinction effects. Values for the lattice constants were obtained by the method of least squares from measurements of 29 powder lines on photographs taken with Cu $K\alpha$ radiation (λ=1·542 Å) in a powder camera of 5·73 cm. nominal radius. It is of interest to compare our values with those obtained in previous investigations; this comparison is made in Table 1.

Table 1. *Lattice constants of urea*

	a_0 (Å)	c_0 (Å)	c/a
Mez (1902)	—	—	0·8333
Mark & Weissenberg (1923)	5·62	4·69	0·835
Hendricks (1928)	(5·71)	4·76	(0·8333)
Wyckoff (1930)	5·659	4·717	0·8335
This work	5·661	4·712	0·8325

Refinement of parameters

The atomic parameters were refined by both Fourier and least-squares methods. Since the urea molecules lie in planes parallel to (110) and (1$\bar{1}$0), only the section $\varrho(x, \frac{1}{2}+x, z)$ of the three-dimensional electron-density function was calculated. During the parameter refinement by the Fourier method twelve such sections were computed. Since the space group $P\bar{4}2_1m$ lacks a center of symmetry it is necessary to assign a general phase angle, based on a calculated structure factor, to each observed value $|F_{hkl}|$. The terms in the Fourier series for the electron-density distribution thus depend not only on the observed structure-factor amplitudes, but on the values of the atomic parameters used, the atom form factors, and the contributions of the hydrogen atoms as well. The Fourier refinement included variation of all of these factors.

In the least-squares treatment of the data (Hughes, 1941) the observational equations used were of the form

$$\sqrt{w_{hkl}}\cdot\left\{ln(|F_o|/|F_c|) = -\Delta K-\Delta B_1(h^2+k^2)-\Delta B_2 l^2+ \right.$$
$$\left. +\frac{1}{|F_c|}\left(\frac{\partial F_c}{\partial z_C}\Delta z_C+\frac{\partial F_c}{\partial z_C}\Delta z_O+\frac{\partial F_c}{\partial z_N}\Delta z_N+\frac{\partial F_c}{\partial x_N}\Delta x_N\right)\right\},$$

where e^K is the scale factor and B_1 and B_2 are temperature factors. The approximate anisotropic temperature factors were determined from the peak shapes in the electron-density functions. Logarithms were used because $\ln|F_c|$ is linear in the temperature and scale factors, and because the logarithmic derivatives with respect to the atomic coordinates do not contain the temperature factor. Atom form factors of James & Brindley were used in the structure factor calculations (*Internationale Tabellen*).

An IBM 604 calculating punch was used to calculate the coefficients in the observational equations, for the reduction to normal equations, and for structure-factor calculations. The solution of the seven normal equations was carried out in the following steps: (1) the four equations for the atomic positions were solved by neglecting the off-diagonal terms; (2) three iterations were made to complete the solution of these four

equations; (3) these solutions were put into the three scale and temperature-factor equations, which were then solved exactly for K, B_1, and B_2; (4) these results were put into the equations for the atomic positions to obtain a final solution for these parameters. Neglect of the off-diagonal terms in the solution of the equations for the atomic positions leads to an average change of 0·003 Å and a maximum change of 0·004 Å. Neglect of the scale and temperature-factor terms in solving these equations produced an even smaller effect, the average change being 0·0003 Å and the maximum 0·0006 Å. We may thus conclude that the usual practice of solving for scale and temperature factors separately produces no significant error, and that neglect of the off-diagonal terms in the equations of atomic position leads to only a small decrease in accuracy, as has been previously pointed out (Shoemaker, Donohue, Schomaker & Corey, 1950).

In the final electron-density calculation the phases were determined from structure factors for which an anisotropic temperature factor $\exp[-\sin^2\theta/\lambda^2(3{\cdot}9\sin^2\varphi_h+1{\cdot}9\cos^2\varphi_h)]$ was applied to the form factors of carbon and oxygen, and the factor $\exp[-\sin^2\theta/\lambda^2(3{\cdot}9\sin^2\varphi_h+1{\cdot}9\cos^2\varphi_h+5{\cdot}7\sin^2\varphi_h\sin^2\theta_h)]$ was applied to the form factor of nitrogen, where φ_h and $(\theta_h+45°)$ are the polar coordinates of the reciprocal-lattice vector, $h=2\sin\theta/\lambda$. The numerical values in these factors were obtained from the least-squares treatment described above, and from measurement of the peak shapes in the electron-density distributions. In addition, the contributions of the hydrogen atoms, the positions of which were suggested in the first electron-density sections, were included in the phase-angle determinations. The four hydrogen atoms were placed so that N–H = 1·00 Å and ∠ C–N–H = 120°. The final electron-density section, $\varrho(x, \frac{1}{2}+x, z)$, is shown in Fig. 2; an electron density section, $\varrho(x, y, z_N)$, is shown in Fig. 3. Fourier sections $\varrho(x, \frac{1}{2}+x, z)$ were also calculated using calculated phases and amplitudes, in order to correct for series-termination errors (Booth, 1946). The anisotropic atom form factors were used in two such calculations, one of which included the scattering of the hydrogen atoms, the other did not. Atomic parameters at various stages of the work are presented in Table 2, together with the results of the previous work on urea.

The average between the second least-squares results and the final Fourier results were taken as the best values for the atomic parameters. The two methods gave answers which are in satisfactory agreement; the average difference is 0·005 Å, the largest difference being that of 0·016 Å in the case of x_N. In three other crystals for which both three-dimensional Fourier and least-squares were used, threonine (Shoemaker *et al.*, 1950), hydroxyproline (Donohue & Trueblood, 1952) and serine (Shoemaker, Barieau, Donohue & Lu, 1952), the average differences

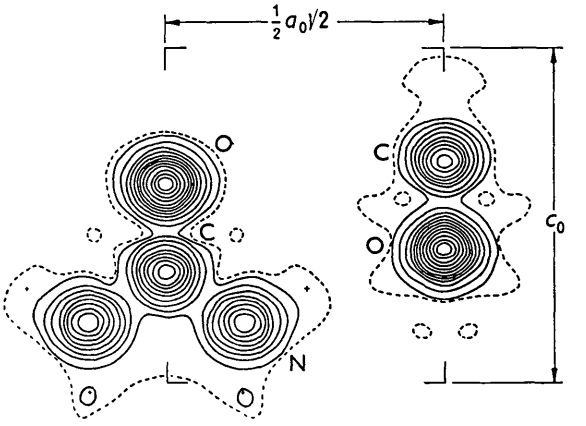


Fig. 2. Electron-density section $\varrho(x, \frac{1}{2}+x, z)$. Solid contour lines are drawn at 1, 2, 3, ... e.Å^{-3}; the broken contour line is at $\frac{1}{2}$ e.Å^{-3}. The small crosses indicate the positions assigned to the hydrogen atoms.

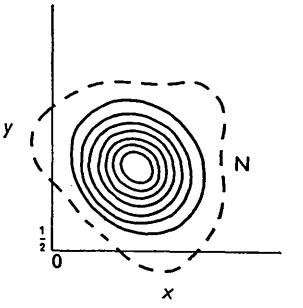


Fig. 3. Electron-density section $\varrho(x, y, z_N)$, showing the anisotropic shape of the nitrogen peak due to torsional motion of the molecule.

Table 2. *Atomic parameters in urea*

	z_C	z_O	x_N	z_N
1. Mark & Weissenberg	—	—	0·13	—
2. Hendricks	0·32	0·57	0·13	0·20
3. Wyckoff	0·32	0·58	0·14	0·17
4. Wyckoff & Corey	0·335	0·60	0·145	0·18
5. From 1st least squares	0·3319	0·5983	0·1450	0·1844
6. From 2nd least squares	0·3305	0·5984	0·1443	0·1848
7. $\varrho(x, \frac{1}{2}+x, z)$; phases without hydrogen contributions	0·3322	0·5985	0·1421	0·1852
8. $\varrho(x, \frac{1}{2}+x, z)$; phases with hydrogen contributions	0·3310	0·5990	0·1415	0·1849
9. Final values; average of 6 and 8	0·3308	0·5987	0·1429	0·1848

Table 3. *Observed and calculated structure factors*

hkl	$\|F_o\|$	$\|F_c\|$	α (°)	*hkl*	$\|F_o\|$	$\|F_c\|$	α (°)	*hkl*	$\|F_o\|$	$\|F_c\|$	α (°)
200	10·88	12·38	0	521	1·70	1·01	14·1	113	3·19	2·58	221·2
400	2·20	0·69	0	621	1·40	1·75	147·1	213	5·82	5·94	137·9
600	3·42	3·62	0	331	2·44	2·53	24·6	313	5·59	5·61	184·1
110	30·17	28·17	180	431	2·88	2·11	264·0	413	1·93	1·82	57·8
210	11·80	11·51	0	531	1·70	1·75	335·4	513	1·75	2·02	166·0
310	4·44	4·30	180	631	< 0·43	0·35	117·2	613	1·16	1·31	57·7
410	1·15	1·68	180	441	1·50	1·29	192·3	223	4·62	3·99	28·1
510	3·85	3·44	180	541	0·64	0·43	116·2	323	3·10	3·41	296·9
610	0·88	1·11	180	551	1·61	1·09	32·4	423	1·45	1·63	345·6
710	1·97	2·09	180	002	9·70	9·73	180	523	2·03	1·97	222·2
220	10·34	11·69	0	102	9·89	9·80	90	333	0·64	0·40	240·0
320	3·13	3·17	180	202	2·27	2·34	0	433	2·24	2·14	80·9
420	6·14	5·94	0	302	2·89	2·37	270	533	0·80	1·02	168·5
520	2·18	2·33	0	402	1·80	2·57	0	443	< 0·55	0·33	14·1
620	2·50	1·99	0	502	0·68	0·08	90	004	4·17	4·59	180
330	8·85	9·71	180	602	< 0·69	0·82	180	104	1·30	0·64	90
430	1·62	0·58	180	112	5·76	5·19	61·1	204	3·62	3·62	180
530	3·57	3·17	180	212	4·73	4·70	206·4	304	6·34	6·64	90
630	< 0·48	0·37	180	312	3·11	2·93	142·0	404	1·82	1·91	180
440	4·65	4·61	0	412	1·03	0·99	33·1	504	2·20	2·13	90
540	< 0·59	0·47	180	512	1·09	1·34	266·5	114	4·98	5·11	320·6
550	0·80	1·53	180	612	0·69	0·88	313·9	214	4·69	4·34	268·5
001	4·02	4·33	180	222	5·30	4·85	262·8	314	2·95	2·89	334·6
101	10·66	10·19	90	322	1·92	2·56	58·5	414	3·24	3·72	270·6
201	10·88	10·87	180	422	1·21	1·09	119·7	514	1·84	1·88	42·3
301	7·10	7·65	270	522	1·26	1·34	158·9	224	4·44	4·72	126·7
401	6·24	6·31	180	622	0·64	0·65	96·2	324	2·63	2·06	89·1
501	< 0·60	0·66	270	332	1·64	1·98	12·8	424	2·04	2·01	205·2
601	1·29	1·17	180	432	2·16	1·96	278·5	524	1·05	1·36	92·1
701	0·97	1·10	90	532	0·91	0·53	308·5	334	2·16	2·01	348·6
111	11·74	14·25	53·2	632	< 0·43	0·28	44·4	005	5·14	5·17	0
211	5·83	5·18	25·2	442	1·03	1·07	191·2	105	1·46	2·76	270
311	8·77	8·64	16·7	542	< 0·48	0·43	52·8	205	0·91	0·34	0
411	2·01	2·72	104·9	003	2·45	3·21	180	305	< 0·46	0·07	270
511	2·90	3·40	324·6	103	7·45	7·58	270	115	2·95	2·83	202·2
611	1·23	0·86	239·2	203	5·25	5·40	0	215	2·34	2·60	28·7
711	0·75	0·72	358·4	303	2·16	1·72	270	315	0·68	0·65	317·6
221	10·88	10·56	227·5	403	4·92	5·60	0	225	1·46	1·59	52·8
321	1·38	1·49	152·4	503	1·30	1·31	270	325	1·68	1·56	236·1
421	3·20	3·37	155·7	603	0·79	0·34	0	006	2·27	2·43	0

were 0·005, 0·008 and 0·006 Å, and the maximum differences were 0·016, 0·023 and 0·013 Å, respectively.

Application of the theory of errors to the least-squares results gives standard errors of 0·015, 0·009, 0·012 and 0·008 Å in z_C, z_O, x_N and z_N, respectively; the average is 0·011 Å. If we assume that the 0·004 Å r.m.s. deviation from the average of parameters determined by least-squares and Fourier methods represents an approximate standard error due to choice of the method of interpretation (Shoemaker *et al.*, 1950), the total average standard error in a single parameter is 0·012 Å, which corresponds to a probable error of 0·008 Å. Probable errors in the molecular parameters are thus 0·011 Å for C–O, 0·009 Å for C–N, 0·45° for ∠ O–C–N and 0·9° for ∠ N–C–N.

Table 3 gives the observed and calculated values of $|F_{hkl}|$. The calculated values include the contributions of the hydrogen atoms. The average discrepancy, R, is 9·9%. If the hydrogen scattering is neglected R is 11·0%. If the torsional motion of the nitrogen atom is also neglected R is 12·4%.

Discussion

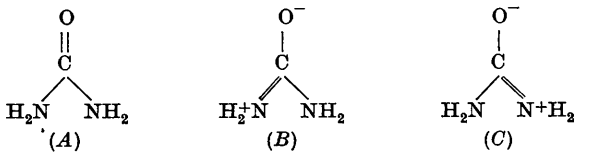
Interatomic distances and interbond angles in the urea crystal are listed in Table 4.

Table 4. *Bond distances and angles in urea*

C–O	1·262 Å
C–N	1·335 Å
∠ N–C–N	118·0°
∠ N–C–O	121·0°
N · · · O	2·989 Å
∠ C–N · · · O	98·5°
N · · · O′	3·035 Å
∠ C–N · · · O′	129·3°

The hydrogen bond distances, N · · · O = 2·99 and 3·04 Å, correspond to rather weak bonds, since both are considerably longer than the N · · · O distances of 2·7 to 2·9 Å found in the amino acids. This relative lengthening is without doubt due to (1) the acceptance of a large number (four) of hydrogen bonds by the oxygen atom in urea, and (2) the smaller formal charge on the nitrogen atom in urea; in the amino acids this atom, because of the *Zwitterion* structure, has a charge of +1, while in urea it has a smaller

positive charge, depending on the relative contributions of the following resonance structures:



In order to estimate the relative contributions of these structures we shall use the familiar equation of Pauling (1940) which relates bond length with double-bond character. It is then necessary to have reliable values for the lengths of carbon–oxygen and carbon–nitrogen single and double bonds. Satisfactory agreement with observed results is obtained if the revised values 1·20 Å for C=O and 1·24 Å for C=N are used, combined with the values 1·42 Å for C–O and 1·48 Å for C–N adopted by Schomaker & Stevenson (1941). The values for C=O and C=N are 0·015 and 0·025 Å shorter than those given by Pauling (1940). In carboxylic acids there is resonance between two forms,  and  , the relative contributions of the two forms depending on the situations of the two oxygen atoms, i.e. the number and strengths of the hydrogen bonds they are accepting, or whether one of them is covalently bonded to a hydrogen atom, as in the case of the free acids. The total double-bond character to be distributed between the two carbon oxygen bonds is 1·00, so that if a percentage of double bond character is assigned to one bond the lengths of both bonds can be predicted. Comparison of observed with calculated bond lengths is made in Table 5.

The data in Table 5 show that excellent agreement between calculated and observed bond distances is obtained by the use of Pauling's equation and the distances C–O = 1·42 Å and C=O = 1·20 Å.

Calculated and observed values for carbon–oxygen and carbon–nitrogen bonds in peptides and related compounds are presented in Table 6. The agreement is again satisfactory. The results indicate that in urea structures *A*, *B* and *C* contribute 40%, 30% and 30%, respectively. We therefore expect the entire urea molecule, including the hydrogen atoms, to be planar, a conclusion verified by the electron-density sections, the disposition of the hydrogen bonds, and the results of spectroscopic investigations of urea crystals with polarized infra-red (Keller, 1948; Waldron & Badger, 1950). Moreover, unsymmetrical structures such as

Table 5. *Bond lengths in carboxylic acids*

Compound	Double-bond character	C–O, obs. (Å)	C–O, calc. (Å)	Reference
Adipic acid	0·30	1·29	1·30	Morrison & Robertson (1949*b*)
	0·70	1·23	1·23	
Glutaric acid	0·30	1·30	1·30	Morrison & Robertson (1949*d*)
	0·70	1·23	1·23	
Sebacic acid	0·40	1·27	1·27	Morrison & Robertson (1949*c*)
	0·60	1·24	1·24	
Succinic acid	0·30	1·30	1·30	Morrison & Robertson (1949*a*)
	0·70	1·25	1·23	
Acetylglycine	0·25	1·31	1·31	Carpenter & Donohue (1950)
	0·75	1·19	1·22	
Alanine	0·35	1·27	1·28	Donohue (1949)
	0·65	1·21	1·23	
Glycylglycine	0·35	1·27	1·28	Hughes & Moore (1949)
	0·65	1·21	1·23	
Hydroxyproline	0·40	1·27	1·27	Donohue & Trueblood (1952)
	0·60	1·25	1·24	
Serine	0·50	1·27	1·26	Shoemaker, Barieau, Donohue & Lu (1952)
	0·50	1·26	1·26	
Threonine	0·50	1·25	1·26	Shoemaker, Donohue, Schomaker & Corey (1950)
	0·50	1·24	1·26	
Formic Acid monomer	0·10	1·37	1·37	Schomaker & O'Gorman (1947)
	0·90	1·21	1·21	
Carbonate ion	0·33	1·31	1·29	Elliott (1937)

Table 6. *Bond distances in peptides and related compounds*

Compound	Bond	Double-bond character	Distance, obs. (Å)	Distance, calc. (Å)	Reference
Acetamide	C–O	0·70	1·28	1·23	Senti & Harker (1940)
	C–N	0·30	1·38	1·35	
Acetylglycine	C–O	0·60	1·24	1·24	Carpenter & Donohue (1950)
	C–N	0·40	1·32	1·32	
Diketopiperazine	C–O	0·60	1·25	1·24	Corey (1938)
	C–N	0·40	1·33	1·32	
Glycylglycine	C–O	0·60	1·23	1·24	Hughes & Moore (1949)
	C–N	0·40	1·29	1·32	
Polypeptide chain	C–O	0·65	1·23	1·23	Corey & Donohue (1950)
	C–N	0·35	1·32	1·33	
Urea	C–O	0·40	1·26	1·27	This work
	C–N	0·30	1·34	1·35	
Urea.H_2O_2	C–O	0·50	1·24	1·25	Lu, Hughes & Giguere (1941)
	C–N	0·25	1·34	1·36	

$-O-C\begin{matrix}NH\\ \diagdown\\ \diagup\\ N^+H_3\end{matrix}$, which Clow (1937) proposed on the basis of magnetic susceptibility measurements, are not in agreement with the above results.

Also of interest is the result that the interatomic distances in the polypeptide chain structure formulated by Corey & Donohue (1950) correspond to contributions of 65% and 35% for the structures

$$\begin{matrix}O & H\\ \| & |\\ -C-N-\end{matrix} \quad \text{and} \quad \begin{matrix}O^- & H\\ | & |\\ -C=N^+-\end{matrix}$$

The contribution of the second of these structures leads to the planarity of the peptide group and its associated atoms, a situation which has already been emphasized and incorporated into detailed considerations of the configuration of polypeptide chains in protein molecules (Pauling & Corey, 1950; Pauling, Corey & Branson, 1951).

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