ALPHA-SILICON CARBIDE, TYPE 51R

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ABSTRACT

The morphological and *x*-ray crystallography, including indexed powder diffraction data, optical properties, etching behavior and chemical analyses of a very rare modification of α-SiC, formerly designated type V, are given in detail. Referred to the smallest hexagonal cell, $a_0=3.073$ Å; $c_0=128.17$ Å. Formula weights in this cell $=51$. Space group $=C_{3v}^5-R3m$. Densities: observed $=3.218$; calculated $=3.217$.

INTRODUCTION

Twenty years ago Ott (1928) described a modification of silicon carbide containing 51 formula weights per hexagonal unit cell and designated it SiC, type V. Although a morphological study of the crystal could not be made, *x*-ray study by means of rotation and oscillation photographs established the following constants:

Hexagonal unit: $a_0=3.09_5$ Å; $c_0=129.0_3$ Å; $Z=51$.
Rhombohedral unit: $a_{rh}=43.1_5$ Å; $\alpha=4°\ 06'$; $Z=17$.

During the author's comprehensive study of the SiC types (Thibault, 1944) no crystals of this modification were encountered, but more recently a large, very well-developed specimen of this type was found quite by accident. Preliminary data on this crystal were given in a paper presented before the Crystallographic Society, March 1946 (Thibault, 1946). Ramsdell (1947) has published Weissenberg data obtained from *x*-ray photographs made of the same crystal, and has substantiated the structure which he had previously deduced (Ramsdell, 1946) from Ott's original data.

Professor Ramsdell has also suggested a more logical method of designating the different modifications. This consists of the number of formula weights in the hexagonal unit cell (rhombohedral types being referred to the hexagonal unit) followed by the letter "H" or "R" depending upon whether the unit cell is hexagonal or rhombohedral. This method of notation appears to be quite satisfactory and is followed in the present paper.

Quite independently Zhdanov and Minervina (1945*a*, *b*, *c*) determined the same structure using Ott's original data.

OPTICAL PROPERTIES

The crystal upon which the present paper is based is illustrated by Fig. 1. It was approximately 18×10×5 mm., being attached to a dense mass of SiC at the 18×5 mm. section.

Although macroscopically jet black, a plate cut about 1 mm. thick parallel to the *c*-axis showed the following pleochroism: ϵ = medium blue; ω = dark blue. Therefore absorption: $\omega > \epsilon$.

Indices of refraction were determined by using sulfur-selenium melts and a black crystal of α-SiC, type 6H, as control. As nearly as could be determined by the method used, ω was the same as that of the control, namely, about 2.63 for the essentially Li-light transmitted by the melt.

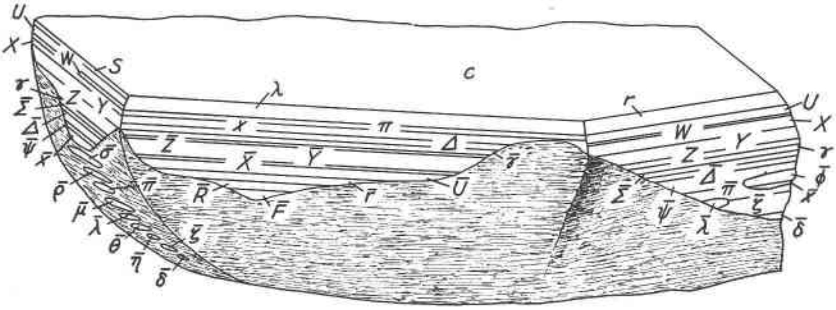

Fig. 1. Alpha-SiC, type 51R (formerly type V).

The index ϵ could not be determined, but it was greater than 2.66. The interference figure was uniaxial positive.

Morphological Crystallography

Fortunately the crystal was extremely well-developed, containing 76 well-established and 5 somewhat uncertain faces; it was, moreover, entirely free from intergrowths with any of the other α-SiC types.

Table 1 gives the complete morphological data for type 51R; Table 2 includes an angle table, while Table 3 lists the uncertain forms. Although showing rhombohedral-like face development with different series of forms in alternate pyramid zones, these series were entirely different from any of the other SiC types, only the basal pinacoid and the first order pyramids r–$\bar{r}$ and x–$\bar{x}$ being equivalent to forms found on any of the other types.

The observed axial ratio offering the greatest simplification of form indices and yielding simple arithmetical series of forms characteristic of crystals with rhombohedral lattices was 41.699. This is well within experimental error rationally related to the axial ratio of type 6H by a factor of $8\frac{1}{2}$. Because the $c:a$ value of type 6H was determined with great precision, and because the axial ratio of all the other types have also been rationally related to type 6H within experimental error, $4.9070 \times 8\frac{1}{2}$ or 41.710 is accepted as the axial ratio of type 51R.

TABLE 1. MORPHOLOGICAL DATA, α-SiC, TYPE 51R (FORMERLY TYPE V)

Form	Number Times Observed	Quality	Angle Between Form and Base		
			Measured Range	Weighted Average	Calculated Value
c 0001	1	A	—	—	0° 00′
$\underline{F}$ $1.0.\bar{1}.43$	1	E	48° 17′	48° 17′	48° 14½′
$\bar{F}$ $0.1.\bar{1}.\overline{43}$	1	C	48° 14½′	48° 14½′	
$\bar{R}$ $0.1.\bar{1}.\overline{40}$	1	C	50° 18′	50° 18′	50° 17½′
r $1.0.\bar{1}.34$	2	C-D	54° 45½′–54° 49′	54° 47½′	54° 47′
$\bar{r}$ $0.1.\bar{1}.\overline{34}$	2	B-C	54° 44′ –54° 47½′	54° 46′	
S $1.0.\bar{1}.31$	2	C-D	57° 15′ –57° 16′	57° 15½′	57° 14′
T $1.0.\bar{1}.28$	1	C	59° 52′	59° 52′	59° 49½′
$\underline{U}$ $1.0.\bar{1}.25$	3	C-D	62° 34½′–62° 35½′	62° 35′	62° 34′
$\bar{U}$ $0.1.\bar{1}.\overline{25}$	2	A-C	62° 33′ –62° 34′	62° 33½′	
V $1.0.\bar{1}.22$	1	D	65° 28′	65° 28′	65° 27′
$\underline{W}$ $1.0.\bar{1}.19$	3	C-E	68° 25′ –68° 32′	68° 28′	68° 28′
$\bar{W}$ $0.1.\bar{1}.\overline{19}$	1	E	68° 28′	68° 28′	
$\underline{X}$ $1.0.\bar{1}.16$	3	B-C	71° 37½′–71° 40′	71° 38½′	71° 37½′
$\bar{X}$ $0.1.\bar{1}.\overline{16}$	2	C-D	71° 30′ –71° 38′	71° 35′	
$\underline{Y}$ $1.0.\bar{1}.10$	2	C-D	78° 21′ –78° 24′	78° 22′	78° 16′
$\bar{Y}$ $0.1.\bar{1}.\overline{10}$	2	B	78° 13′ –78° 18½′	78° 16′	
$\underline{Z}$ $10\bar{1}7$	3	B-C	81° 39½′–81° 53′	81° 45′	81° 44′
$\bar{Z}$ $01\bar{1}\bar{7}$	2	B-C	81° 42½′–81° 44′	81° 43′	
γ $10\bar{1}1$	3	C	88° 46½′–88° 49′	88° 47½′	88° 48½′
$\bar{\gamma}$ $01\bar{1}\bar{1}$	2	C	88° 46′ –88° 49½′	88° 48′	
$\bar{\delta}$ $1.0.\bar{1}.\overline{50}$	2	C-E	43° 54½′–43° 56½′	43° 56′	43° 55½′
$\bar{\zeta}$ $1.0.\bar{1}.\overline{44}$	2	B-E	47° 36′ –47° 41′	47° 37′	47° 35′
$\bar{\eta}$ $1.0.\bar{1}.\overline{41}$	1	D	49° 35′	49° 35′	49° 35½′
$\bar{\theta}$ $1.0.\bar{1}.\overline{38}$	1	C	51° 44½′	51° 44½′	51° 43½′
$\underline{\lambda}$ $0.1.\bar{1}.35$	2	C	54° 00′	54° 00′	53° 59½′
$\bar{\lambda}$ $1.0.\bar{1}.\overline{35}$	2	E	53° 59′ –54° 06′	54° 02½′	
$\bar{\mu}$ $1.0.\bar{1}.\overline{32}$	1	D	56° 22½′	56° 22½′	56° 24′
π $0.1.\bar{1}.26$	2	C	61° 38′ –61° 40′	61° 39′	61° 38½′
$\bar{\pi}$ $1.0.\bar{1}.\overline{26}$	3	B-D	61° 33′ –61° 38′	61° 35½′	
$\bar{\rho}$ $1.0.\bar{1}.\overline{23}$	1	C	64° 25′	64° 25′	64° 28½′
σ $0.1.\bar{1}.20$	1	E	67° 21′	67° 21′	67° 27′
$\bar{\sigma}$ $1.0.\bar{1}.\overline{20}$	1	B	67° 25′	67° 25′	

TABLE I—*Continued*

Form	Number Times Observed	Quality	Angle Between Form and Base		
			Measured Range	Weighted Average	Calculated Value
x $0.1.\bar{1}.17$	2	B-D	70° 33′ -70° 38′	70° 36′	70° 33½′
$\bar{x}$ $1.0.\bar{1}.\overline{17}$	3	C-E	70° 27½′-70° 30′	70° 29′	
$\bar{\phi}$ $1.0.\bar{1}.\overline{14}$	1	E	73° 50½′	73° 50½′	73° 47½′
$\bar{\psi}$ $1.0.\bar{1}.\overline{11}$	3	C-E	77° 03′ -77° 06′	77° 04½′	77° 08′
Δ $01\bar{1}8$	2	D	80° 33½′-80° 35½′	80° 34½′	80° 34′
$\bar{\Delta}$ $10\overline{18}$	3	B-C	80° 30′ -80° 36′	80° 33½′	
$\bar{\Sigma}$ $10\overline{12}$	3	C-E	87° 35½′-87° 41′	87° 38½′	87° 37½′

TABLE 2. α-SIC, TYPE 51R (FORMERLY TYPE V), ANGLE TABLE

Hexagonal $-R$; ditrigonal pyramidal $-3m$

$a:c = 1:41.710$ $\alpha = 4°\,07'$

$p_0:r_0 = 48.163:1$ $\lambda = 119°\,57'$

Lower	Upper		ϕ	ρ	A_1	A_2
	c	0001	—	0° 00′	90° 00′	90° 00′
$\bar{F}$	F	$1.0.\bar{1}.43$	+30° 00′	48° 14½′	49° 45½′	90° 00′
$\bar{R}$		$1.0.\bar{1}.40$	+30° 00′	50° 17½′	48° 13½′	"
$\bar{r}$	r	$1.0.\bar{1}.34$	"	54° 47′	44° 58′	"
	S	$1.0.\bar{1}.31$	"	57° 14′	43° 16′	"
	T	$1.0.\bar{1}.28$	"	59° 49½′	41° 31½′	"
$\bar{U}$	U	$1.0.\bar{1}.25$	"	62° 34′	39° 46′	"
	V	$1.0.\bar{1}.22$	"	65° 27′	38° 01½′	"
$\bar{W}$	W	$1.0.\bar{1}.19$	"	68° 28′	36° 20′	"
$\bar{X}$	X	$1.0.\bar{1}.16$	"	71° 37½′	34° 43½′	"
$\bar{Y}$	Y	$1.0.\bar{1}.10$	"	78° 16′	32° 01′	"
$\bar{Z}$	Z	$10\bar{1}7$	"	81° 44′	31° 01′	"
$\bar{\gamma}$	γ	$10\bar{1}1$	"	88° 48½′	30° 01½′	"
$\bar{\delta}$		$0.1.\bar{1}.50$	−30° 00′	43° 55½′	90° 00′	53° 04½′
$\bar{\zeta}$		$0.1.\bar{1}.44$	"	47° 35′	"	50° 15½′
$\bar{\eta}$		$0.1.\bar{1}.41$	"	49° 35½′	"	48° 44½′
$\bar{\theta}$		$0.1.\bar{1}.38$	"	51° 43½′	"	47° 10′
$\bar{\lambda}$	λ	$0.1.\bar{1}.35$	"	53° 59½′	"	45° 31½′
$\bar{\mu}$		$0.1.\bar{1}.32$	"	56° 24′	"	43° 50′
$\bar{\pi}$	π	$0.1.\bar{1}.26$	"	61° 38½′	"	40° 21′
$\bar{\rho}$		$0.1.\bar{1}.23$	"	64° 28½′	"	38° 36′
$\bar{\sigma}$	σ	$0.1.\bar{1}.20$	"	67° 27′	"	36° 53½′
$\bar{x}$	x	$0.1.\bar{1}.17$	"	70° 33½′	"	35° 15′
$\bar{\phi}$		$0.1.\bar{1}.14$	"	73° 47½′	"	33° 44′
$\bar{\psi}$		$0.1.\bar{1}.11$	"	77° 08′	"	32° 24½′
$\bar{\Delta}$	Δ	$01\bar{1}8$	"	80° 34′	"	31° 19′
$\bar{\Sigma}$		$01\bar{1}2$	"	87° 37½′	"	30° 05′

TABLE 3. UNCERTAIN FORMS, α-SiC, TYPE 51R (FORMERLY TYPE V)

Form	No. Times Observed	Angle Between Form and Base	
		Observed	Calculated
$\overline{T}$ $0.1.\bar{1}.\overline{28}$	1	59° 12′	59° 49½′
$10\bar{1}4$	1	85° 50′	85° 15′
$1.0.\bar{1}.\overline{47}$	1	45° 31′	45° 42′
$10\bar{1}\bar{5}$	2	83° 45′ 83° 57½′	84° 04½′

ETCHING FIGURES

Small chips removed from the crystal were etched both by immersion in a borax melt at red heat for about 2 hours, and by partial chlorination at about 1200° C. followed by removal of the resulting carbon by oxidation.

In both cases the etch figures were very similar in appearance to those formed on α-SiC, types 15R, 21R, and 33R (Thibault, 1944), and the same symmetry elements are indicated. The crystal class of type 51R is thus ditrigonal-pyramidal, or $3m$ of the hexagonal system. The single basal pinacoid observed on the 51R crystal is the upper form, in conformity with the arbitrary decision made in this respect when the other types were studied in detail.

CHEMICAL ANALYSES AND MEASURED DENSITY

After the crystal had been studied optically and morphologically, additional small pieces were chipped off the upper base (each chip including basal pinacoid and first order pyramid faces) for equi-inclination Weissenberg and powder diffraction studies. Then the remainder of the crystal was crushed in a steel mortar until it all passed a 200 mesh screen. After ignition in an inclined tube furnace at 900° C. in oxygen for 15 minutes to remove any free carbon which might have been present, the sample was treated with HF-HNO_3 to remove any SiO_2 or free Si as well as iron introduced by powdering the sample. An *x*-ray powder photograph of a portion of the sample so prepared showed exactly the same pattern as obtained from the chips. There was, therefore, no morphological or *x*-ray evidence of the presence of any other SiC modification.

Mr. R. M. Rebert, Norton Worcester laboratories, determined the density, d at 30°/4° C. = 3.218, using a 5 cc. pycnometer, and xylene as the displacing liquid. This is the same density as he previously obtained from light green crystals of α-SiC, type 6H (Thibault, 1944).

Mr. Rebert then analyzed the sample according to the method outlined by Lamar (1939). In the following table this analysis is compared with a spectrographic analysis of a portion of the same sample as reported by

Mr. W. M. Hazel, Norton Chippawa laboratories, and with the analysis of light green, type 6H, crystals as previously reported.

Element	α-SiC, type 51R		Theoretical	α-SiC, type 6H
	Spectrographic Analysis	Quantitative Analysis		(Thibault, 1944)
Si	major	69.64%	70.03%	69.78%
C	not det.	29.91	29.97	29.99
Al	minor	0.05		0.01
Fe	minor	0.20		0.10
Ca	minor	0.16		0.16
Mg	minor	<0.01		0.01
Cu	faint trace	not det.		not det.
Ti	faint trace	not det.		not det.
Na	faint trace	not det.		not det.
B	faint trace	not det.		not det.
Total		99.96		100.05

Both analyses indicate a carbon content close to theoretical, the minor elements apparently substituting for Si in the structure. The higher content of iron and/or aluminum in type 51R is probably the cause of its black color compared with the transparent green of the type 6H crystals analyzed.

X-RAY CRYSTALLOGRAPHY

Powder Diffraction Studies. In order to be certain that the sample used for powder diffraction studies was entirely type 51R it was prepared from chips removed from the crystal adjacent to the upper base where morphological study had indicated no intergrowth with any other α-SiC type. The chips were crushed in a steel mortar until all passed a 200 mesh screen, the magnetic removed with an Alnico hand magnet, and the sample further ground for some time in a boron carbide ("Norbide") mortar. For use with the Norelco Geiger-Counter X-ray Spectrometer a portion of the ground sample was flowed onto a glass slide using a few drops of dioxane as the liquid medium, no binder being employed. For use in the powder camera, a portion of the sample was mixed with a minute amount of library paste which itself gave no interfering pattern and extruded in the form of a rod approximately $\frac{1}{2}$ mm. in diameter using the technique described by Lukesh (1940).

Complete scans of nearly 90° (2θ) were made using the Brown recorder with chart travel of $\frac{1}{2}''$ per minute coupled to the Norelco spectrometer which was operated under the following conditions:

x-ray slit width	1 mm.
x-ray slit length	6 mm.
Geiger slit width	$\frac{1}{4}$ mm.
Geiger slit length	6 mm.
Amplitude	Maximum
Damping	Minimum
Time constant	5 seconds
Scanning speed	1° (2θ) per min.

Scans were made both with filtered copper and with filtered iron radiations. Although the former is to be preferred because of the greater intensity of the reflections and much more satisfactory recording of the weaker ones, the pattern obtained by the use of iron radiation was very useful because of better resolution of closely-spaced reflections. Figure 2 is a reproduction of the pattern obtained with filtered iron radiation of

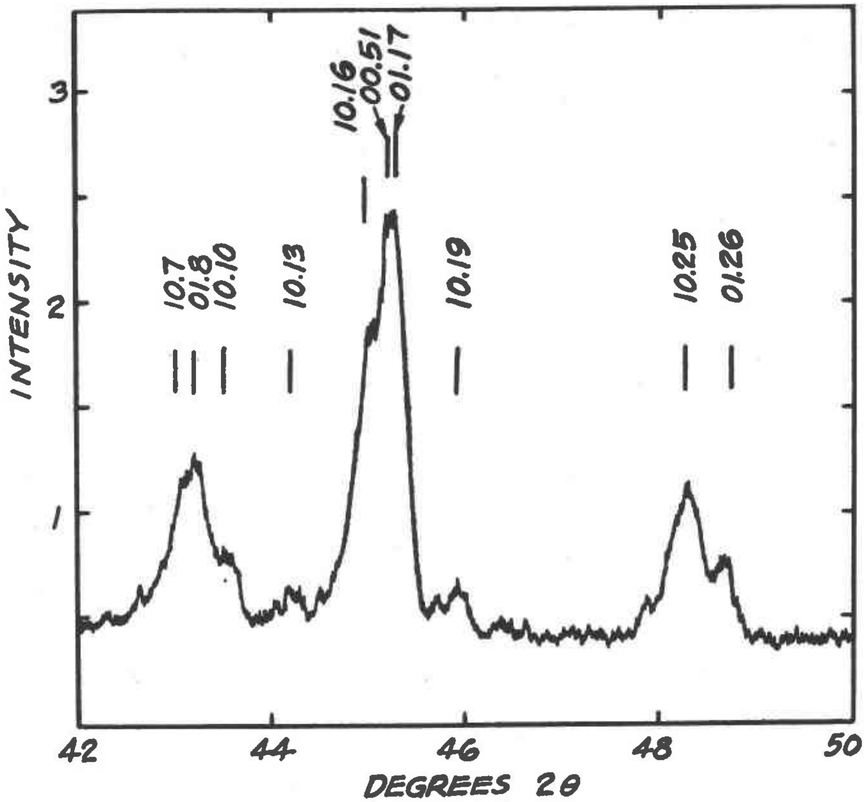

FIG. 2. Pattern obtained from α-SiC, type 51R, using Norelco x-ray spectrometer with filtered iron radiation. Interval: 42°–50° 2θ. *Calculated* positions of the reflections indicated by vertical lines.

the first few reflections in the powder pattern, the interval being 42° – 50° (2θ). Note the very excellent agreement between the observed position of the reflections as recorded and the calculated positions indicated by the vertical lines in the upper portion of the figure.

Regular powder photographs were made with the Norelco one radian camera using filtered copper radiation and a collimated beam approximately $\frac{1}{2}$ mm. in diameter. The specimen was rotated but not translated during exposure. Figure 3 is a reproduction of the powder pattern obtained, the portion covered by the scan of Fig. 2 being indicated by the arcs drawn adjacent to the photograph.

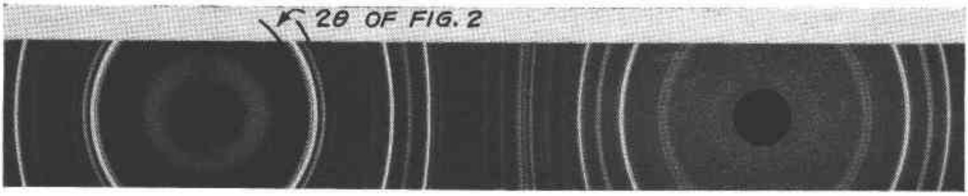

FIG. 3. *X*-ray powder photograph of α-SiC, type 51R. CuK_α radiation. Camera diameter about 57.3 mm. Portion included in Fig. 2 indicated by the arcs.

Data obtained from the various powder diffraction studies are given in Table 4. Values are for the $CuK\alpha_1$ reflections where resolved, otherwise for $CuK\alpha_1$ and α_2. For 2θ up to 90°, the intensity data were derived largely from the spectrometer curves made with filtered copper radiation. Here the strongest reflection was arbitrarily designated 10, the weakest, 1. Reflections not observed on the spectrometer curves, but visible on the powder photographs were assigned intensity values of <1. For 2θ greater than 90°, intensity data were derived from the powder photographs. The reflections were indexed by correlation with Weissenberg exposures, *a*-axis rotations, zero and first levels. Although indexing of the reflections offered little difficulty in the forward reflecting position, considerable uncertainty was often present in the back reflection because of the great number of planes which might contribute to the powder pattern. In many cases where the reflections from two or more planes are practically coincident, it is often impossible to determine whether some of the possible planes actually contributed to the observed reflections. Such cases are indicated by question marks in Table 4. The calculated values for $d_{hk.l}$ were derived from $a_0 = 3.073$ Å, $c_0 = 128.17$Å,* the accepted hexagonal unit cell dimensions of type 51R. The rhombohedral unit cell is $a_{rh} = 42.76$ Å, $\alpha = 4°07'$.

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* To be consistent with the earlier work on SiC, the cell dimensions are given in Å, although they are actually kX units.

TABLE 4. POWDER DIFFRACTION DATA FOR α-SIC, TYPE 51R, (FORMERLY TYPE V)

"Line" No.	Intensity	*hk.l*	Equipment*	$d_{hk\,l}$ (Å)	
				Observed	Calculated
1	5	10.7	G, Fe	2.630	2.634
1a		01.8	G, Fe	2.625	2.625
2	2	10.10	G, Fe	2.604	2.606
3	2	10.13	G, Fe	2.567	2.569
4	3	10.16	G, Fe	2.526	2.526
5	10	00.51 01.17	G, Fe	2.511	2.513 2.510
6	2	10.19	G, Fe	2.478	2.476
7	1	10.22	G, Cu	2.422	2.421
8	4	10.25	G, Fe	2.362	2.362
9	3	01.26	G, Fu	2.343	2.342
10	1	01.32	G, Cu	2.214	2.217
11	2	10.34	G, Cu	2.173	2.174
12	2	01.35	G, Cu	2.152	2.152
13	1	01.41	G, Cu	2.026	2.026
14	1	10.43	G, Cu	1.984	1.985
15	<1	10.58	P, Cu	1.704	1.700
16	1	01.59	G, Cu	1.685	1.683
17	1	10.61	G, Cu	1.650	1.649
18	<1	10.64	P, Cu	1.596	1.600
19	2	10.67	G, Cu	1.553	1.555
20	5	01.68 11.0	G, Cu	1.538	1.538 1.537
21	<1	10.70	P, Cu	1.510	1.509
22	<1	10.73	P, Cu	1.459	1.466
23	2	10.76	G, Cu	1.426	1.425
24	2	01.77	G, Cu	1.414	1.411
25	1	01.83	G, Cu	1.328	1.335
26	5	02.16 10.85 11.51 20.17?	G, Cu	1.313	1.313 1.312 1.311 1.310
27	1	01.86	G, Cu	1.302	1.300
28	1	02.25	G, Cu	1.290	1.288
29	1	20.26	G, Cu	1.287	1.285
30	1	00.102 02.34?	G, Cu	1.256	1.257 1.255
31	1	20.35	G, Cu	1.253	1.251
32	<1	02.43 10.94	P, Cu	1.214	1.215 1.214
33	1	20.59	G, Cu	1.137	1.135
34	<1	02.61	P, Cu	1.125	1.124
35	1	02.67	G, Cu	1.095	1.092
36	1	20.68	G, Cu	1.090	1.087
37?	<1	02.70	P, Cu	1.073	1.077
38	1	02.76	P, Cu	1.042	1.045
39	1	20.77	P, Cu	1.037	1.039

TABLE 4—(*continued*)

"Line" No.	Intensity	*hk.l*	Equipment*	$d_{hk.l}$ (Å)	
				Observed	Calculated
40	1	10.118 12.8? 21.10? 01.119 02.85 21.16	P, Cu	1.001	1.006 1.004 1.003 .998 .998 .998
41	2	12.17 20.86	P, Cu	.995	.997 .993
42	1	21.25 12.26 10.121?	P, Cu	.984	.987 .986 .984
43	2	11.102 21.34 12.35	P, Cu	.971	.973 .972 .970
44	<1	02.94	P, Cu	.952	.952
45	1	10.127	P, Cu	.942	.944
46	1	01.128	P, Cu	.936	.937
47	<1	12.59	P, Cu	.913	.913
48	<1	21.61	P, Cu	.906	.907
49	<1	21.64	P, Cu	.898	.899
50	1	21.67 10.136	P, Cu	.889	.890 .888
51	4	30.0 12.68	P, Cu	.886	.887 .887
52?	<1	01.137 21.70	P, Cu	.882	.883 .882
53	2	21.76	P, Cu	.864	.864
54	2	12.77	P, Cu	.861	.861
55	<1	02.118	P, Cu	.841	.841
56	5	10.145 00.153 30.51 20.119 21.85	P, Cu	.837	.839 .838 .837 .837 .837
57?	<1	12.86 02.121?	P, Cu	.832	.834 .829
58	<1	02.124 12.92	P, Cu	.816	.816 .816
59	<1	21.94	P, Cu	.809	.809
60	2	12.95? 02.127	P, Cu	.805	.806 .804
61	2	20.128	P, Cu	.801	.800
62	<1	20.131	P, Cu	.787	.788
63	1	20.134	P, Cu	.777	.777

* Equipment used: G = Norelco Geiger-Counter *X*-ray Spectrometer; P = Photographic method, 1 radian camera; Fe = filtered iron radiation; Cu = filtered copper radiation.

Weissenberg Photographs. Professor L. S. Ramsdell kindly made zero and first level, *a*-axis rotation Weissenberg exposures of the type 51R crystal. A number of planes which are common to most of the α-SiC modifications are indexed in Fig. 4, the zero level, *a*-axis rotation Weissenberg which has also been reproduced by Ramsdell (1947).

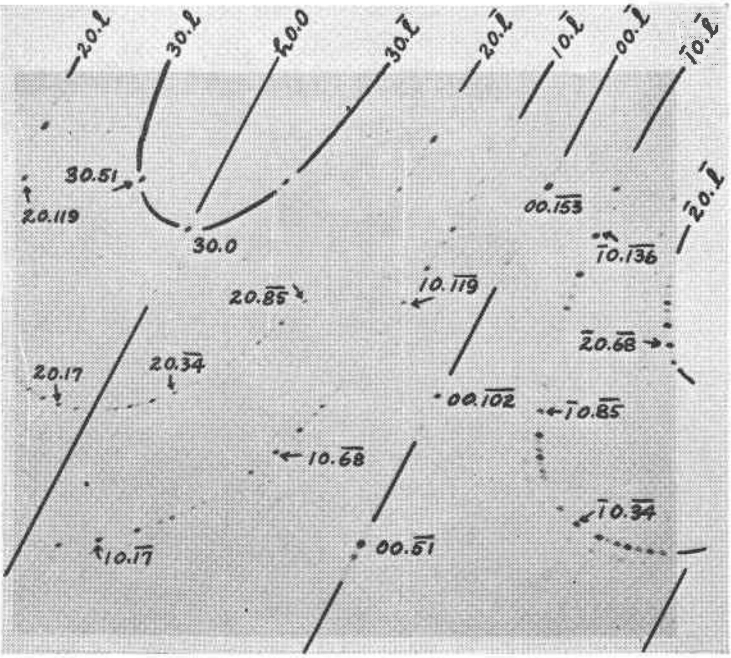

FIG. 4. Equi-inclination Weissenberg photograph of α-SiC, type 51R; *a*-axis rotation, zero level.

The space group of type 51R is obviously the same as that of the other rhombohedral types, C_{3v}^5-R3m, and the calculated density is 3.217, in good agreement with the observed density of 3.218.

ACKNOWLEDGMENTS

The author is indebted to Miss Pauline Krukonis for making many of the calculations necessary in the course of the study, and to Norton Company for permission to publish the paper.

REFERENCES

All published papers on the crystallography of SiC which have appeared since the author's previous work (Thibault, 1944) as well as the articles cited in the present paper are included here.

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