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Chemistry

MASTER

DETERMINATION OF THE CRYSTAL SYSTEM AND
LATTICE PARAMETERS OF PREVIOUSLY
UNREPORTED COMPOUNDS

C. E. Holcombe

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE Y-12 PLANT

operated for the **ATOMIC ENERGY COMMISSION** *under* **U. S. GOVERNMENT Contract W-7405 eng 26**

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ABSTRACT

Computer programs have been developed in BASIC language, for the CEIR 420 teletype computer system, in order to index patterns of cubic, tetragonal, and hexagonal materials whose crystal systems were previously unidentified by analytical methods.

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SUMMARY

Computer programs have been developed in BASIC language, for the CEIR 420 teletype computer system, in order to index patterns of cubic, tetragonal, and hexagonal materials whose crystal systems were previously unidentified by analytical methods. In all the crystal systems with which these programs apply, only the average lattice parameters (not the least squares average) were found. Thus, the lattice parameters calculated by these programs should be refined by an appropriate technique.

INTRODUCTION

A knowledge of crystal structure is a necessary prerequisite to any understanding of such phenomena as plastic deformation, alloy formation, phase transformations, and phase purity. The methods of determining structure from powder patterns are often of interest when new substances are synthesized in materials research, particularly where single crystals are difficult or almost impossible to obtain.

The crystal structure of a substance determines the diffraction pattern of that substance; ie, the shape and size of the unit cell determines the angular positions of the diffraction lines, and the arrangements of the atoms within the unit cell determines the relative intensities of the lines.

The determination of an unknown structure proceeds in three major steps:

1. The shape and size of the unit cell are deduced from the angular positions of the diffraction lines. An assumption is first made as to which of the seven crystal systems the unknown structure belongs, and then, on the basis of this assumption, the correct Miller indices (hkl values) are assigned to each reflection. This step is called "indexing the pattern" and is only possible when the correct choice of a crystal system has been made. In many cases, when there is no evidence for assuming a certain structure, it is necessary to begin with cubic, then tetragonal, then hexagonal, and finally, more complicated structures. Once a pattern has been indexed, the shape of the unit cell is known (from the crystal system), and its size is calculable from the positions and Miller indices of the diffraction lines. Analytical or graphical methods are normally used to index the powder patterns.
2. The number of atoms per unit cell is then computed from the shape and size of the unit cell, the chemical composition of the specimen, and its measured density.
3. The positions of the atoms within the unit cell are deduced from the relative intensities of the diffraction lines. This approach is a difficult process and is simplified by the use of single crystals, although it can be deduced from the powder diffraction patterns.

Since the problem of structure determination is one of finding a structure which will account for all the lines on the pattern (in both position and intensity), it is necessary to make sure at the outset that the observed pattern does not contain any extraneous lines. There are two sources of extraneous lines: (1) diffraction of X rays having wavelengths different from that of the principal component of the radiation, and (2) diffraction by substances other than the unknown. These extraneous lines should be accounted for and eliminated before attempting to index the pattern of the unknown substance. Furthermore, systematic errors in measurements of d -spacings or $\sin^2\theta$ values inevitably result from such things as film shrinkage, incorrect camera radius,

off centering of the specimen, and absorption in the specimen when Debye-Scherrer cameras are used. When diffractometers are used, systematic errors result from misalignment of the instrument, use of a flat specimen instead of a curved specimen to conform to the focusing circle, absorption in the specimen, displacement of the specimen from the diffractometer axis (this is usually the largest source of error), and vertical divergence of the incident beam. These errors are not large enough to cause any difficulty in indexing patterns of cubic crystals, but they can seriously interfere with the determination of some noncubic structures. The best method of removing such errors from the data is to calibrate the camera or diffractometer with a substance of known lattice parameter, mixed with the unknown. The difference between the observed and calculated values of $\sin^2\theta$ for the standard substance gives the error in $\sin^2\theta$, and this error can be plotted as a function of the observed values of $\sin^2\theta$. The errors represented by the ordinates of such a curve can then be applied (added or subtracted) to each of the observed values of $\sin^2\theta$ for the diffraction lines of the unknown substance.

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WORK ON THE PROBLEM

ACCOMPLISHMENTS

Since determination of the shape and size of the unit cell of an unknown substance (indexing the powder pattern) can be accomplished by analytical means, a computerized solution can readily be employed. Therefore, computer programs were written in BASIC language for the CEIR 420 teletype computer system in order to index patterns of cubic, tetragonal, and hexagonal materials with crystal systems that could not be previously identified by analytical methods.

These programs were verified with copper (example of a face-centered cubic), yttrium sesquioxide (body-centered cubic), and zinc (hexagonal), and gave excellent agreement with reported values.

DISCUSSION

These computer programs (in combination) can be used to determine the best index of the diffraction pattern of an unknown substance, if it belongs to the cubic, tetragonal, or hexagonal crystal system. It is often necessary to use all three crystal systems as assumed structures, calculate the best values for each system, and choose the best index as the correct one. These indexing programs follow essentially the same procedure as that described by Cullity.^(a)

The cubic indexing program (CUINDEX) solves the following equation for a set of integers, S , which will yield a constant quotient when divided one by one into the observed $\sin^2\theta$ values:

$$\frac{\sin^2\theta}{S} = \frac{\lambda^2}{4a^2},$$

where:

$\sin^2\theta$ is calculated from the observed X-ray diffraction lines,
 $S = (H^2 + K^2 + L^2)$,
 λ is the wavelength of the radiation, and
 a is the lattice parameter for the cubic cell.

The program lists the average value, a , and distinguishes among simple, body-centered, and face-centered cubic lattice types.

(a) Cullity, B. D.; Elements of X-Ray Diffraction; Addison-Wesley Publishing Company, Inc, Reading, Massachusetts (1959).

Tetragonal and hexagonal indexing programs solve the following equation for A and C:

$$\sin^2\theta = A(S) + CL^2,$$

where:

$$\begin{aligned} A &= \lambda^2/4a^2, \\ C &= \lambda^2/4c^2, \text{ and} \\ S &= (H^2 + K^2) \text{ for the tetragonal system.} \end{aligned}$$

For the hexagonal system:

$$\begin{aligned} A &= \lambda^2/3a^2, \\ C &= \lambda^2/4c^2, \\ S &= (H^2 + HK + K^2), \text{ and} \end{aligned}$$

a and c are the lattice parameters for the tetragonal and hexagonal systems.

The value of A is obtained from HK0 lines, using either the TENDX1 (tetragonal) or the HXNDX1 (hexagonal) program. The value of C is obtained from either the TENDX2 (tetragonal) or the HXNDX2 (hexagonal) program.

The procedure for using the CEIR 420 teletype computer system for determining the crystal system and lattice parameters of previously unreported compounds is as described in the sections that follow:

Indexing Patterns of Cubic Crystals

1. Call CUINDX by typing OLD and responding CUINDX to the computer question PROBLEM NAME. Wait for the computer to print READY.
2. Type KEY. Wait for READY.
3. Type 440 DATA D where D is the number of d-spacings that are being used to index the unknown material.

Usually, the first 15 lines in the pattern are enough. The statement "442 DATA E, I1, N, L1" contains the number of possible $H^2 + K^2 + L^2$ values, E, for a simple cubic; the number of $H^2 + K^2 + L^2$ values for a face-centered cubic, I1; the number of $H^2 + K^2 + L^2$ values for a body-centered cubic, N; and the wavelength, L1, which is normally 1.54178 for CuK α radiation. The statement "450, 455, and 456 DATA" contains the values of $H^2 + K^2 + L^2$ for the simple cubic; "460 DATA" contains the values of $H^2 + K^2 + L^2$ for the face-centered cubic; and "465 and 466 DATA" contains the values of $H^2 + K^2 + L^2$ for the body-centered cubic. Therefore, leave 442, 450, 455, 456, 460, 465, and 466 DATA unchanged unless a larger number of $H^2 + K^2 + L^2$ values is needed, in which case the DIM statements (5 and 10) must be increased accordingly.

4. Type 445 DATA $d_1, d_2, d_3, \dots, d_n$, where d stands for the d -spacings observed. 445-449 DATA is reserved for d -values.
5. Type SAVE. Wait for READY. Then type LISTNH:445. This information provides a list of the data and makes it possible to see if any data are left in 445-449 that are not the appropriate data. If such extra data are found (ie, line 446 DATA), type KEY; when the computer responds READY, type 446. This step erases the data there.
6. Type RUN.
7. The program is written such that if the absolute value of the difference between $\sin^2\theta$ values divided by a set of integers is less than a certain amount, the hkl indices of each line can be found. Thus, statement 112 in the saved version is as follows:

112 FOR U = 0.00005 to 0.007 STEP 0.00005.

This provision causes the difference to increase in increments from 0.00005 to 0.007 in steps of 0.00005. Therefore, when a set of d -spacings is indexed, the best index is the first index that is printed out. This fact is true because the difference, U , increases in increments. If two different indexed patterns are printed out for the same U -value, it may be necessary to change the increment in statement 112 (ie, 112 for $U = 0.00005$ to 0.007 STEP 0.00001). When only one index is printed out, followed by ALL LINES CHECKED FOR A DIFFERENCE LESS THAN U , then this is the best index on the basis of a cubic system.

8. Note that d -spacings, corresponding $\sin^2\theta$, $\sin^2\theta/S$ (where S is $H^2 + K^2 + L^2$), S , and $A(o)$ values, will be typed out. Then the average $A(o)$, and type of system (face centered, body centered, or simple cubic) will be typed out, followed by: THE DIFFERENCE IS LESS THAN U and ALL LINES CHECKED FOR A DIFFERENCE LESS THAN U when all possibilities for a certain difference, U , are printed out.
9. If the $A(o)$ values differ very much, the system may not be cubic, in which case, tetragonal should be tried next, then hexagonal if it is not indexed on tetragonal.

Indexing Patterns of Tetragonal Crystals

Two programs are used: the first, TENDX1, calculates $A(o)$; the second, TENDX2, calculates $C(o)$.

1. Call TENDX1 by typing OLD and responding TENDX1 to the computer question PROBLEM NAME. Wait for READY.
2. Type KEY. Wait for READY.

3. Type 350 DATA D, E, L1, where D is the number of d-spacings; E is normally five, which is the number of $H^2 + K^2$ values for tetragonal ($L = 0$); L1 is the wavelength, usually $\text{CuK}\alpha$ and 1.54178. The statement "390 DATA" contains the five values of $H^2 + K^2$.
4. Type 355 DATA $d_1, d_2, \dots, d_n$ as for cubic. Data can be put into lines 355-389. Type SAVE; wait for READY. Then type LISTNH:355; and, if extra data are present from some other run, erase it by typing the statement number.
5. Type RUN.
6. In this program, V is the value that the difference must be less than. In the saved version, it is as follows:

67 FOR V = 0.00005 to 0.007 STEP 0.00005.

7. Note that the maximum number of equal values (the maximum number of HK0 lines) d-spacings, $\sin^2\theta, \sin^2\theta/S$ ($S = H^2 + K^2$), ($H^2 + K^2, L = 0$), and $A(o)$ values will be printed out, followed by the average $A(o)$, the number of remaining lines to be indexed, AVER CAP A for TET2, THE DIFFERENCE IS LESS THAN V, and ALL LINES CHECKED FOR A DIFFERENCE LESS THAN V, MAX = B.
8. Call TENDX2 as before for TENDX1.
9. Add data in 450 DATA D, E, L2, L1, A, where D is the number of remaining lines from TENDX1; E is normally ten, which is the number of $H^2 + K^2$ values; L2 is normally six, which is the number of L values; L1 is the wavelength, usually for $\text{CuK}\alpha$ and is 1.54178; A is AVER CAP A from TENDX1. The statement "460 DATA" contains the ten ($H^2 + K^2$) values; "465 DATA" contains the six L values.
10. Type 455 DATA $d_1, d_2, \dots, d_n$, where only the d-spacings not indexed in TENDX1 are used. Data can be put in lines 455-459. Type SAVE; wait for READY. List the data to be sure no extra data from past runs are included. If data are present, erase them by typing the statement number.
11. Type RUN.
12. Statement 132 in the saved version contains the value that the difference must be less than. The statement in the saved version is as follows:

132 FOR U = 0.0005 to 0.007 STEP 0.0005.

The starting point (0.0005) and the increment were increased by a factor of ten over that in TENDIX1 because of the longer time required for execution of TENDX2.

13. If the $C(o)$ values have large variations, it may be necessary to try the hexagonal system. However, it is possible due to the values of the difference in $\sin^2\theta$ values in this program for the $(H^2 + K^2)$ and L values to be the same for different d -spacings, which is impossible. Therefore, if, for the lowest difference (U value), all the d -spacings are indexed with nearly equal $C(o)$ values, but the program has printed extra (or double) values of $(H^2 + K^2)$ and L^2 for different d -spacings, the pattern is indexed, but the average $C(o)$ must be calculated from the d -spacings that have been indexed and give equal $C(o)$ values. Of course, different planes have the same d -spacing and this can account for some of the double values.

Indexing Patterns of Hexagonal Crystals

This series of programs follows the same procedure as for the tetragonal. $HXNDX1$ calculates $A(o)$; $HXNDX2$ calculates $C(o)$. The following are differences in $HXNDX1$ (saved version) compared to $TENDX1$:

67 FOR $V = 0.00005$ to 0.007 STEP 0.00005 ,

355-389 DATA $d_1, d_2, \dots, d_n$, and

350 DATA $D, E, L1$, where D is the number of d -spacings; E is normally four, which is the number of $H^2 + HK + K^2$ values ($L = 0$); $L1$ is usually 1.54178 .

The following are differences in $HXNDX2$ (saved version) compared to $TENDX2$:

450 DATA $D, E, L2, L1, A$, where D is the number of d -spacings not indexed by $HXNDX1$; E is the number of $H^2 + HK + K^2$ values (normally ten); $L2$ is six, which is the number of L values; $L1$ is usually 1.54178 ; A is AVER CAP A for $HEX2$ from $HXNDX1$,

455-459 DATA $d_1, d_2, \dots, d_n$, and

132 FOR $U = 0.0005$ to 0.007 STEP 0.0005 .

COMPUTER PROGRAMS

A listing of each of the computer programs used to index the patterns of cubic, tetragonal, and hexagonal materials is as shown.

CUINDEX Program

CUINDEX 08:49 CEIR THU. 05/01/69

```

5 DIM H(20),Z(20),S(50),X(20,50),O(30),R(25),G(25),A(25),B(25)
10 DIM J(25),Y(30)
15 READ D,E,I1,N,L1
20 FOR I = 1 TO D
25 READ H(I)
30 LET Z(I) = (L1+2/(4*H(I)+2))
35 NEXT I
40 FOR J = 1 TO E
45 READ S(J)
50 NEXT J
55 FOR V = 1 TO I1
60 READ U(V)
65 NEXT V
70 FOR V = 1 TO N
75 READ Y(V)
80 NEXT V
85 FOR I = 1 TO D
90 FOR J = 1 TO E
95 LET X(I,J) = Z(I)/S(J)
100 NEXT J
105 NEXT I
110 LET I = 1
112 FOR U = .00005 TO .007 STEP .00005
115 FOR L = 1 TO E
120 LET M = 1
125 LET K = 1
130 FOR J = 1 TO E
135 IF (I+K)>D THEN 240
140 IF J <= L THEN 160
145 LET Q = ABS(X(I,L)-X((I+K),J))
150 IF Q<U THEN 190
155
160 NEXT J
165 GO TO 180
170 LET K = K+1
175 GO TO 160
180 NEXT L
185 GO TO 470
190 LET A(I) = X(I,L)
195 LET O(I) = S(L)
200 LET R(I) = Z(I)
205 LET G(I) = H(I)
210 LET M = M+1
215 LET A(M) = X((I+K),J)
220 LET O(M) = S(J)
225 LET G(M) = H(M)
230 LET R(M) = Z(I+K)
235 GO TO 170
240 IF M<D THEN 180
245 LET T = 0
250 FOR M = 1 TO D
255 LET B(M) = SQR((L1+2)/(4*A(M)))
260 LET T = T+B(M)
265 NEXT M
270 LET F = T/M
275 PRINT "D SPAC","SNSQTA","SQT/S","S","A(0)"
280 FOR M = 1 TO D
285 PRINT H(M),R(M),A(M),O(M),B(M)
290 NEXT M

```

```

295 PRINT "AVERAGE VALUE OF A(0) =" F
300 LET W = 0
305 FOR M = 1 TO D
310 FOR V = 1 TO I1
315 LET Q = ABS(O(M)-U(V))
320 IF Q=0 THEN 340
325 NEXT V
330 NEXT M
335 GO TO 350
340 LET W = W+1
345 GO TO 330
350 IF W<D THEN 365
355 PRINT "FACE CENTERED CUBIC"
357 PRINT "THE DIFFERENCE IS LESS THAN " U
360 GO TO 180
365 LET W = 0
370 FOR M = 1 TO D
375 FOR V = 1 TO N
380 LET Q = ABS(O(M)-Y(V))
385 IF Q = 0 THEN 405
390 NEXT V
395 NEXT M
400 GO TO 415
405 LET W = W+1
410 GO TO 395
415 IF W<D THEN 430
420 PRINT "BODY CENTERED CUBIC"
422 PRINT "THE DIFFERENCE IS LESS THAN " U
425 GO TO 180
430 PRINT "SIMPLE CUBIC"
432 PRINT "THE DIFFERENCE IS LESS THAN " U
435 GO TO 180
440 DATA 7
442 DATA 50,20,28,1.54178
445 DATA 5.6,4.03,3.6,2.49,2.35,2.06,1.778
446 DATA
450 DATA 1,2,3,4,5,6,8,9,10,11,12,13,14,16,17,18,19,20,21,22,24,25,26
455 DATA 27,29,30,32,33,34,35,36,37,38,40,41,42,43,44,45,46,48,49
456 DATA 50,51,52,53,54,56,57,58
460 DATA 3,4,8,11,12,16,19,20,24,27,32,35,36,40,43,44,48,51,52,56
465 DATA 2,4,6,8,10,12,14,16,18,20,22,24,26,30,32,34,36,38,40,42,44
466 DATA 46,48,50,52,54,56,58
470 PRINT "ALL LINES CHECKED FOR A DIFFERENCE LESS THAN" U
472 NEXT U
475 STOP
480 END

```

TENDX1 Program

TENDX1 10:45 CEIR THU. 05/01/69

```

5 DIM H(10),Z(10),S(10),X(10,10),A(10),U(150),C(10),O(10)
6 DIM R(10),G(10),F(10)
10 READ D,E,L1
15 FOR I = 1 TO D
20 READ H(I)
25 LET Z(I) = (L1*2)/(4*H(I)+2)
30 NEXT I
35 FOR J = 1 TO E
40 READ S(J)
45 NEXT J
50 FOR I = 1 TO D
55 FOR J = 1 TO E
60 LET X(I,J) = Z(I)/S(J)

```

```

62 NEXT J
64 NEXT I
65 LET F = 1
67 FOR V = .00005 TO .007 STEP .00005
69 LET T = 0
70 FOR I = 1 TO D
75 FOR L = 1 TO E
80 LET M = 1
81 LET K = 1
85 FOR J = 1 TO E
86 IF (I+K)>D THEN 117
87 IF J<=L THEN 105
90 LET Q = ABS(X(I,L)-X((I+K),J))
95 IF Q<V THEN 135
105 NEXT J
110 LET K = K+1
115 GO TO 85
117 LET T = T+1
119 LET U(T) = M
120 NEXT L
125 NEXT I
130 GO TO 145
135 LET M = M+1
140 GO TO 110
145 LET B = 0
150 FOR I = 1 TO T
155 IF U(I)<=B THEN 170
160 LET B = U(I)
170 NEXT I
171 IF B<=F THEN 395
172 LET F = B
175 PRINT "MAX NO. OF EQUAL VALUES="B
176 FOR I = 1 TO D
177 FOR L = 1 TO E
181 LET K = 1
182 LET M = 1
185 FOR J = 1 TO E
190 IF (I+K)>D THEN 272
192 IF J<=L THEN 205
195 LET Q = ABS(X(I,L)-X((I+K),J))
200 IF Q<V THEN 225
205 NEXT J
210 LET K = K+1
215 GOTO 185
216 NEXT L
218 NEXT I
220 GO TO 395
225 LET C(1) = X(I,L)
230 LET O(1) = S(L)
235 LET R(1) = Z(I)
240 LET G(1) = H(I)
245 LET M = M+1
250 LET C(M) = X((I+K),J)
255 LET O(M) = S(J)
260 LET R(M) = Z(I+K)
265 LET G(M) = H(I+K)
270 GO TO 210
272 IF M<B THEN 216
275 LET Y = 0
280 LET W = 0
285 FOR A = 1 TO M
290 LET F(A) = SQR((L1+2)/(4*C(A)))
295 LET W = W + F(A)
300 LET Y = Y + C(A)
305 NEXT A

```

```

308 LET P = W/M
311 LET N= Y/M
315 PRINT "D SPAC","SNSQTA","SQT/S","H+2+K+2,L=0","A(0)"
320 FOR A = 1 TO M
325 PRINT G(A),R(A),C(A),O(A),F(A)
330 NEXT A
335 PRINT " AVERAGE VALUE OF A(0) ="P
340 PRINT "REMAINING LINES ="(D-M)
345 PRINT "AVER. CAP. A FOR TET2=" N
346 PRINT "THE DIFFERENCE IS LESS THAN" V
347 GO TO 216
350 DATA 7.5,1.54178
355 DATA 5.6,4.03,3.6,2.49,2.35,2.06,1.778
390 DATA 1,2,4,5,8
395 PRINT "ALL LINES CHECKED FOR A DIFFERENCE LESS THAN" V      "MAX ="B
397 NEXT V
400 STOP
405 END

```

TENDX2 Program

TENDX2 08:59 CEIR THU. 05/01/69

```

5 DIM H(10),Z(10),S(10),L(10),X(10,10),C(30),O(30),R(30)
10 DIM G(30),Y(30),B(30)
15 READ D,E,L2,L1,A
20 FOR I = 1 TO D
25 READ H(I)
30 LET Z(I) = (L1+2)/(4*H(I)+2)
35 NEXT I
40 FOR I = 1 TO E
45 READ S(I)
50 NEXT I
55 FOR I = 1 TO L2
60 READ L(I)
65 NEXT I
90 FOR I = 1 TO D
95 FOR J = 1 TO E
100 LET X(I,J) = Z(I) -A*S(J)
105 IF X(I,J)<=.0005 THEN 125
110 NEXT J
115 NEXT I
120 GO TO 135
125 LET X(I,J) = 0
130 GO TO 110
132 FOR U = .0005 TO .007 STEP .0005
135 FOR I = 1 TO D
140 FOR W = 1 TO E
145 LET M = 1
150 FOR N = 1 TO L2
155 LET P = 1
160 LET K = 1
161 IF I = D THEN 163
162 GO TO 165
163 LET K = 0
165 LET P = P+1
170 FOR J = 1 TO E
175 IF (I+K)>D THEN 370
180 IF (I+P)<1 THEN 255
185 IF X(I,W) = 0 THEN 270
190 IF P<0 THEN 205
195 IF X((I+K),J) = 0 THEN 245
200 GO TO 215

```

```

205 IF X((I+P),J) = 0 THEN 165
210 GO TO 225
215 LET Q = ABS((L(N)+2)*X(I,W)-X((I+K),J))
220 GO TO 230
225 LET Q = ABS((L(N)+2)*X(I,W)-X((I+P),J))
230 IF Q<U THEN 280
235 NEXT J
240 IF P<0 THEN 165
245 LET K = K+1
250 GO TO 170
255 NEXT N
260 GO TO 385
265 NEXT W
270 NEXT I
275 GO TO 470
280 LET C(1) = X(I,W)
285 LET O(1) = S(W)
290 LET R(1) = Z(I)
295 LET G(1) = H(I)
300 LET Y(1) = 1
305 LET M = M+1
310 IF P<0 THEN 340
315 LET C(M) = (X((I+K),J))/(L(N)+2)
320 LET O(M) = S(J)
325 LET R(M) = Z(I+K)
330 LET G(M) = H(I+K)
335 GO TO 360
340 LET C(M) = (X((I+P),J))/(L(N)+2)
345 LET O(M) = S(J)
350 LET R(M) = Z(I+P)
355 LET G(M) = H(I+P)
360 LET Y(M) = L(N)
365 GO TO 240
370 IF I>1 THEN 380
375 GO TO 255
380 GO TO 160
385 IF M<D THEN 265
390 LET T=0
395 FOR V = 1 TO M
400 LET B(V) = SQR((L1+2)/(4*C(V)))
405 LET T = T + B(V)
410 NEXT V
415 LET F = T/M
420 PRINT "D SPAC","SNSQTA","H+2+K+2","L VALUE","C(0)"
425 FOR V = 1 TO M
430 PRINT G(V),R(V),O(V),Y(V),B(V)
435 NEXT V
440 PRINT "AVERAGE VALUE OF C(0)="F
442 PRINT "THE DIFFERENCE IS LESS THAN" U
445 GO TO 265
450 DATA 4,10,6,1.54178,.00458
455 DATA 2.49,2.35,2.06,1.778
460 DATA 0,1,2,4,5,8,9,10,13,16
465 DATA 1,2,3,4,5,6
470 PRINT "ALL LINES CHECKED FOR A DIFFERENCE LESS THAN" U
472 NEXT U
475 STOP
480 END

```

HXNDX1 Program

HXNDX1 10:37 CEIR THU. 05/01/69

```

5 DIM H(10),Z(10),S(10),X(10,10),A(10),U(150),C(10),O(10)
6 DIM R(10),G(10),F(10)
10 READ D,E,L1

```

```

15 FOR I = 1 TO D
20 READ H(I)
25 LET Z(I) = (L1+2)/(4*H(I)+2)
30 NEXT I
35 FOR J = 1 TO E
40 READ S(J)
45 NEXT J
50 FOR I = 1 TO D
55 FOR J = 1 TO E
60 LET X(I,J) = Z(I)/S(J)
62 NEXT J
64 NEXT I
65 LET F = 1
67 FOR V = .00005 TO .007 STEP .00005
69 LET T = 0
70 FOR I = 1 TO D
75 FOR L = 1 TO E
80 LET M = 1
81 LET K = 1
85 FOR J = 1 TO E
86 IF (I+K)>D THEN 117
87 IF J<=L THEN 105
90 LET Q = ABS(X(I,L)-X((I+K),J))
95 IF Q<V THEN 135
105 NEXT J
110 LET K = K+1
115 GO TO 85
117 LET T = T+1
119 LET U(T) = M
120 NEXT L
125 NEXT I
130 GO TO 145
135 LET M = M+1
140 GO TO 110
145 LET B = 0
150 FOR I = 1 TO T
155 IF U(I)<=B THEN 170
160 LET B = U(I)
170 NEXT I
171 IF B<=F THEN 395
172 LET F = B
175 PRINT "MAX NO. OF EQUAL VALUES="B
176 FOR I = 1 TO D
177 FOR L = 1 TO E
181 LET K = 1
182 LET M = 1
185 FOR J = 1 TO E
190 IF (I+K)>D THEN 272
192 IF J<=L THEN 205
195 LET Q = ABS(X(I,L)-X((I+K),J))
200 IF Q<V THEN 225
205 NEXT J
210 LET K = K+1
215 GOTO 185
216 NEXT L
218 NEXT I
220 GO TO 395
225 LET C(I) = X(I,L)
230 LET O(I) = S(L)
235 LET R(I) = Z(I)
240 LET G(I) = H(I)
245 LET M = M+1
250 LET C(M) = X((I+K),J)
255 LET O(M) = S(J)
260 LET R(M) = Z(I+K)
265 LET G(M) = H(I+K)

```

```

270 GO TO 210
272 IF M<B THEN 216
275 LET Y = 0
280 LET W = 0
285 FOR A = 1 TO M
290 LET F(A) = SQR((L1+2)/(3*C(A)))
295 LET W = W + F(A)
300 LET Y = Y + C(A)
305 NEXT A
308 LET P = W/M
311 LET N= Y/M
315 PRINT "DSPAC","SNSQTA","SQT/S","H+2+HK+K+2, L=0","A(0)"
320 FOR A = 1 TO M
325 PRINT G(A),R(A),C(A),O(A),F(A)
330 NEXT A
335 PRINT " AVERAGE VALUE OF A(0) ="P
340 PRINT "REMAINING LINES ="(D-M)
345 PRINT "AVER. CAP. A FOR HEX2="N
346 PRINT "THE DIFFERENCE IS LESS THAN" V
347 GO TO 216
350 DATA 7,4,1.54178
355 DATA 2.473,2.308,2.091,1.687,1.342,1.332,1.237
390 DATA 1,3,4,7
395 PRINT "ALL LINES CHECKED FOR A DIFFERENCE LESS THAN" V      "MAX=" 8
397 NEXT V
400 STOP
405 END

```

HXNDX2 Program

HXNDX2 09:09 CEIR THU. 05/01/69

```

5 DIM H(10),Z(10),S(10),L(10),X(10,10),C(30),O(30),R(30)
10 DIM G(30),Y(30),B(30)
15 READ D,E,L2,L1,A
20 FOR I = 1 TO D
25 READ H(I)
30 LET Z(I) = (L1+2)/(4*H(I)+2)
35 NEXT I
40 FOR I = 1 TO E
45 READ S(I)
50 NEXT I
55 FOR I = 1 TO L2
60 READ L(I)
65 NEXT I
90 FOR I = 1 TO D
95 FOR J = 1 TO E
100 LET X(I,J) = Z(I) -A*S(J)
105 IF X(I,J)<=.0005 THEN 125
110 NEXT J
115 NEXT I
120 GO TO 135
125 LET X(I,J) = 0
130 GO TO 110
132 FOR U = .0005 TO .007 STEP .0005
135 FOR I = 1 TO D
140 FOR W = 1 TO E
145 LET M = 1
150 FOR N = 1 TO L2
155 LET P = 1
160 LET K = 1
161 IF I = D THEN 163

```



```

162 GO TO 165
163 LET K = 0
165 LET P = P-1
170 FOR J = 1 TO E
175 IF (I+K)>D THEN 370
180 IF (I+P)<1 THEN 255
185 IF X(I,W) = 0 THEN 270
190 IF P<0 THEN 205
195 IF X((I+K),J) = 0 THEN 245
200 GO TO 215
205 IF X((I+P),J) = 0 THEN 165
210 GO TO 225
215 LET Q = ABS((L(N)+2)*X(I,W)-X((I+K),J))
220 GO TO 230
225 LET Q = ABS((L(N)+2)*X(I,W)-X((I+P),J))
230 IF Q<U THEN 280
235 NEXT J
240 IF P<0 THEN 165
245 LET K = K+1
250 GO TO 170
255 NEXT N
260 GO TO 385
265 NEXT W
270 NEXT I
275 GO TO 470
280 LET C(1) = X(I,W)
285 LET O(1) = S(W)
290 LET R(1) = Z(I)
295 LET G(1) = H(I)
300 LET Y(1) = 1
305 LET M = M+1
310 IF P<0 THEN 340
315 LET C(M) = (X((I+K),J))/(L(N)+2)
320 LET O(M) = S(J)
325 LET R(M) = Z(I+K)
330 LET G(M) = H(I+K)
335 GO TO 360
340 LET C(M) = (X((I+P),J))/(L(N)+2)
345 LET O(M) = S(J)
350 LET R(M) = Z(I+P)
355 LET G(M) = H(I+P)
360 LET Y(M) = L(N)
365 GO TO 240
370 IF I>1 THEN 380
375 GO TO 255
380 GO TO 160
385 IF M<D THEN 265
390 LET T=0
395 FOR V = 1 TO M
400 LET B(V) = SQR((L+2)/(4*C(V)))
405 LET T = T + B(V)
410 NEXT V
415 LET F = T/M
420 PRINT "DSPAC","SNSQTA","H+2+HK+K+2","LVALUE","C(0)"
425 FOR V = 1 TO M
430 PRINT G(V),R(V),O(V),Y(V),B(V)
435 NEXT V
440 PRINT "AVERAGE VALUE OF C(0)="F
442 PRINT "THE DIFFERENCE IS LESS THAN " U
445 GO TO 265
450 DATA 5,10,6,1.54178,.0268786
455 DATA 5,6,4,03,3,6,2,49,2,06
460 DATA 0,1,3,4,7,9,12,13,16,19
465 DATA 1,2,3,4,5,6
470 PRINT "ALL LINES CHECKED FOR A DIFFERENCE LESS THAN" U

```

472 NEXT U
475 STOP
480 END

Examples of the CUINDEX Program

Copper (ASTM value $A_0 = 3.6153A$, face-centered cubic)

```

CUINDEX      09:40      CEIR      THU. 05/01/69

D SPAC      SNSQTA      SQT/S      S      A(0)
2.088      .136309      4.54363E-2      3      3.61652
1.808      .181798      4.54494E-2      4      3.616-
1.278      .363851      4.54813E-2      8      3.61473
1.09      .500186      4.54715E-2      11     3.61512-
1.0436     .545653      4.54711E-2      12     3.61514-
.9038      .727512      4.54695E-2      16     3.6152-
.8293      .864095      4.54787E-2      19     3.61483-
.8083      .909577      4.54789E-2      20     3.61483-
* AVERAGE VALUE OF A(0) = 3.6153
FACE CENTERED CUBIC
THE DIFFERENCE IS LESS THAN .00005
D SPAC      SNSQTA      SQT/S      S      A(0)
2.088      .136309      2.27181E-2      6      5.11453
1.808      .181798      2.27247E-2      8      5.1138-
1.278      .363851      2.27407E-2      16     5.112-
1.09      .500186      2.27357E-2      22     5.11255-
1.0436     .545653      2.27355E-2      24     5.11257-
.9038      .727512      2.27347E-2      32     5.11266
.8293      .864095      2.27393E-2      38     5.11215-
.8083      .909577      2.27394E-2      40     5.11214
AVERAGE VALUE OF A(0) = 5.1128
BODY CENTERED CUBIC
THE DIFFERENCE IS LESS THAN .00005
ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .00005-

```

Note that the first index (*) was used, since the second contains S values which are a multiple of two times the first and is therefore not the value of the smallest unit cell.

Yttrium Sesquioxide (ASTM value $A_0 = 10.604A$, body-centered cubic)

```

CUINDEX      09:59      CEIR      THU. 05/01/69

D SPAC      SNSQTA      SQT/S      S      A(0)
4.34      3.15504E-2      5.25841E-3      6      10.6308-
3.06      6.34661E-2      5.28884E-3      12     10.6002-
2.652     8.44963E-2      5.28102E-3      16     10.608-
2.5      9.50834E-2      5.28241E-3      18     10.6066-
2.372     .105622      5.28111E-3      20     10.6079-
2.261     .116248      5.28398E-3      22     10.605
2.165     .126785      5.28272E-3      24     10.6063
2.08      .137359      5.28305E-3      26     10.606-

```

1.936	.158553	5.2851E-3	30	10.6039-
1.874	.169218	5.28805E-3	32	10.6009-
1.818	.179803	5.28833E-3	34	10.6007
1.769	.189902	5.27505E-3	36	10.614-
1.72	.200876	5.28621E-3	38	10.6028-
1.677	.211309	5.28273E-3	40	10.6063-
1.636	.222033	5.28651E-3	42	10.6025-
1.599	.232428	5.28245E-3	44	10.6066-
1.563	.243258	5.28821E-3	46	10.6008-
1.531	.253533	5.28194E-3	48	10.6071
1.499	.264473	5.28946E-3	50	10.5995
1.47	.275011	5.28867E-3	52	10.6003

AVERAGE VALUE OF A(0) = 10.6058
 BODY CENTERED CUBIC
 THE DIFFERENCE IS LESS THAN .00005
 ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .00005-

Calculation of A(o) and C(o) for Zinc (ASTM value A(o) = 2.664A, C(o) = 4.945, hexagonal)

The following example shows the results of two programs necessary to index tetragonal and hexagonal materials. Since two equal possibilities occurred in the HXNDX1 program, the smaller value was used in HXNDX2. Because a complete index was obtained with the smaller value, it was concluded to be correct since the unit cell is the smallest repeatable unit in a structure. An asterisk indicates the smaller value of A(o) used:

Part I - Calculation of A(o)

HXNDX1 10:22 CEIR THU. 05/01/69
 ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .00005 MAX= 1
 MAX NO. OF EQUAL VALUES= 2
 DSPAC SNSQTA SQT/S H+2+HK+K+2, L=0
 A(0)
 2.473 .097171 .097171 1 2.85557
 1.237 .38837 9.70924E-2 4 2.85673-
 AVERAGE VALUE OF A(0) = 2.85615
 REMAINING LINES = 5
 AVER. CAP. A FOR HEX2= 9.71317E-2
 THE DIFFERENCE IS LESS THAN .0001
 DSPAC SNSQTA SQT/S H+2+HK+K+2, L=0
 A(0)
 2.308 .111561 .111561 1 2.66505-
 1.332 .334947 .111649 3 2.664-
 *AVERAGE VALUE OF A(0) = 2.66452
 REMAINING LINES = 5
 AVER. CAP. A FOR HEX2= .111605
 THE DIFFERENCE IS LESS THAN .0001
 ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .0001 MAX= 2

Part II - Calculation of C(o)

HXNDX2 10:29 CEIR THU. 05/01/69

ALL LINES CHECKED FOR A DIFFERENCE LESS THAN 0

ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .0005

DSPAC	SNSQTA	H+2+HK+K+2	LVALUE	C(O)
2.091	.135918	1	1	4.94395-
1.687	.208812	1	2	4.94509-
2.473	.097171	0	2	4.946-
1.342	.329974	1	3	4.94901
1.237	.38837	0	4	4.948-

AVERAGE VALUE OF C(O) = 4.94641

THE DIFFERENCE IS LESS THAN .001

ALL LINES CHECKED FOR A DIFFERENCE LESS THAN .001

When accurate (corrected) d-spacing values are obtained, as seen from the examples, these programs provide an excellent method of indexing powder diffraction patterns. For copper (the face-centered cubic example), the calculated value of A(o) agreed with the ASTM value to the nearest ten-thousandths of an angstrom. For yttrium sesquioxide, the calculated value of A(o) differed from the ASTM value by only 0.0018 A. For the hexagonal example (zinc), the difference in A(o) was correct to the nearest thousandth of an angstrom; C(o) differed from the ASTM value by 0.00141 A. It should be noted that these lattice parameters represent numerical average values and should be further refined by standard methods.