NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Technical Report 32-1201

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Abstract

Tensile fracture stress data at room temperature are presented for a series of hot-pressed polycrystalline MgO and TaC specimens. An unacceptably large number of failures in the end section of the MgO indicated the need for improved fabrication techniques. Data for the $TaC_{0.93}$ indicate a fracture stress of approximately 25 kg/mm^2 (35,000 psi) and a Young's modulus of 2.8×10^4 to 2.9×10^4 kg/mm^2 (39×10^6 psi). Termination of the program before its completion prevented such improvement and the generation of sufficient data for constructive analysis. Consequently, data are presented in detail to permit subsequent analysis.

The Mechanical Behavior of Tantalum Carbide and Magnesium Oxide

I. Introduction

Additional knowledge is greatly needed on the mechanical behavior of polycrystalline ceramics, since mechanical failure is the most frequently encountered shortcoming of ceramics. Also, pure structural applications of ceramics are infrequently encountered, and the selection of a ceramic for a particular application is usually based on its thermal, electrical, or chemical properties. However, such components often fail by fracture, even during installation, before they have been exposed to service conditions.

Since such mechanical failure is so important, and since grain boundaries can be expected to have a large influence on this behavior in polycrystalline ceramics, an extensive study of this interrelationship has been underway at the Jet Propulsion Laboratory. The mechanical property data that are available are often not useful in such a research study, since much of it is concerned with single crystals, while the interest in this study is in real polycrystalline material. In addition, characterization can never be total; for these studies, detailed knowledge of the nature of the grain boundary is needed.

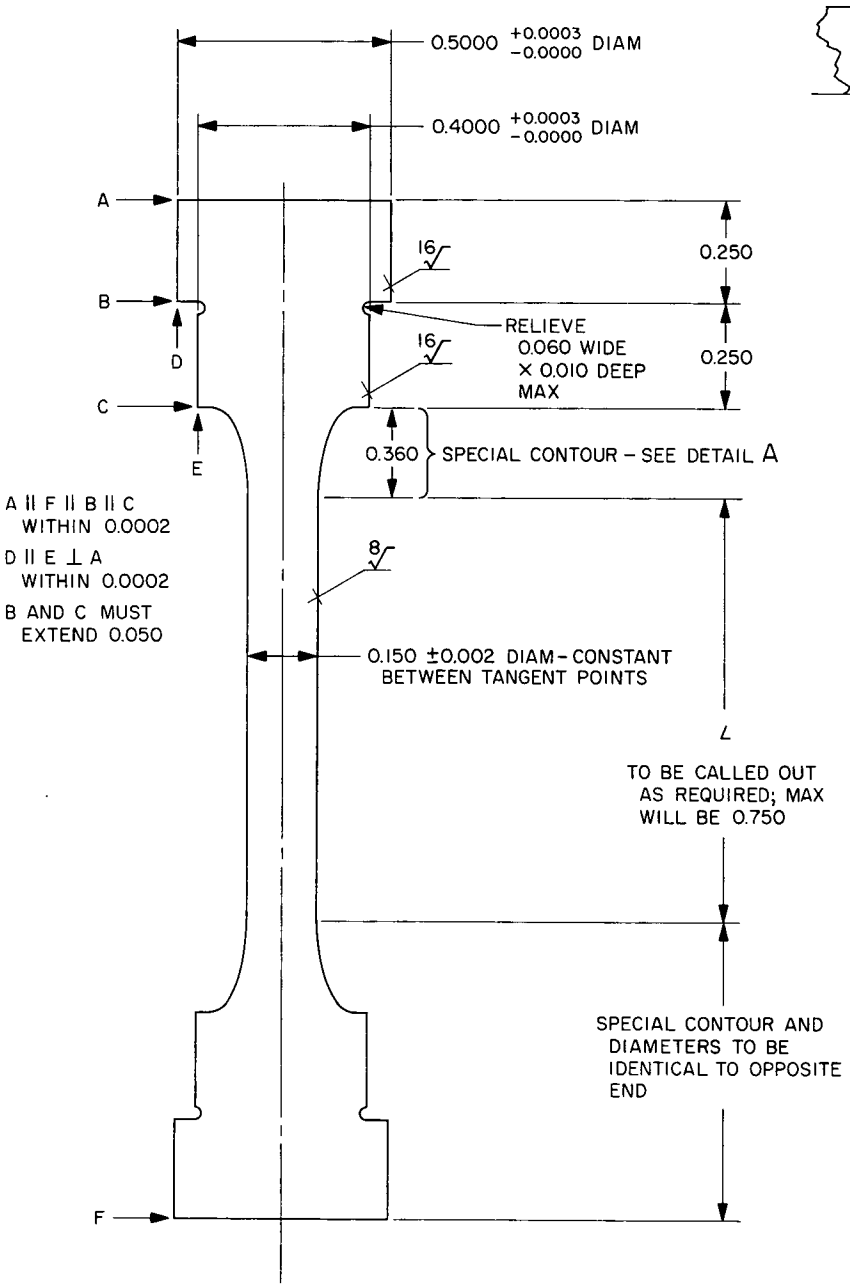
These factors have shown the need for additional mechanical data to be generated as part of this overall program to understand the role of grain boundaries in mechanical behavior. The concepts invoked in the design and selection of experiments toward clarifying this relationship are reported elsewhere (Ref. 1). Likewise, other reports describe the development of a mechanical tension testing facility for brittle materials (Ref. 2) and the studies directed toward the understanding of grain boundary structure (Ref. 3).

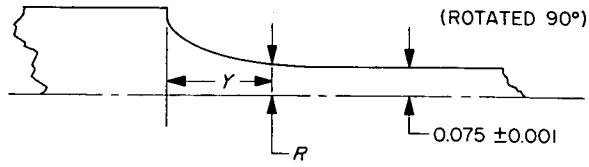
The area covered by this report is limited strictly to the specific preparation of specimens and the observations and results recorded during their mechanical testing. It should be noted that the results described in this report, as well as the other phases of this activity previously referenced, are incomplete as a result of termination of this activity. For this reason, these results are presented in detail in an effort to provide usable information for subsequent analysis. Little in the way of general conclusions can be drawn.

The materials being investigated in this study were selected primarily on the basis of their typifying brittle ceramics. The two materials chosen are magnesium oxide and tantalum carbide. Specimens of both types were produced from blanks densified by means of hot-pressing.

The blanks (¾-in. diameter × 2½-in. long) were diamond ground to a diameter of 0.55 in. and were checked for density and visual flaws. If satisfactory, the blanks were pot-chucked and then finished ground to the contour shown in Fig. 1. In many cases, a final grinding to each end of the specimen was performed in a surface grinder with the specimen held in a vertical position by a machined jig block.

The specimens were then loaded into the grips using the procedure described in the appendix of this report, and were tested to fracture. The testing procedure employed a crosshead rate of 0.005 cm/min; however, the presence of the air-floated bearings introduced considerable *softening* in the load system, and the actual rate of elongation in the specimen was approximately 2×10^{-4} min^{-1}. The postfracture analysis consisted primarily of preparing the





Y	R	Y	R
0.0000	0.1228	0.090	0.0890
0.0006	0.1210	0.120	0.0851
0.0015	0.1195	0.150	0.0821
0.003	0.1180	0.180	0.0802
0.006	0.1152	0.210	0.0789
0.012	0.1112	0.240	0.0778
0.018	0.1080	0.280	0.0770
0.024	0.1053	0.300	0.0764
0.030	0.1031	0.360	0.0755
0.045	0.0983	∞	0.0750
0.060	0.0945		

DETAIL A

NOTES:
1. ALL DIAMETERS MUST BE PARALLEL AND CONCENTRIC TO AXIS WITHIN 0.0002 *T/R*.
2. SURFACE FINISH OF CONTOURS AND 0.150 ±0.002 DIAM IS CRITICAL.
3. TRANSITION BETWEEN CONTOURS AND 0.150 DIAM MUST BE SMOOTH WITHOUT REDUCTION OF AREA; SCRATCHES, NICKS, OR DISCONTINUITY OF CONTOUR WILL NOT BE ACCEPTED.

(ALL DIMENSIONS ARE IN INCHES)

Fig. 1. Tensile specimen contour

metallographic sections and photographs of the specimen and fracture surface.

II. Tantalum Carbide Specimen Preparation

The tantalum carbide specimens were produced to the required ¾-in. diameter × 2½-in. length by hot-pressing with graphite dies and punches at 2050°C and 9000 psi. The charge consisted of slugs approximately 50% theoretical density of isostatically pressed powder that was obtained commercially (CIBA Corp., Grade 1000) and had a particle size of approximately 0.1 μm. During the development of the procedures for the fabrication of these specimens, considerable variation was encountered in the behavior of the supposedly equivalent lots of powder.

The details of the TaC investigation and conclusions are more extensively noted in a separate report (Ref. 4). For the purpose of preparing these blanks, the various lots of powder were blended and hot-pressed under the conditions noted. The pressed blanks ranged from 94 to 98% of density and showed a lattice parameter of 4.446 Å and corresponded to $TaC_{0.93}$. A typical chemical analysis of hot-pressed TaC (blended lots) was as follows:

Type	Wt %
Combined C	5.8
Free C	≤0.02
N_2	≤0.01
O_2	≤0.02
Total metallic	≤0.05

III. Magnesium Oxide Specimen Preparation

The magnesium oxide compacts used in this study were also formed by means of hot-pressing. The starting materials were either a 99.5% pure MgO from a commercial source (Fisher M-300), or a special grade produced at JPL, as reported in Refs. 1 and 5. One sample was produced from a commercial source* of $Mg(OH)_2$ calcined at JPL (Ref. 1).

The blanks prior to machining into specimens were ¾ in. in diameter by approximately 2½ in. long. The blanks were hot-pressed either in graphite dies at 4000 psi and 1600°C for ½ h, or in composite die cases at 13,000 psi and at temperatures of 975°C for the JPL material and 1050°C for the commercial materials. The composite die cases consisted of an aluminum oxide liner with a molybdenum TZM outer jacket. The two parts were sized such that, at the temperatures used, the internal stresses were supported by the molybdenum jacket.

*Kanto Chemical Works, Tokyo, Japan.

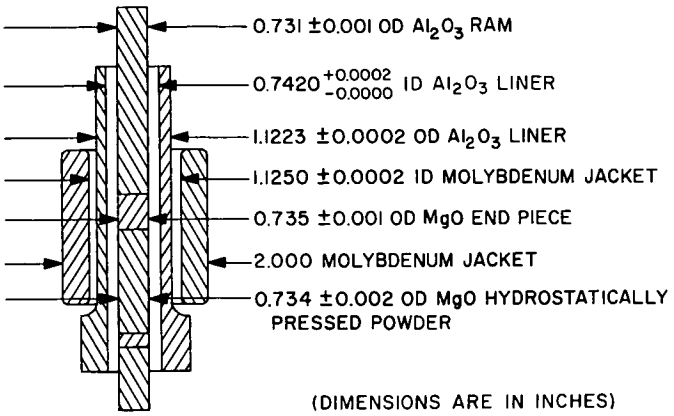


Fig. 2. Configuration used in composite die hot-pressing of MgO

Figure 2 shows the sizes and tolerances of the various components used in this system. Table 1 gives the typical chemical analysis for the two primary types of specimens.

The specimens fabricated in the composite dies used a combination of hot-pressing and hot-pressed welding to obtain the desired length of specimen. A compact of isostatically pressed powder was placed between two end sections of conventionally sintered 99% pure 97% dense

Table 1. Mass spectrographic analyses of hot-pressed Fisher M-300 MgO and JPL MgO

Element	M-300, part/10^6 at.	JPL, part/10^6 at.
H[a]	100	50
B	300	ND[b]
N	100	70
F	3000	300
Na	61	7
Al	100	250
Si	1800	100
P	120	8
S	420	56
Cl	80	38
K	64	2
Ca	310	7
Ti	66	ND
Fe	510	5

[a]Additional H at m/ε = 17 (OH); absolute value is approximate.
[b]ND = not detected.

MgO.* Approximately ½ g of loose powder was placed between the compact and each section end for alignment and to hold the die case in place. This procedure was necessary since the travel required to compact the full length in one operation was beyond the capability of the facility.

Initial evaluation of these welds by examination of the microstructure suggested that the procedure was capable of producing a sound specimen; however, later testing of specimens in the test frame did not support this assumption, as a significant number of specimens failed by weld fracture. The reported density of these specimens was calculated from the known density of the end pieces and the measured density of the compact.

The specimens fabricated in the graphite die system used a conventional charging technique. However, some discoloration existed from the graphite dies and larger grain sizes resulted from the higher pressing temperatures. Some of the specimens pressed in graphite dies were reheated to 1700°C in air for ½ h prior to testing.

IV. Results

The data developed during the course of these tests is summarized in Tables 2, 3, and 4. The total indicator reading (TIR) listed refers to that observed at the lower

*Kaiser Refractories, Milpitas, Calif.

Table 2. Mechanical tensile behavior of polycrystalline MgO (Fisher M-300) at 25°C

Run	Specimen	Density, g/cm³	Fracture stress, kg/mm²	Grain size, μm	Young's modulus, kg/mm² × 10⁻⁴	TIR, in. × 10⁻³	Remarks
Hot-pressed in composite die and tested as pressed							
3	170	3.55	—	ND[a]	—	—	Fractured during assembly
5	285	3.55	—	ND	—	—	Fractured during assembly
11	275	3.54	(8.2)	—	1.8	0.3	Failed in weld
13	279	3.54	9.6	1[b]	2.2	0.6	
16	287	3.56	—	ND	—	—	Fractured during assembly
17	255	3.55	—	ND	—	—	Fractured during assembly
18	300	3.56	(3.75)	ND	ND	1.5	Failed in end section
19	268	3.54	—	ND	—	0.4	Fractured during assembly
21	275	3.52	0.0	ND	ND	ND	Repeat after epoxy; failed in weld
22	197	3.55	—	ND	—	0.5	Fractured during assembly
23	287	3.56	(3.1)	ND	—	ND	Repeat after epoxy; failed in epoxy
24	198	3.57	9.85	1[b]	3.1	0.6	
Hot-pressed in composite die and reheated to 1100°C							
30	283	3.56	(7.9)	ND	2.8	0.5	Fractured in end section
31	245	3.57	(4.45)	ND	ND	0.8	Fractured in end section
32	282	3.57	—	ND	—	—	Fractured during assembly
43	274	3.54	5.4	1[b]	ND	0.5	
Hot-pressed in graphite die and tested as pressed							
6	536	3.50	9.82	22	2.8	0.8	
7	537	3.54	(0.5)	ND	—	ND	Failed at pressing; flaw in end
12	537	3.54	—	ND	—	5.0	Repeat epoxy; joint failed
36	539	3.52	7.75	15	2.0	0.4	
37	540	3.53	11.0	15	2.9	0.2	
49	548	3.50	8.9	27	2.8	0.2	
Hot-pressed in graphite die and reheated to 1700°C							
15	545	3.54	9.8	ND	2.0	0.3	
25	546	3.52	(7.8)	33	2.1	0.5	Fractured in end section
34	544	3.53	(6.61)	26.3	3.3	0.5	Fractured in end section
35	547	3.53	9.20	21	3.4	0.5	
30	541	3.52	10.51	16	3.0	0.2	

[a]ND = not determined.
[b]Estimated.

end of the assembled load train during rotation about the upper end (see appendix). Previous analysis (Ref. 2) indicated that 0.001-in. TIR was equivalent to 2.7% stress variation.

The Young's modulus values were taken from the most linear portion of the stress-strain curves. (Occasional non-linearity was observed during the initial 10^{-4} strain as shown in Fig. 3.)

Grain sizes were determined using a line-count method (Ref. 6) from a single micrograph. When the grain size was too small for optical resolution, the size was estimated from previous determinations.

Table 3. Mechanical tensile behavior of polycrystalline MgO (JPL) at 25°C

Run	Specimen	Density, g/cm³	Fracture stress, kg/mm²	Grain size, μm	Young's modulus, kg/mm² × 10^{-4}	TIR, in. × 10^{-3}	Remarks
Hot-pressed composite die and tested as pressed							
26	350	3.52	4.7	25	3.4	0.3	
27	344	3.56	—	ND[a]	—	0.5	Fractured during assembly
29	134	3.55	3.75	2.0	3.45	0.6	One large grain evident (0.3-mm diameter)
Hot-pressed in composite die and reheated to 1100°C							
41	322	3.49	4.2	9.1	ND	0.5	
42	353	3.53	9.9	ND	3.32	0.3	Fractured in end section
44	172	3.49	—	ND	—	0.5	Fractured during assembly
Kanto $Mg(OH)_2$ caloried at JPL; hot-pressed in composite die and tested as pressed							
33	1.76	3.52	6.22	1[b]	2.8	0.6	

[a]ND = not determined.
[b]Estimated.

Table 4. Mechanical tensile behavior of polycrystalline TaC at 25°C

Run	Specimen	Density, g/cm³	Fracture stress, kg/mm²	Grain size, μm	Young's modulus, kg/mm² × 10^{-4}	TIR, in. × 10^{-3}	Remarks
9	148	14.31	29.7	32.1	2.5	0.3	
10	118	14.29	9.8	26.5	2.5	0.4	Flaw in specimen
14	137	14.05	14.9	31.7	2.9	0.3	
28	161	13.94	28.2	9.2	ND[a]	0.3	
38	173	13.65	13.2	6.2	2.9	0.2	
39	172	13.58	15.2	12.5	2.8	0.1	
40	120	14.05	16.8	23.0	3.6	1.2	
45	162	13.58	20.0	7.2	2.9	0.2	
46	160	13.41	16.8	6.01	2.8	0.5	
47	119	14.03	—	ND	3.0	—	Dimensions inaccurate
48	149	14.11	26.4	32	2.9	1.1	
51	170	13.65	17.5	ND	3.4	0.2	
52	174	13.66	—	ND	—	—	Dimensions inaccurate
53	144	13.24	22.0	ND	2.6	0.4	
54	145	14.30	19.1	ND	3.2	0.2	

[a]ND = not determined.

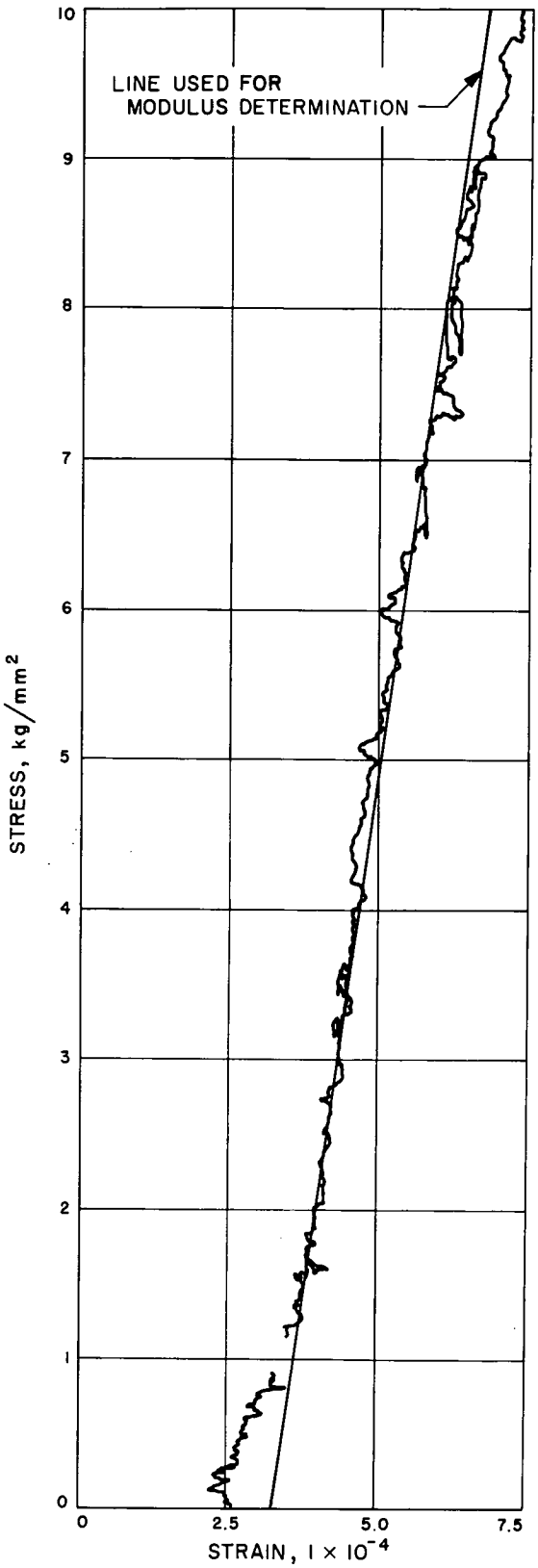


Fig. 3. Stress-strain diagram for MgO run 6

V. Discussion

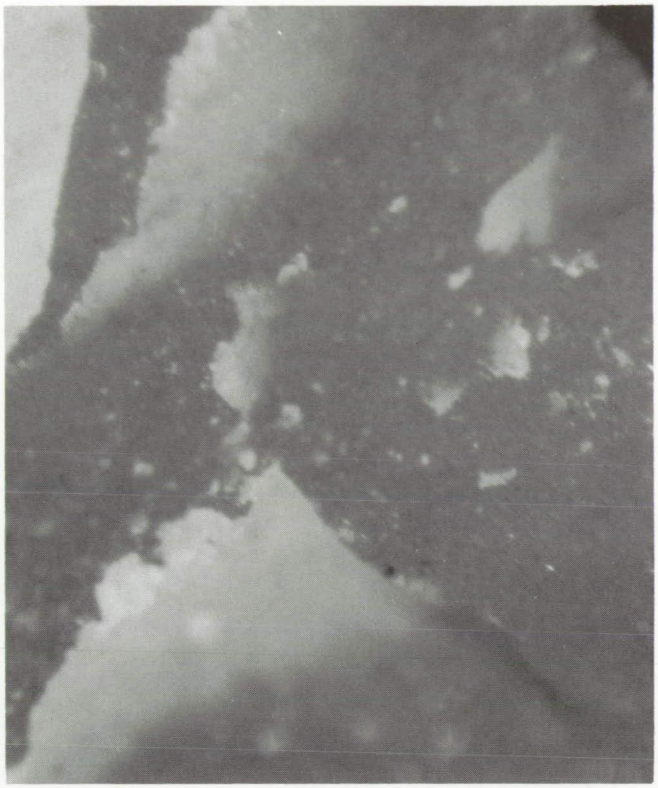
Since there was no opportunity to extensively analyze the data, only very cursory inferences may be drawn. However, these inferences can be valuable in contributing to future activity in the field of tensile testing ceramic materials.

It appears from Table 2 that improvements are needed in the fabrication procedures and in the end portions of the specimen design. Previous analysis of microstructure of the hot-press welded pieces indicated that they were structurally sound; however, several breaks occurred in this region (see runs 18, 22, 30, and 31). Some of these breaks were related to failures in the end sections of the ceramic with the fracture parallel to and near the weld, but not directly in it. Such failures indicated that, although the strength of the hot-pressed MgO was not high, it was apparently greater than that of the conventionally fabricated end pieces.

Several breaks also occurred at the relief groove in the end section of the specimen. It is believed that the failure here was the result of the lower strength of the end section coupled with stress concentrations stemming from the relief groove. The increased cross sectional area at this point was designed to compensate for the groove, but was apparently insufficient. Failure from both of these sources could be reduced by higher-strength end sections. Such material is available from hot-pressing and should be used.

Attempts were made to salvage the data from such specimens by epoxy cementing the two pieces together again and retesting. Alignment was maintained during this procedure by shutting off the air to the air-floated bearings before the two parts of the specimen were completely separated. Because of the counterbalancing of the load train parts, the upper and lower portions remained on the same vertical line and, with the air bearings deactivated, the lower parts of the specimen could be dropped free with the crosshead; the epoxy was applied and then the parts were driven back into place. Although alignment appeared adequate, failure was usually anomalous (either in the epoxy or in the end section); the practice was discontinued.

Examination of the fracture surfaces in the MgO gave evidence for multiple cracking during failure (see Fig. 4). Such multiple cracking could explain the difference between the energy absorbed in the propagation of a crack in a ceramic and the energy involved in the generation of



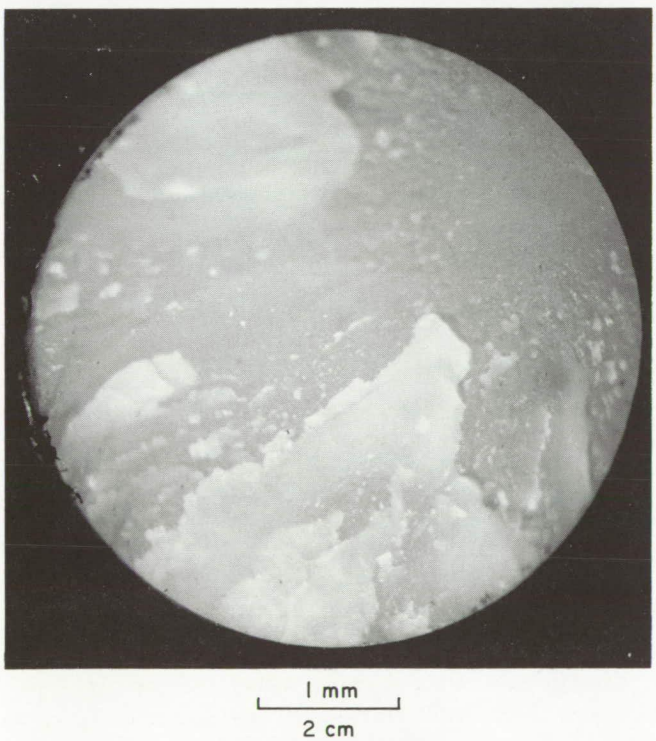


Fig. 4. Fracture surface of MgO run 24 showing extensive multiple cracking

new surface. This difference is sometimes ascribed to plastic work during crack propagation.

The maximum fracture stresses observed in the MgO tested were disappointingly low. The large-grain specimens exhibited strength reasonably consistent with reported values for MgO (Ref. 7). However, the fine-grain specimens were expected to have been considerably stronger. The lack of high strength in these fine-grain materials has been ascribed to the presence of volatile impurities, since increases in strength were observed after reheating the specimens (Ref. 7). Since it is known that these materials contain considerable amounts of volatile impurities, an attempt was made to verify this finding by reheating several specimens for 4 h at 1100°C. However, no increase in strength was noted, but this limited testing should be taken as a disagreement.

The TaC specimens were not a composite piece, therefore, no difficulties were encountered with either failures

Fig. 5. Fracture surface of TaC run 48 showing planar grain surfaces

at weld joints or failures due to the presence of weaker sections. All specimens failed between the shoulders, although, several did exhibit failure in the contour portion of the specimen.

Examination of the fracture surfaces in the TaC exhibited many flat grains suggesting that intragranular fracture is the primary failure mode (Fig. 5). Intergranular facture is to be expected in a material of this relatively coarse-grain size.

Reasonable strengths were obtained of the order of 25 kg/mm^2 in the relatively large-grain material. However, previous work indicated that improvements to strength by means of decreasing the grain size would not be simple, because of the difficulties in controlling the powder characteristics to permit reproducible lower-temperature hot-pressing.

Further, the variation in grain size and density exhibited by these specimens (produced under identical conditions from the same large lot of powder)indicates the inability to precisely reproduce conditions. Such variation likely results from variations in die friction, especially for specimens of such large length-to-diameter ratio.

VI. Conclusions

In general, the status of these results indicates general agreement with available data, and supplies some new data for TaC. However, anomalous failure in many of these early specimens and insufficient time to correct such conditions prevent more than cursory conclusions. These are:

(1) Improved fabrication techniques are required for the production of dense polycrystalline ceramics for mechanical testing in tension.

(2) Fracture-stress increases (in MgO with decrease in grain size) were not noted from this small amount of data. Further, no increase in fracture stress with 1100°C anneal was noted in fine-grain MgO.

(3) Fracture stress of polycrystalline TaC at room temperature appears to be of the order of 25 kg/mm^2 (35,000 psi) in tension for approximately 95% dense and 20-μm grain size. The Young's modulus was approximately 2.8×10^4 to 2.9×10^4 kg/mm^2 (39×10^6 psi).

(4) Transgranular tracking in such TaC was noted.

Appendix
Specimen Assembly

The procedure used in assembling the specimens in the grips was developed to assure axial alignment and concentricity. The components and their assembly are more completely described elsewhere (Ref. 2). The procedure relied on independent evaluation of alignment during assembly using a 0.001-in. dial indicator and a pair of differential distance detectors.*

The upper pull-rod is held in a vertical position in a dummy upper collet, and the specimen and upper grip are assembled. Either the dial indicator or the distance detector may be used to align the lower end of the specimen while the upper grip is tightened. The other pull-rod is then placed in the dummy collet, and the remaining grip assembled on the specimen.

The distance detector is used to align the lower end of the previously assembled grip pull-rod while the second grip is tightened. The distance detector rather than the dial indicator is required for this second operation, because the side loads introduced by the dial indicator cause bending in the specimen resulting in inaccurate readings.

The amount of misalignment was noted and generally was $\leq$ 0.005-in. TIR. Analysis of the bending moments indicates that such misalignment produces uniform distribution of stress to within 3%.

*Bently Scientific Co., Berkeley, Calif.

References

1. Leipold, M. H., Nielsen, T. H., and de Wys, E. C., *The Effect of Grain Boundaries on Mechanical Behavior in Polycrystalline Ceramics*, Technical Report 32-1200. Jet Propulsion Laboratory, Pasadena, Calif. (in preparation).
2. Leipold, M. H., and Becker, P. F., *A Mechanical Tension Testing Facility for Brittle Materials*, Technical Report 32-1179. Jet Propulsion Laboratory, Pasadena, Calif., Oct. 1, 1967.
3. de Wys, E. C., and Leipold, M. H., *The Structure of Grain Boundaries in Ceramics*, Technical Report 32-1211. Jet Propulsion Laboratory, Pasadena, Calif. (in preparation)
4. Leipold, M. H., and Becker, P. F., *Fabrication and Characterization of Hot-Pressed Tantalum Carbide*, Technical Report 32-1204. Jet Propulsion Laboratory, Pasadena, Calif., Oct. 1, 1967.
5. Nielsen, T. H., and Leipold, M. H., "Hot-Pressed High Purity Polycrystalline MgO," *Bull. Am. Ceram. Soc.*, Vol. 45, No. 3, p. 281, 1966.
6. Fullman, R. L., "Measurement of Particle Sizes in Opaque Bodies," *Trans. AIME*, Vol. 197, pp. 447–529, Mar. 1953.
7. Rice, R. W., and Hunt, J. G., *Identifying Optimum Parameters of Hot Extrusions*, joint Interim Report III from The Boeing Company, Space Div., and Whittaker Corporation, Nuclear Metals Div., May 1967. (Available through National Aeronautics and Space Administration, Office of Scientific and Technical Information, Attention AFSS-A, Washington, D.C.)