15th International Plansee Seminar, Eds. G. Kneringer, P. Rödhammer and H. Wildner, Plansee Holding AG, Reutte (2001), Vol. 4

Fracture Toughness of 6.4 mm (0.25 inch) Arc-Cast Molybdenum and Molybdenum-TZM Plate at Room Temperature and 300°C

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Summary:

The fracture toughness of 6.4 mm (0.25 inch) low carbon arc-cast (LCAC) molybdenum and arc-cast molybdenum-TZM alloy plate were measured at room temperature and 300°C using compact tension specimens. The effect of crack plane orientation (longitudinal vs. transverse) and annealing practice (stress-relieved vs. recrystallized) were evaluated. Depending upon the test temperature either a standard K_{IC} or a J-integral analysis was used to obtain the toughness value. At room temperature, regardless of alloy, orientation, or microstructure, fracture toughness values between 15 and 22 MPa $m^{1/2}$ (14 and 20 ksi $in^{1/2}$) were measured. These K_{IC} values were consistent with measurements by other authors. Increasing temperature improves the toughness, due to the fact that one takes advantage of the ductile-brittle transition behavior of molybdenum. At 300°C, the fracture toughness of recrystallized LCAC and arc-cast TZM molybdenum were also similar at approximately 64 MPa $m^{1/2}$ (58 ksi $in^{1/2}$). In the stress-relieved condition, however, the toughness of arc-cast TZM (91 MPa $m^{1/2}$ / 83 ksi $in^{1/2}$) was higher than that of the LCAC molybdenum (74 MPa $m^{1/2}$ / 67 ksi $in^{1/2}$).

Keywords:

molybdenum, TZM, refractory metals, fracture toughness, K_{IC}, J Integral

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1. Introduction:

Background: Brittle failure analysis has become a common consideration in the design of many structures. As a result, fracture toughness properties necessary to conduct brittle fracture analysis of many common materials such as iron, aluminum and titanium alloys have been well characterized. Relatively little effort has been devoted to the characterization of the fracture toughness of molybdenum and molybdenum alloys. Because molybdenum's ductile-to-brittle transition temperature (DBTT) is typically within approximately 100°C of room temperature, fracture - critical components have not typically used molybdenum in this temperature range. Electronic heat sinks, for example, rely on the thermal expansion and conductivity of the material, but do not include fracture considerations in component design. Likewise, the high-temperature applications for molybdenum and its alloys (heat treating furnaces, glass-melting furnace components, e.g.), concern themselves more with long-term creep behavior than with fracture resistance.

In order, for molybdenum and molybdenum alloys to be considered for applications that must exhibit fracture resistance, it is necessary to characterize the toughness of the material. The temperature range between ambient and 1200°C forms the region of most interest with respect to toughness. It is in this region that components such as forging dies, metalworking tooling, and furnace structural parts must operate, and in this range where additional characterization of fracture toughness must be performed.

Prior Work: A few authors have investigated the toughness of molybdenum and molybdenum alloys. Measurements of toughness values were performed on pre-cracked bend bars of extruded arc-cast molybdenum as early as 1963 (1). These measurements used thick cross-sections, and calculated the energy of fracture. Other early work investigated sheet molybdenum and TZM alloy (2), employing thin sheet specimens that frequently displayed plasticity. The state-of-the-art at this time did not permit an appropriate stress-analysis solution for such tests. Furthermore the authors were unable to fatigue pre-crack the pure molybdenum samples without fracture, and relied upon Electro-Discharge Machining (EDM) to produce notches in the specimens. More recently the fracture toughness of molybdenum rod was measured (3). These investigators solved the pre-cracking problem by using compression - compression fatigue, and obtained K_{IC} values for the specimen geometries with which they worked.

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In the preceding work, toughness measurements were obtained at ambient temperatures. Russian investigators have measured the toughness of molybdenum to temperatures of about 500°C (4,5), as have German investigators (6). However, there is no data on the fracture toughness of molybdenum or molybdenum alloys covering temperatures above 500°C, and very little well-characterized data reported between ambient and 500°C.

The results of measurements performed in the preceding references are summarized in Table 1. The toughness numbers vary widely probably due to limitations of the tests, specimen geometries employed, and analytical techniques available at the time of the individual investigations. The most reliable values are probably those of Romine (1), Chen, et. al. (3), and Rödig, et. al. (6).

Current Tests: The testing and analytical tools available to the experimenter today are much improved over those of 20-30 years ago. Analytical techniques like the J-Integral approach (7) have allowed extension of the art into the realm of tests displaying plasticity. The pre-cracking techniques of Chen, et. al. (3) and Rödig, et. al. (6) permit the introduction of sharp notches into less ductile materials like molybdenum. Furthermore, there is increased interest in the fracture behavior of molybdenum and its alloys, particularly at elevated temperatures where the materials are on the upper shelf of their transition curve.

For these reasons, we have set out to measure the fracture toughness of molybdenum and TZM plate at temperatures up to 1100°C using improved techniques. This paper covers the initial measurements made at ambient temperature and at 300°C.

Table 1. K_{IC} measurements of molybdenum and molybdenum alloys at ambient temperatures.

Ref	Product Form	Specimen Type	Crack Dir.	Material	Microstructure	K_{IC}, MPa m$^{1/2}$
1	Extrusion, Mid-rad.	NB	?	A/C Mo	Extruded	17.6[a]
1	Extrusion, OD	NB	?	A/C Mo	Extruded	20.6[b]
2	Sheet	CN	Trans.	A/C Mo	SR	35 - 65[c]
2	Sheet	CN	Trans.	A/C Mo	RX	27 - 33[c]
2	Sheet	CC	Trans.	A/C TZM	SR	40[d]
3	Forged bar	CT	Radial	P/M Mo	Forged	8 ± 5%
3	Forged bar	CT	Radial	P/M TZM	Forged	19 ± 5%
3	Forged bar	SC	Radial	P/M Mo	Forged	17[e]
3	Forged bar	SC	Radial	P/M TZM	Forged	17
3	Swaged bar	SC	Radial	P/M Mo	Swaged	17 ± 11%[f]
3	Swaged bar	SC	Radial	P/M Mo	RX	18 ± 10%[f]
3	Swaged bar	SC	Radial	P/M TZM	RX	31 ± 7%[f]
4	Sheet	CT	Trans.	Mo-1% Zr	RX	15 - 17
5	Sheet	?	⊥Rolling	Mo	?	15 - 60
5	Sheet	?	In-plane	Mo	?	10 - 40
6	Bar	DCT	Radial	P/M TZM	SR	15 - 17

Key: NB = notched bend; CN = center notched; CC = center cracked; CT = compact tension; SC = surface crack; DCT = dia. compact tension; ? = not reported; SR = stress relieved; RX= recrystallized

[a] Average of 5 tests, calculated from G_{IC}; [b] Average of 11 tests, calculated from G_{IC}; [c] K_C value.

[d] Significant plasticity, K_i calculated at deviation of elastic line from linearity.

[e] Specimen displayed sub-critical crack growth, calc at initiation of crack growth.

[f] Specimen displayed sub-critical crack growth, calc using the deepest point of semi-elliptical crack

2. Material Description:

The alloys used in this investigation were consolidated from powder precursors using the Press-Sinter-Melt (PSM) process developed in the 1950s (8). In this process, powder is fed from a hopper enclosed in the vacuum melting furnace, into a pressing die where it is pressed into a disc and fed down into the melting chamber. Repeating this process allows an electrode to be built up which is melted *in vacuo* by an AC arc and solidified in a water-cooled copper crucible.

The cast ingot is then extruded at elevated temperature to break up the large grain structure associated with arc melting. Analytical samples are obtained after extrusion, and Table 2 summarizes the chemical analysis of the two materials. Interstitial elements (C, N, and O) were determined by LECO inert gas fusion and trace elements determined by DC spark/optical emission spectroscopy. The rectangular extrusion is then sectioned and recrystallized before hot rolling to final dimensions.

Table 2. Chemical analysis of molybdenum plates in weight percent.

Material	C	O	N	Ti	Zr	Fe	Ni	Si
Mo Ingot 50839	.005	<.001	<.001	-	-	.003	<.001	<.001
ASTM B386 (365)	≤.010	≤.0015	≤.002	-	-	≤.010	≤.002	≤.010
TZM Ingot 61756	.021	.0008	.0009	.48	.104	<.001	<.001	<.001
ASTM B386 (363)	.01-.03	≤.0030	≤.002	.40-.55	.06-.12	≤.010	≤.002	≤.010

The material used in this investigation was rolled to 6.4 mm (0.25 inch) thick, and subjected to either a final stress-relief or a final recrystallization anneal. The rolling direction was perpendicular to the extrusion direction. Following the final heat treatment, the surface was etched to remove the resulting decarburized zone (≈ 25μm, 1 mil). Table 3 summarizes the average transverse tensile properties measured on each plate. The microstructures of the various material heat treatment combinations are shown in Figure 1. The tensile

Table 3. Mean transverse tensile properties of molybdenum plates at 25°C.

Material	Structure	No. of Tests	σ_y, MPa (ksi)	σ_u, MPa (ksi)	Elongation $\Delta l/l_0$, %
Mo Ingot 50839	SR	2	712(103)	778 (113)	21.0
Mo Ingot 50839	RX	3	377 (55)	498 (72)	52.8
TZM Ingot 61756	SR	2	846(123)	965 (140)	15.3
TZM Ingot 61756	RX	1	476 (69)	623 (90)	50.9

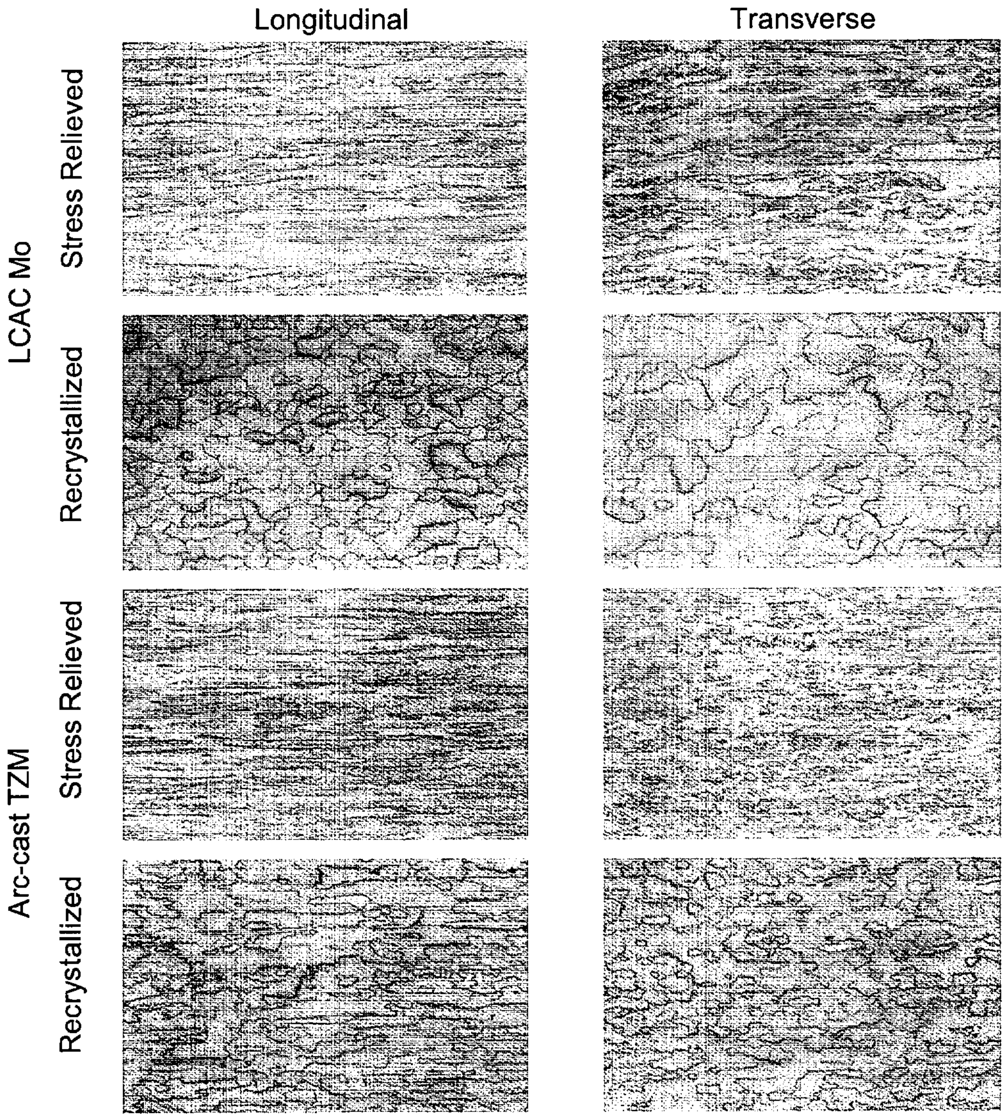

Figure 1. Representative Optical Photomicrographs of LCAC and arc-cast TZM molybdenum; Murakami's etch. ├──┤ 150μm

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properties and microstructures are typical of the two materials in the conditions obtained. Additionally, the mechanical properties exceed ASTM B386 minimum requirements for yield strength and ductility.

3. Experimental Procedures:

Sample Machining: Square compact tension (CT) specimens were machined from the 6.4 mm (0.25 inch) thick plate of LCAC molybdenum and arc-cast TZM molybdenum using EDM to provide both longitudinal and transverse notch orientations. The specimen design is shown in Figure 2. The notch width made using a 127 μm (5 mil) diameter EDM wire was consistently between 177 and 254 μm (7 and 10 mils) in width and 10 mm (0.400 inches) past the load line of the sample. The surface finish was according to ASTM E-399.

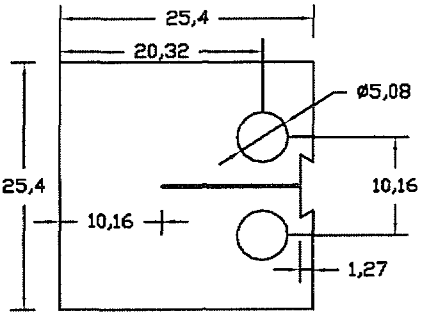

Figure 2. Sketch of 6.4 mm (0.25 inch) thick compact tension specimen design (dimensions are shown in mm).

Cyclic Pre-cracking: A cyclic compressive load was used for initiating the pre-crack (3). Benefits of this method include pre-crack stability, symmetry and low risk of fracture for brittle specimens. Specimens were cycled 2500 times and visually inspected before the maximum load was increased. A typical loading sequence was 2500 cycles (20 to 40Hz, R=0.1) at 455 kg (1000 lbs) maximum load, followed by 2500 cycles at 909 kg (2000 lbs) maximum load etc. Cycling was continued in this manner until a crack was observed from some point at the notch root. If a crack was visible and growing, no further increase in load was applied until crack advance stopped. Using this method, specimens were pre-cracked at maximum loads less than 4545 kg (10,000 lbs) and total cycle count below 40,000.

<u>Test Method:</u> Room temperature toughness testing followed ASTM E-399 (7) guidelines wherever possible. Even though the pre-cracking method described above does not meet E-399 requirements, the testing yielded meaningful results by following the other guidelines as highlighted below:

- Loading rate 0.55 to 2.75 MPa $m^{1/2}$/sec (30 to 150 ksi $in^{1/2}$/min).
- Pre-crack length must be a minimum of 1.2 mm (50 mils) past the notch root.
- Pre-crack must be within 10° of the notch plane.
- Surface crack lengths between 85% and 115% of the internal average.
- Maximum load at failure no more than 110% of that used for K_{IC} calculation.

From a sample thickness standpoint, E-399 requires the specimen thickness to be greater than 2.5 times $(K/\sigma_y)^2$. Only one of the room temperature specimens, as noted in the results, showed enough plasticity that the required specimen thickness should have exceeded the 6.4mm (0.25 inch) plate thickness. As temperature and toughness increase, and yield strength decreases, it would not be unusual for certain materials to require very thick (75 mm / 3 inch) samples for valid K tests using E-399. Because this is impractical for most cases of testing, other toughness test methods have been developed. The ASTM E-1820 (7) procedure applies to materials tested under these conditions. A quantity known as J is defined based on crack length, sample geometry and plastic energy absorbed during testing. Like the quantity K described above, J is also a measure of the energy required to create an additional unit of crack advance.

The J test induces either unstable crack extension or stable crack extension (tearing) in the sample. Instability results in a single-point value of toughness, while stable tearing results in a continuous fracture toughness versus crack-extension relationship (R-curve) from which significant point values may be determined. This method requires continuous measurement of load versus load-line displacement crack mouth opening as tearing occurs. In the work presented here the load line opening is not measured; rather crack opening is measured on the edge of the sample as in ASTM E-399. The J method can be

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used to determine toughness with a single sample if high-resolution crack opening measurements can be made using a compliance unloading technique to determine in-situ crack lengths. An example of the load versus crack opening displacement measurements and derived R curve that were used to calculate the J values is shown in Figure 3. The values of K_{IC} are then calculated from the measured J values for comparison to the room temperature values. A full description of these test methods and surrounding calculations is determined in ASTM E-1820 (7). Although not all ASTM validity requirements were strictly met, the values obtained in this investigation are a good first approximation of the fracture toughness of molybdenum alloys.

4. Results and Discussion:

The room temperature and 300°C fracture toughness test results are shown in Table 4. As can be seen from the listed data, toughness measurements of the duplicate specimens showed good agreement. At both test temperatures, the orientation of the crack did not appear to have a significant effect on the toughness. This is most likely due to the similarity between the longitudinal and transverse microstructures in each heat treatment condition, Figure 1. The similarity in microstructure is developed by the recrystallization of the billet following extrusion and then further working the billet (rolling) perpendicular to the extrusion direction.

At room temperature, regardless of alloy, microstructure, and crack orientation, the fracture toughness values were all between 15 and 22 MPa $m^{1/2}$ (14 and 20 ksi $in^{1/2}$) with the LCAC molybdenum slightly higher (18 - 22 MPa $m^{1/2}$ / 16 - 20 ksi $in^{1/2}$) than the arc-cast TZM molybdenum (15 - 20 MPa $m^{1/2}$ / 14 - 18 ksi $in^{1/2}$). These fracture toughness values compare quite favorably to the literature values shown in Table 1.

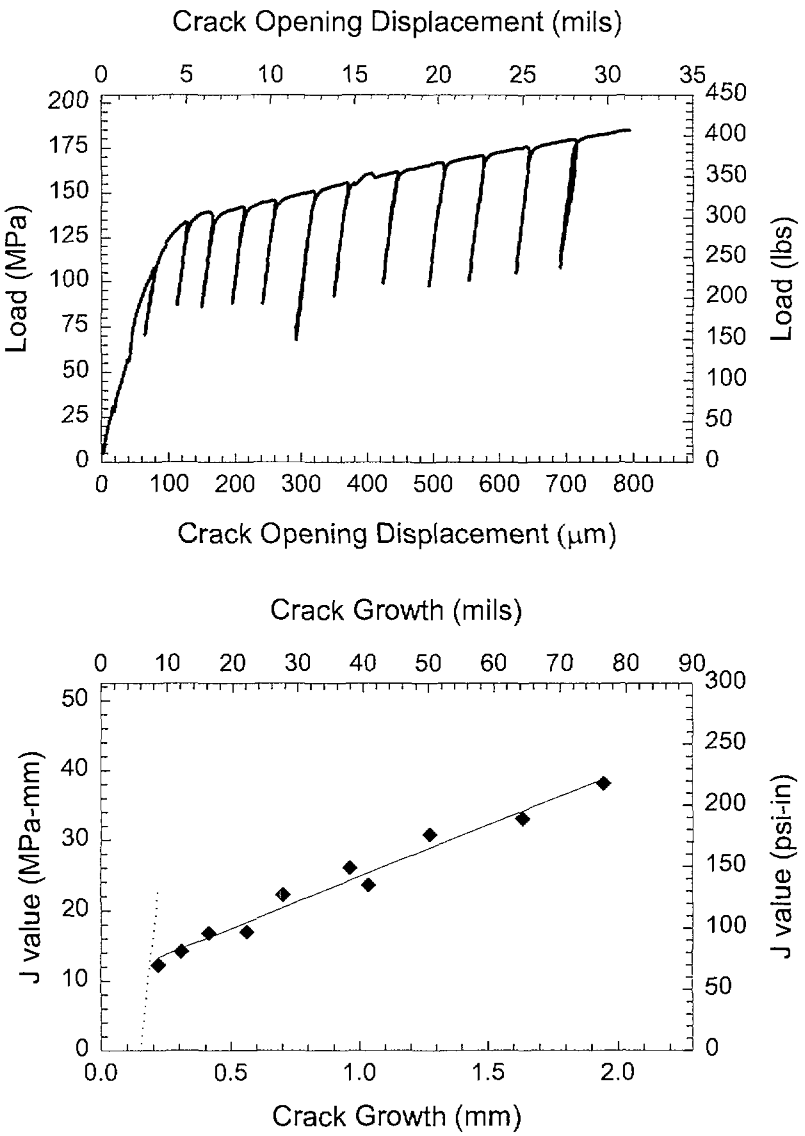

Figure 3. Typical 300°C unloading and R curve used to calculate the J - integral of 0.25 inch thick LCAC molybdenum and arc-cast TZM plate.

Table 4. Fracture toughness of 6.4 mm (0.25 inch) thick LCAC molybdenum and arc-cast TZM plate.

Material	**Test Temperature**	**Microstructure**	**Orientation**	**Toughness** MPa $m^{\frac{1}{2}}$	ksi $in^{\frac{1}{2}}$
LCAC Mo	20°C (68°F)	Stress-relieved	Longitudinal	20, 20	18, 18
			Transverse	21, 22	19, 20
		Recrystallized	Longitudinal	18, 21*	16, 19
			Transverse	19, 19	17, 17
	300°C (572°F)	Stress-relieved	Longitudinal	73, 76	66, 69
			Transverse	69, 77	63, 70
		Recrystallized	Longitudinal	59, 60	54, 55
			Transverse	66, 68	60, 62
Arc-cast TZM	20°C (68°F)	Stress-relieved	Longitudinal	19, 20	17, 18
			Transverse	15, 18	14, 16
		Recrystallized	Longitudinal	15, 18	14, 16
			Transverse	18, 19	16, 17
	300°C (572°F)	Stress-relieved	Longitudinal	84, 99	76, 90
			Transverse	88, 93	80, 85
		Recrystallized	Longitudinal	58, 64	53, 58
			Transverse	67, 67	61, 61

* Did not meet the ASTM E-399 thickness requirement.

At 300°C, the fracture toughness of the recrystallized material was similar for both the LCAC and arc-cast TZM molybdenum ranging from 58 to 68 MPa $m^{\frac{1}{2}}$ (53 to 62 ksi $in^{\frac{1}{2}}$). The toughness of the stress relieved materials, however, was higher than the recrystallized material for both alloys with the arc-cast TZM showing the higher overall toughness. The stress relieved LCAC molybdenum toughness was approximately 74 MPa $m^{\frac{1}{2}}$ (67 ksi $in^{\frac{1}{2}}$) while the arc-cast TZM molybdenum toughness was approximately 91 MPa $m^{\frac{1}{2}}$ (83 ksi $in^{\frac{1}{2}}$).

5. Conclusions:

For molybdenum and molybdenum alloys to be considered for applications that must exhibit fracture resistance, it is necessary to characterize the materials with respect to toughness. The temperature range between ambient and 1200°C forms the region of most interest with respect to toughness since it is in this region that components such as forging dies, metalworking tooling, and furnace

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structural parts must operate. In this investigation, the fracture toughness of low carbon arc-cast (LCAC) molybdenum and arc-cast molybdenum-TZM alloy plate were measured at room temperature and 300°C using compact tension specimens that were fatigue pre-cracked.

At room temperature, regardless of alloy, orientation, or microstructure, fracture toughness values between 15 and 22 MPa $m^{1/2}$ (14 and 20 ksi $in^{1/2}$) were measured. These K_{IC} values were consistent with measurements by other authors testing bar stock and employing radial crack orientations. At 300°C, the fracture toughness of recrystallized LCAC and arc-cast TZM molybdenum were also similar to one another at approximately 64 MPa $m^{1/2}$ (58 ksi $in^{1/2}$). In the stress-relieved condition, however, the arc-cast TZM toughness of (91 MPa $m^{1/2}$ / 83 ksi $in^{1/2}$) was higher than the LCAC molybdenum (74 MPa $m^{1/2}$ / 67 ksi $in^{1/2}$). While it is often thought that molybdenum is an inherently brittle material, the results obtained here and by other investigators have shown that commercially available molybdenum has sufficient toughness at room temperature and at elevated temperatures to be a useful material for load-bearing structural applications.

6. Acknowledgements:

The authors would like to acknowledge the efforts of D. Priebe of CSM Industries for tensile testing and N. Ferri and T. Delahanty of Pittsburgh Materials Technology, Inc. for fracture toughness testing and metallographic analysis.

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