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Author(s)	Matsuda, Fukuhisa; Ushio, Masao; Nakata, Kazuhiro
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Weldability of Molybdenum and Its Alloy Sheet (Report II) † —Ductility of EB and GTA Welds—

Fukuhisa MATSUDA*, Masao USHIO** and Kazuhiro NAKATA***

Abstract

The ductility of the welds in electron-beam welding in high vacuum and GTA welding in pure and air mixed argon atmospheres has been examined for electron-beam melted pure molybdenum (EB-Mo) and TZM alloy by using bending test. The main conclusions were as follows; (1) GTA and EB welds of EB-Mo became much brittle in comparison with base metal. Ductile-to-brittle transition temperature on transverse bend was 60°C for EB welds and 180°C for GTA welds in as-welded condition in comparison with that of base metal of -75°C. (2) The transition temperature of EB-Mo welds was much lower on longitudinal bend than that on transverse bend by 100 - 150°C for GTA and EB welds. (3) Low welding heat input was effective to weld bend ductility. Therefore, EB welds is generally superior to GTA welds. However, extreme low welding heat input gave a detrimental effect to weld bend ductility in EB welds. There is, therefore, an optimum condition for welding heat input in EB welds in order to obtain the welds having the best ductility. (4) Air gas mixed into argon welding atmosphere remarkably increased the transition temperature of GTA welds. Therefore, purity of argon welding atmosphere should be kept as pure as possible. (5) Post heat treatment of stress relieving was beneficial to weld bend ductility of the welds. (6) Rough surface of weld bead was detrimental to bend ductility of the welds.

KEY WORDS: (Molybdenum) (Weldability) (Ductility) (Bend Test) (Brittle Fracture) (Electron Beam Welding) (GTA Welding)

1. Introduction

The research program of this title has been conducting to make clear the weldability of electron-beam melted high pure molybdenum metal (EB-Mo) using electron-beam welding (EBW) in high vacuum and GTA welding in pure or impure argon atmospheres. In the previous report¹⁾, the effects of purity of welding atmosphere on weld defects, such as hot cracking and porosities, and mechanical properties were made clear in comparison with conventional arc-melted (AM) and powder metallurgy - (PM) TZM alloys.

On the other hand, besides weld defects in the above, the low ductility of weld metal and heat affected zone (HAZ) near room temperature is also one of the serious problems for the welding of molybdenum metal. Therefore, in this report the ductility of EB-Mo welds has been investigated by using a simple bend test, and effects of various factors which seem to be related to bend ductility of the welds, such as purity of welding atmosphere, welding heat input, surface condition of weld bead and heat treatment after welding have been examined in comparison with TZM alloys.

2. Materials Used and Experimental Procedures

2.1 Materials used

Same materials as used in the previous report¹⁾ have been used whose chemical compositions are shown in Table 1. EB-Mo which is the most pure molybdenum

Table 1 Chemical compositions of materials used

Material	Chemical composition (ppm)								
	O	N	C	Si	Ni	Fe	Ti	Zr	Mo
EB-Mo(40C)	8	12	40	88	1	2	-	-	Bal.
AM-TZM	3	<1	170	<10	<10	10	4000	910	Bal.
PM-TZM	200	8	380	-	-	-	5000	700	Bal.

Sheet thickness: 1.5 mm

obtained in commercial levels has been mainly used. The thicknesses of those materials are 1.5 mm and are annealed for stress relieving before welding.

2.2 Experimental procedures

2.2.1 Welding method

EB welding in high vacuum and GTA welding in a welding chamber initially evacuated to 1×10^{-3} torr and filled to 760 torr with high purity argon gas (99.99% Ar) or with impure argon gas mixed with air have been per-

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† Received on April 4, 1980
* Professor
** Associate Professor
*** Research Associate

Table 2 Welding conditions used

Welding method	Welding condition	Welding atmosphere
EBW	55mA, 50kV, 2540mm/min 45mA, 50kV, 1000mm/min 80mA, 40kV, 1000mm/min	1×10^{-4} Torr
GTAW (dcsp)	150A, 11V, 100mm/min 170A, 11V, 100mm/min	Ar (99.99%) or Ar+Air (Max. 1.05%) } 1 atm.

formed by bead-on-plate welding with welding conditions to produce full penetration, which are shown in Table 2. Weld specimens were pickled by chromic acid before welding in order to insure a clean surface for welding. The details of welding methods were shown in the previous reports¹⁾.

2.2.2 Test procedure

A simple bend test was used to determine the ductility of welds. A bend test equipment is shown in Fig. 1. Bending stress was applied usually to transverse direction, but partly to longitudinal one to welding direction as shown in Fig. 1. All welds were tested with the top surface of welds in tension during bending. The load was applied at the center of the specimen at a deflection rate of 1 mm/min. Specimens were bent until fracture occurred or to a maximum angle of 120 degree (limit of test equipment) in temperature range from -196 to 350°C . Specimen sizes and testing conditions are listed in Table 3. Data from bend tests are represented in mm of deflection. The deflection of specimen has been correlated with the angle of bending using a relation between bend deflection and bend angle as shown in Fig. 2. The temperature of the ductile-to-brittle transition (T_r) of specimen was designated as the lowest temperature over which complete bending of 120 degree was accomplished without fracture. The welds for the bending test specimens were mostly in as-welded condition, but some of them were ground for their faces by emery paper (# 800) or annealed for stress-relieving after welding.

Metallographic and fractographic examinations were also made in order to evaluate the ductility of welds.

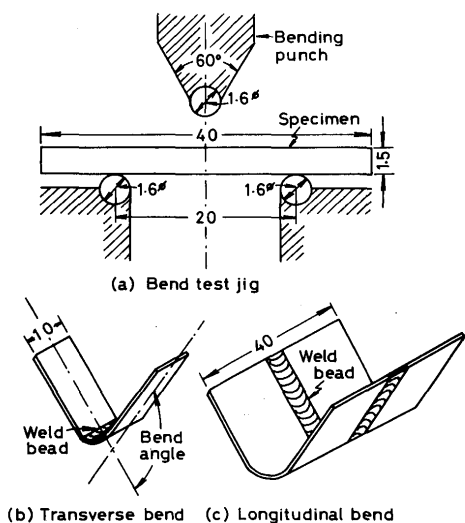


Fig. 1 Nomenclature and dimensions for bend test jig

Table 3 Specimen sizes and testing conditions for bend test

Specimen	: $10^w \times 40^l \times 1.5\text{mm}$ (Trans.)
	: $40^w \times 40^l \times 1.5\text{mm}$ (Longi.)
Test span	: 20 mm
Punch radius	: 0.8 mm
Deflection rate	: 1 mm/min
Testing temperature	: -196 to 350°C
Bend ductility-to-brittle transition temperature, T_r	: Lowest temperature for 120 degree bend without cracking

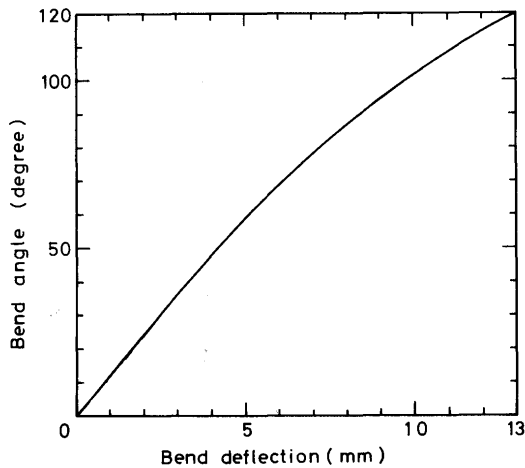


Fig. 2 Relation between bend deflection and bend angle

3. Results and Discussions

3.1 Ductility of welds

3.1.1 Comparison between transverse and longitudinal bending ductilities of welds.

Transverse and longitudinal bending tests have been carried out for EB-Mo welds specimen of EB welding and GTA welding in argon atmosphere. The relation between bend angle and testing temperature are shown in Fig. 3. In both cases, the welds became much brittle in comparison with the base metal whose T_r was -75°C . At transverse bend, the T_r was much higher than those at longitudinal bend by $100 - 150^\circ\text{C}$ for each specimen.

As mentioned in the previous report¹⁾, in fusion zone,

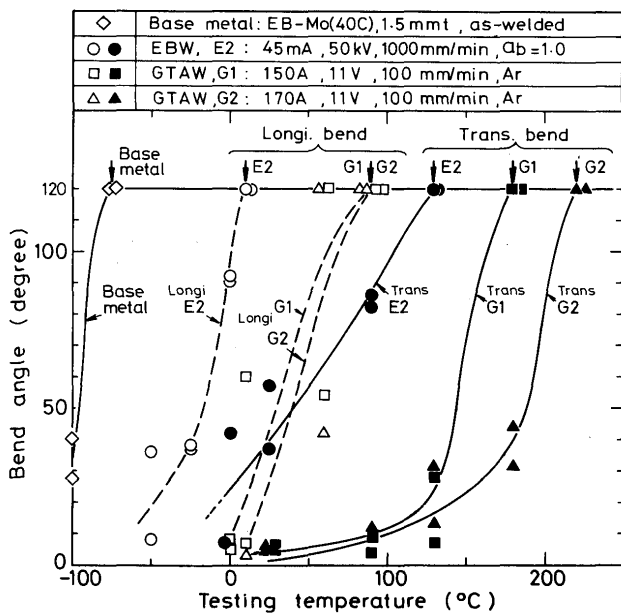


Fig. 3 Comparison of bend transition temperature of longitudinal bend specimen with that of transverse bend specimen of EB and GTA welds (material: EB-Mo (40C))

the coarse columnar crystals preferentially grow to almost same direction of the welding one in weld central zone. In this grain orientation, the bend stresses are applied vertically to these elongated grain boundaries at transverse bend. This is one of the main causes that the T_r at transverse bend was higher than that at longitudinal bend test.

As a result, ductile-to-brittle transition behaviours have been evaluated by the transverse bend test in this study hereafter because it is severer than longitudinal bend test.

3.1.2 Effect of welding heat input

Ductility of molybdenum metal is much sensitive to the grain size. The coarsening of grains in weld metal is one of the causes of the lack of ductility in fusion welds. Decreasing welding heat input is one of the most effective methods to prevent the coarsening of grains in weld metal as shown in previous report¹⁾.

The relation between bend angle and testing temperature are shown in Fig. 4 for EB-Mo in EB welding and GTA welding in argon atmosphere. The welding heat

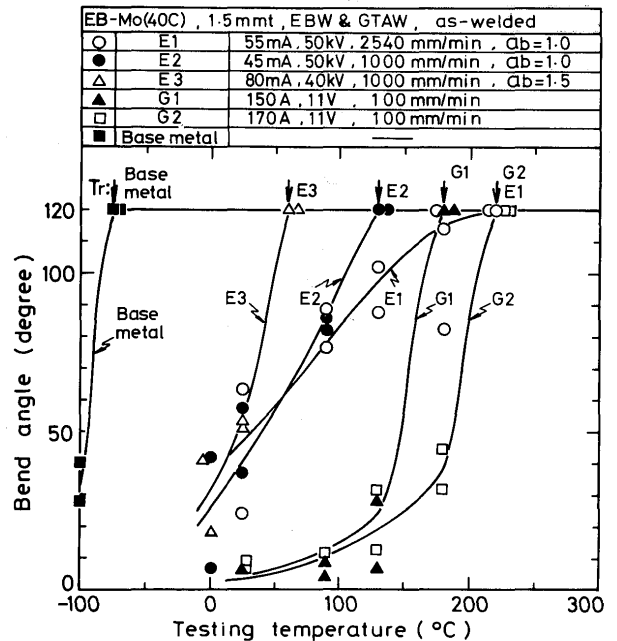


Fig. 4 Effect of welding heat input on transition temperature of transverse bend test for EB and GTA welds (material: EB-Mo (40C))

input was varied for each welding method in the range where full-penetrated weld bead could be obtained. In GTA welds, it is appeared that decreasing the welding heat input considerably lowered the T_r . However, there is a limit to decrease the welding heat input in GTA welding because of insufficient weld penetration. Therefore, the lowest T_r of EB-Mo GTA welds is about 180°C in as-

welded condition.

Very low welding heat input has been obtained in EB welding. The lowest T_r is about 60°C , which is much lowered in comparison with that of GTA welds. Decreasing welding heat input, however, the T_r increased and at the smallest condition, it again became to the same value as in GTA welds. It seems that this is mainly due to the hardness distribution of EB welds. As shown in report I¹⁾, the weld metal and HAZ were considerably softened in comparison with base metal. When this softened range become very narrow, the deformation is concentrated there due to restriction by the hard base metal surrounded. Therefore, the T_r increased as the decrease of welding heat input in EB welds. On the contrary, in GTA welds the softened range is considerably widened.

Consequently, from a viewpoint of improvement of weld bend ductility, the low welding heat input is usually effective. Therefore, the weld bend ductility of EB welds is generally superior to that of GTA welds, but in EB welds, extreme low welding heat input is rather detrimental to the weld bend ductility.

3.1.3 Effect of purity of welding atmosphere in GTA welding

Oxygen and nitrogen in weld metal of molybdenum are very harmful to the weld bend ductility^{2), 3)}. In actual GTA welding process for the industrial purposes, it is usually performed under an open air condition. In this condition, welds are considered to be always contaminated to some degree by air gas penetrated into welding atmosphere from surrounding air.

Figures 5 and 6 show the relations between the bend angle and the testing temperature in GTA welds in pure and impure argon atmospheres for EB-Mo and PM-TZM, respectively. For both materials, air gas added in welding atmosphere increased the T_r . These relationships are

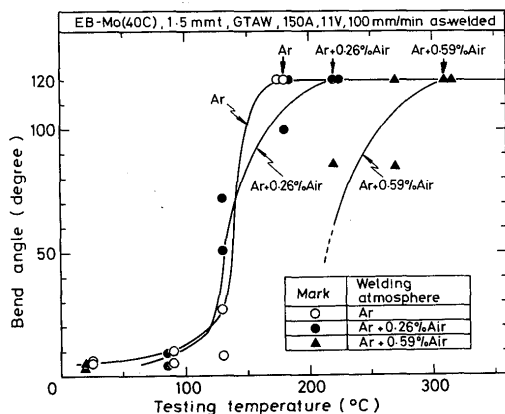


Fig. 5 Influence of air content in welding atmosphere of GTAW on the relations between bend angle and testing temperature for EB-Mo (40C)

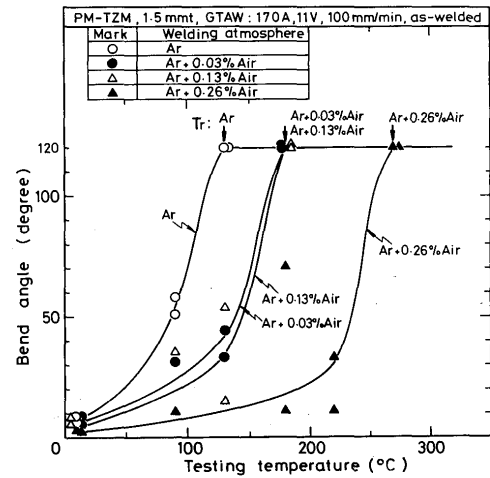


Fig. 6 Influence of air content in welding atmosphere of GTAW on the relations between bend angle and testing temperature for PM-TZM

collectively shown in Fig. 7. In the case of EB-Mo, addition of 0.26% air increased the T_r from 180°C to 220°C . Addition of 0.59% air increased to 310°C . The weld center-bead hot cracking occurred in more air added atmosphere as shown in report I¹⁾. For PM-TZM, addition of only 0.03 – 0.13% air increased the T_r from 130°C to 180°C and at 0.26% air, it was considerably

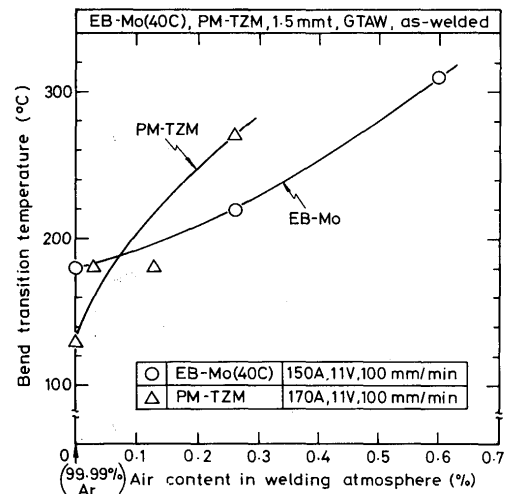


Fig. 7 Effect of air content in welding atmosphere of GTAW on transverse-bend transition temperature for EB-Mo and PM-TZM

increased to 270°C . Next, Figure 8 shows the coordinated effect of oxygen and nitrogen contents in weld metal measured¹⁾ to the T_r . As the increase of total contents of oxygen and nitrogen, the T_r also increased for each material. In spite of small amounts of oxygen and nitrogen in weld metal of EB-Mo, the T_r of it is higher than that of PM-TZM. According to this result, it is considered that as to weld bend ductility, EB-Mo welds

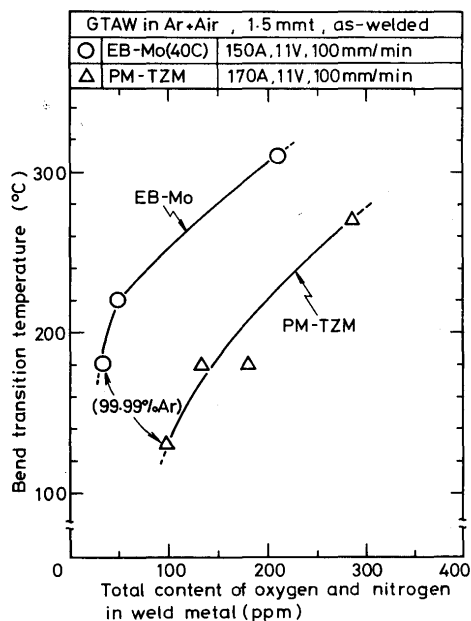


Fig. 8 Effect of total contents of oxygen and nitrogen in weld metal of GTAW on transverse-bend transition temperature for EB-Mo and PM-TZM

are much sensitive to oxygen and nitrogen in weld metal than PM-TZM welds.

As a result, in GTA welding the shielding gas in welding should be kept as pure as possible by preventing the intrusion of air gas into it in order to obtain much ductile welds.

3.1.4 Effect of postheat treatment

Figure 9 shows the relations between the bend angle and the testing temperature with and without postheating after welding for stress relief at 920°C for 1 hour in vacuum. For GTA welds, the Tr was lowered from 180°C

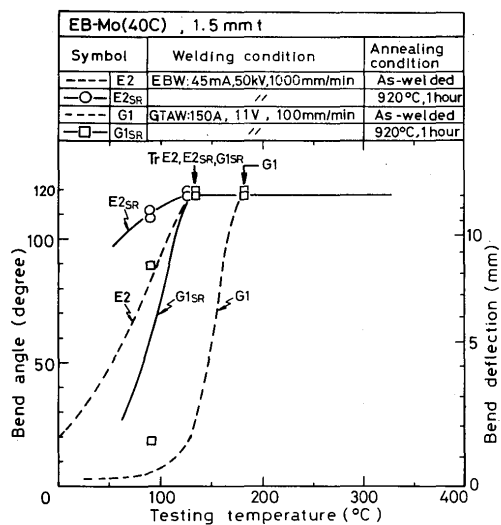


Fig. 9 Effect of postheating of EB and GTA welds on transverse-bend transition temperature (material: EB-Mo (40C))

to 130°C by stress relieving heat treatment. The Tr of EB welds was not changed, but in comparison with the bend angle at the same testing temperature they were increased by the stress relieving.

Therefore, it is considered that postheating for stress relief at 920°C for 1 hour in vacuum is effective to improve the weld bend ductility.

3.1.5 Influence of surface condition on weld bead

The surface of weld bead is fairly rough in as-welded condition, especially for EB welds because of reinforcement and surface ripple of weld bead. This surface condition affects on the weld bend ductility as shown in Fig. 10. In the solid line the weld bead surface treatment

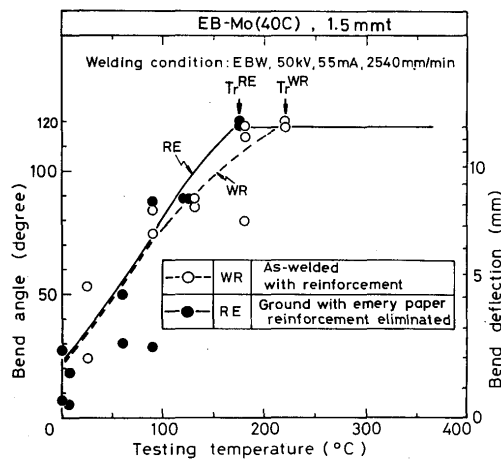


Fig. 10 Influence of surface condition of weld bead on transverse-bend transition temperature for EB welds (material: EB-Mo (40C))

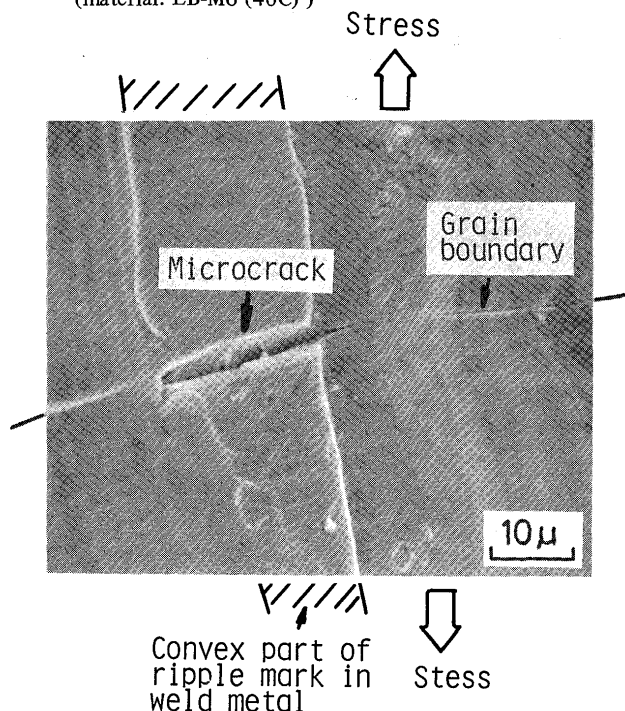


Fig. 11 Microcracking observed on surface of weld bead after bend test at 90°C (material: EB-Mo (40C), EBW(E2))

lowered the T_r of EB welds of EB-Mo from 220°C to 180°C. Figure 11 shows a typical example in SEM photograph that rough surface acts as the crack initiation. Microcrack was usually initiated at grain boundary on the convex part of the ripple mark on weld bead surface when the grain boundary was vertically stressed.

The beneficial effect of surface smoothing of weld bead has been also reported⁴⁾. Therefore, it is better for weld bend ductility to perform the treatment of surface smoothing of welds.

3.1.6 Comparison between EB-Mo and TZM alloy welds

TZM alloys are known as one of the weldable molybdenum alloys. The relations between the bend angle and the testing temperature were compared at same welding conditions. Results are shown in Figs. 12 and 13 for GTA welds in pure argon atmosphere and EB welds, respective-

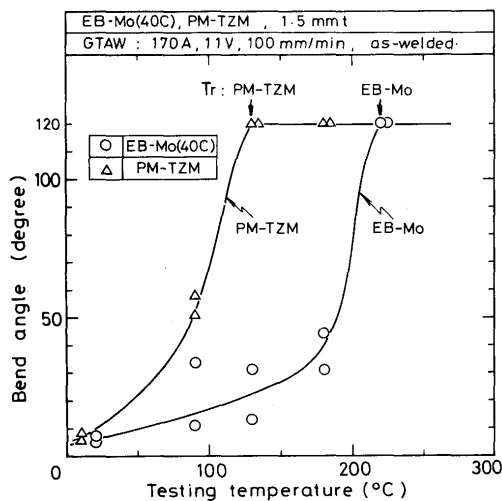


Fig. 12 Relation between bend angle and testing temperature for GTA welds on transverse-bend test in same welding condition (material: EB-Mo (40C) and PM-TZM)

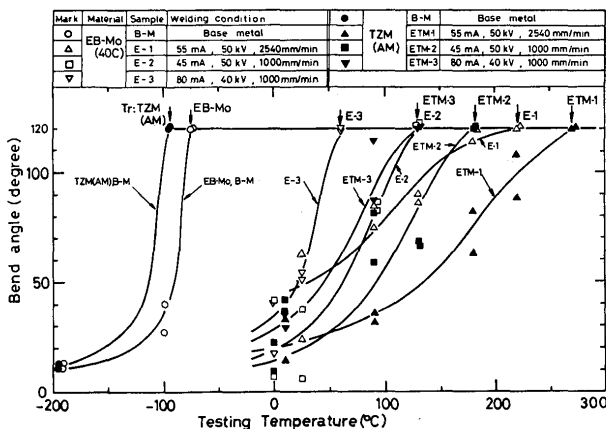


Fig. 13 Relation between bend angle and testing temperature for EB welds on transverse-bend test in various welding conditions (materials: EB-Mo (40C) and AM-TZM)

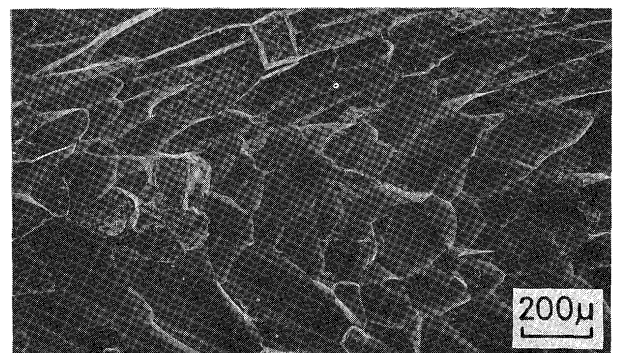
ly. In GTA welds, the T_r of PM-TZM is 130°C, which is lower than that of EB-Mo by 90°C. It seems that this is mainly due to small grain size of PM-TZM weld metal as mentioned in report I¹⁾. On the contrary, in EB welds, the T_r of AM-TZM is higher than that of EB-Mo by 50 – 70°C for each welding condition. Moreover, they became higher as the decrease of welding heat input. This is probably due to the same reason as mentioned in 3.1.2 for EB-Mo. Therefore, it seems that the narrower fusion zone and larger difference in hardness between base metal and weld metal of AM-TZM than those of EB-Mo are one of the reasons why the T_r of AM-TZM is higher than that of EB-Mo.

On the other hand, in the case of PM-TZM, the porous weld bead has been formed with EB welding as mentioned in report I¹⁾.

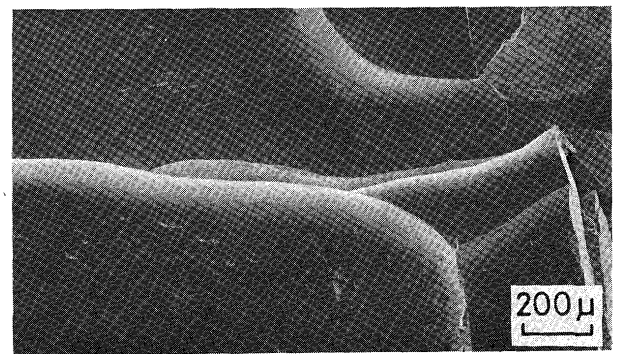
In addition, it is suggested from the results that more suitable method is needed to evaluate the ductile-to-brittle transition behaviour of molybdenum welds with very narrow fusion zone such as EB welds.

3.2 Fractography of fractured surface

At first, the fractured surfaces of GTA welds in pure argon and EB welds of EB-Mo were compared as shown in Fig. 14 (a) and (b), respectively. Fractured surfaces



(a) EBW; 45 mA, 50 kV, 1000 mm/min



(b) GTAW; 170 A, 11 V, 100 mm/min

Fig. 14 SEM photographs for fractured surface of weld metal of EB-Mo (40C) tested at 25°C

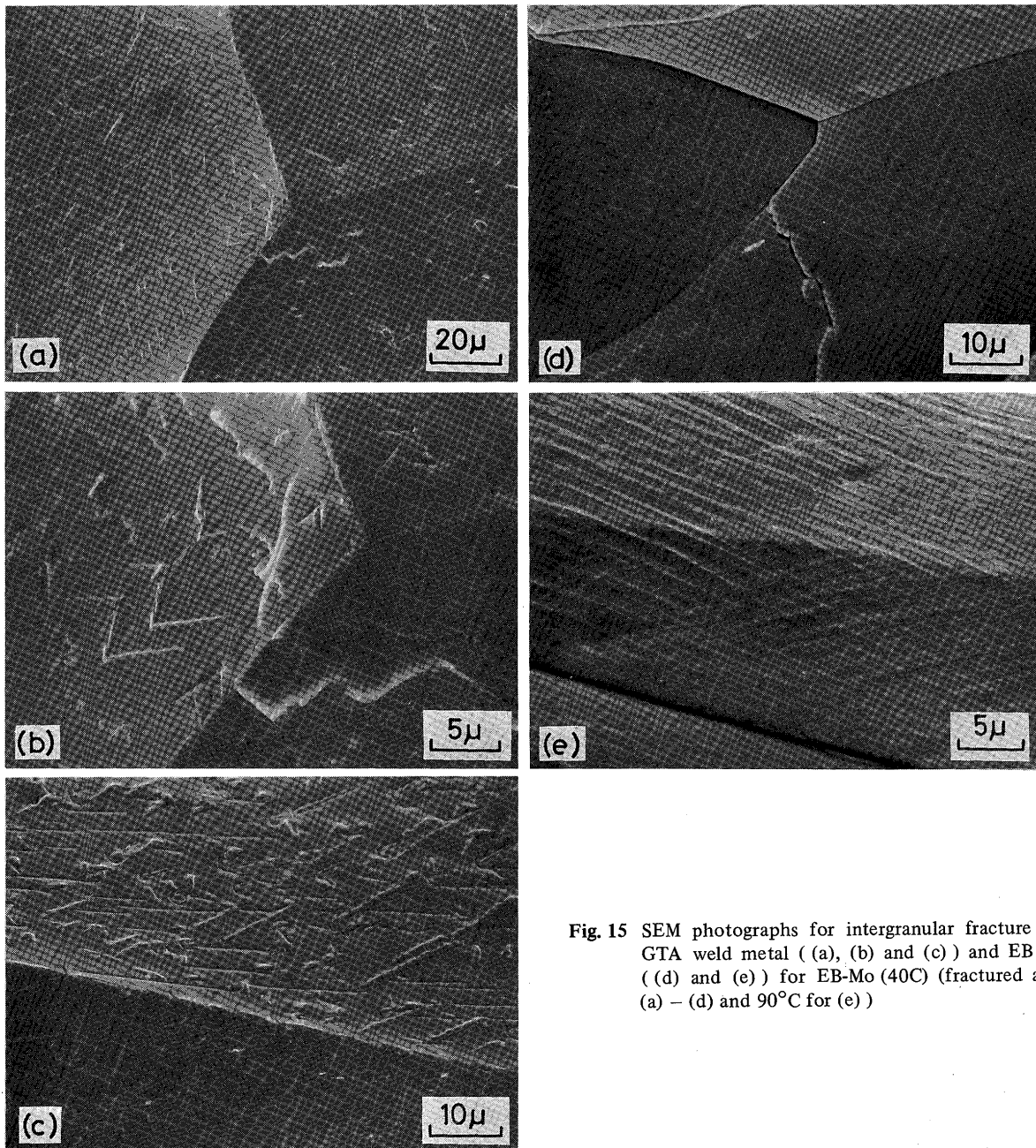


Fig. 15 SEM photographs for intergranular fracture surfaces of GTA weld metal ((a), (b) and (c)) and EB weld metal ((d) and (e)) for EB-Mo (40C) (fractured at 25°C for (a) – (d) and 90°C for (e))

almost consist of intergranular ones along columnar grains and partly cleavage fracture surfaces were also observed. The size of fracture facet in EB welds is apparently much smaller than that in GTA welds. Judging from the shape of the river pattern, it is considered that crack was initiated at grain boundary. Details in intergranular fracture surface in Fig. 14 are shown in Fig. 15. As shown in Fig. 15 (a) for GTA welds, comparably large precipitates have been usually observed, whose shape was different as shown in (b) and (c).

On the contrary, very fine precipitates were observed in EB weld metal as shown in (d) and (e). Figure 15 (e)

also shows the slip lines on intergranular fracture surface which often appeared in EB weld metal at elevated temperature near the T_r . As a result, it is considered that due to the smaller facet size and much finer precipitates on intergranular fracture surface of EB welds, weld bend ductility of EB welds is generally superior to that of GTA welds except in the case of extreme low welding heat input.

Nextly, Figure 16 shows the intergranular fracture surface of EB-Mo in GTA welds in argon with 0.59% air atmosphere. Almost all the fractured surface are covered with lamellar precipitates which are much different from

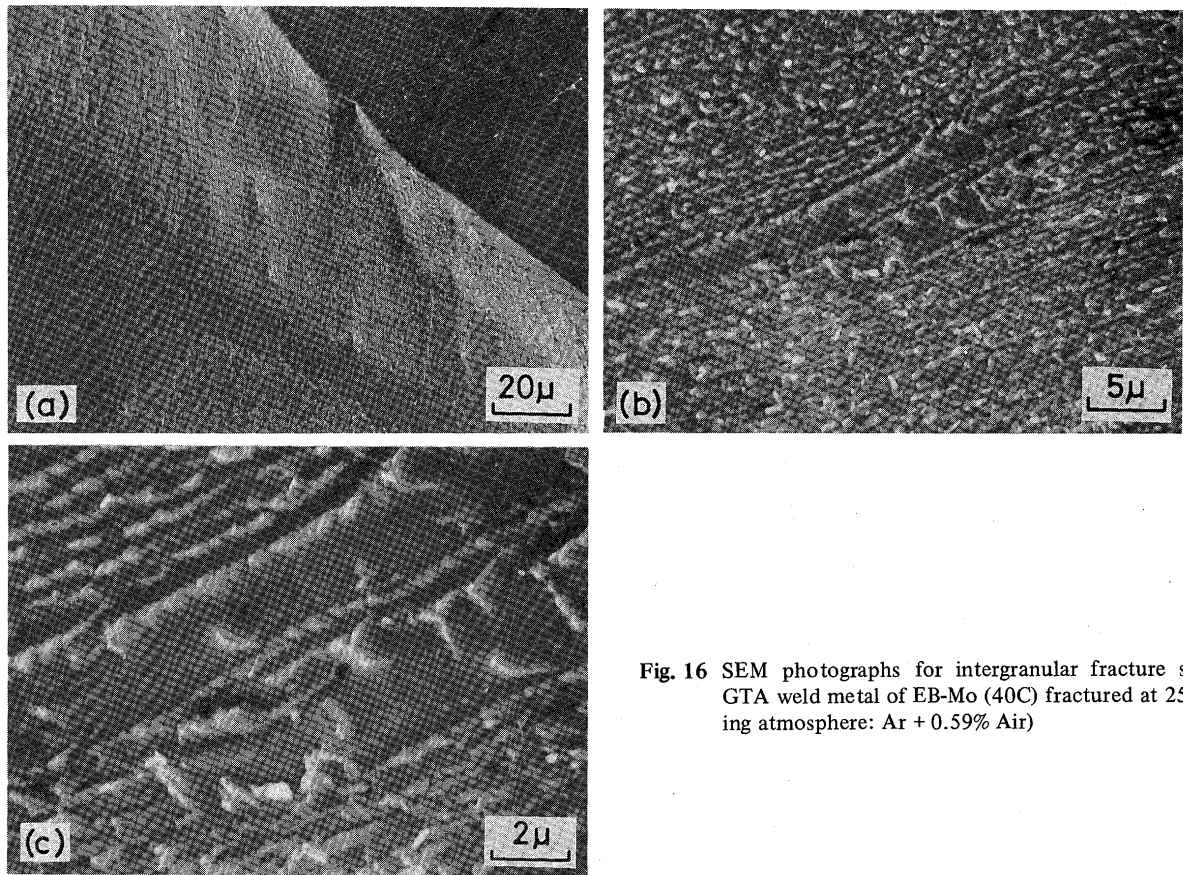
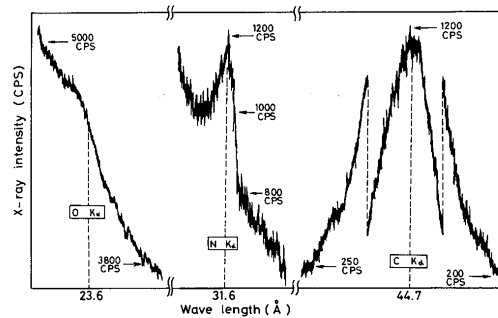
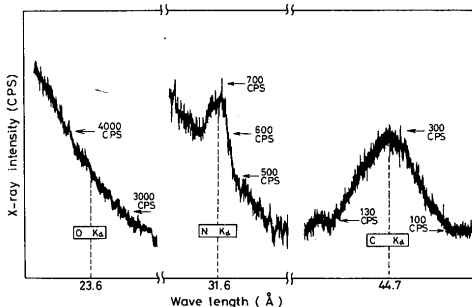


Fig. 16 SEM photographs for intergranular fracture surface of GTA weld metal of EB-Mo (40C) fractured at 25°C (welding atmosphere: Ar + 0.59% Air)



(a) Intergranular precipitates



(b) Matrix (Cleavage fracture surface)

Fig. 17 Results of X-ray analysis by wave dispersive type micro-analyzer on fractured surface of GTA weld metal of EB-Mo (40C)

those in GTA welds with pure argon atmosphere as shown in Fig. 15. Figure 17 (a) shows the results of X-ray analysis about these precipitates by the wave dispersive type X-ray microanalyzer in comparison with those on cleavage fracture surface as shown in (b). Oxygen was detected only on intergranular precipitates. Nitrogen and carbon were detected in each case, though their intensities were higher on intergranular precipitates than cleavage fracture surface. Nitrogen peak in cleavage fracture surface seems to be related to molybdenum nitride precipitated in matrix as shown in report I¹⁾. As a result, these precipitates on intergranular fracture surface seem to consist of molybdenum oxide, nitride and carbide.

Consequently, air intrusion into welding atmosphere of GTA welding made molybdenum weld metal much brittle by forming the intergranular precipitates as mentioned in the above.

The fracture surface in EB welds of AM-TZM was also observed. Typical photographs are shown in Fig. 18. As shown in (a), the area fraction of cleavage fracture surface remarkably increased in comparison with that of EB-Mo in Fig. 14, though the locations of crack initiation were still grain boundaries as shown in (b). The intergranular fracture surface is shown in (c) and (d). All over the intergranular surface was covered with lots of precipitates

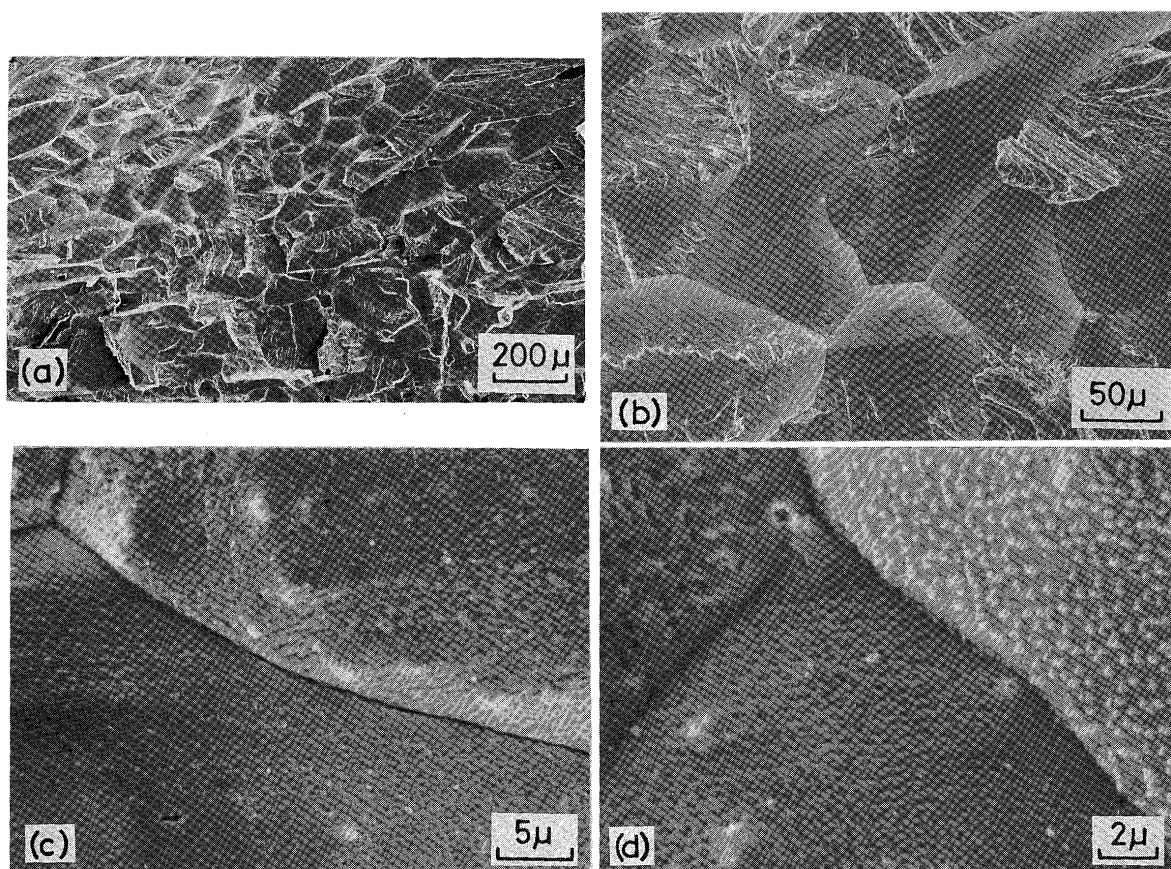


Fig. 18 SEM photographs for fractured surface of weld metal fractured at 0°C (material: AM-TZM, EBW: 80 mA, 40 kV, 1000 mm/min)

whose shapes were much different from those in EB-Mo welds.

3.3 Summary of weld bend test

The bend transition temperature of the welds of EB-Mo evaluated in this study is collectively listed in Table 4 in comparison with those of TZM alloys. The lowest transition temperature at transverse bend was higher than room temperature, that is, about 60°C, even using EB welding which was the most effective welding method in order to minimum the coarsening of grains in welds and the contamination of weld metal from welding atmosphere. These results suggest the strong demand for the development of much more weldable molybdenum metal by alloying some special elements beneficial to weld bend ductility such as carbon⁵⁻⁸, titanium^{2),4),9}, boron¹⁰ and rhenium¹¹ etc.

4. Conclusions

The weld bend ductility of electron-beam melted pure molybdenum (EB-Mo) welds has been investigated and effects of various factors which seemed to be related to

bend ductility of welds, such as purity of welding atmosphere, welding heat input, weld bead surface condition and heat treatment after welding have been examined in comparison with TZM alloys. The main conclusions obtained were as follows;

- (1) GTA and EB welds of EB-Mo became much brittle in comparison with base metal. Ductile-to-brittle transition temperature on transverse bend was 60°C for EB welds and 180°C for GTA welds even as the lowest(best) values at as-welded condition in comparison with that of base metal of -75°C.
- (2) Transition temperature of EB-Mo welds was much lower on longitudinal bend than that on transverse bend by 100 – 150°C for GTA and EB welds.
- (3) Low welding heat input was effective to lower the transition temperature of fusion welds. On the contrary, however, in EB welds the excess decrease of welding heat input increased the transition temperature. Therefore, extreme low welding heat input is detrimental to weld bend ductility for EB welds.
- (4) Air gas intruded into welding atmosphere drastically increased the transition temperature of GTA welds. 0.26% air increased it from 180°C to 220°C and at

Table 4 A list of transition temperatures in bending test of welds evaluated in this study

Material	Welding method	Welding condition	Welding atmosphere	Transverse-bend transition temperature			Longitudinal-bend transition temperature
				As-welded	As-welded & reinforcement eliminated	920°C, 1hour annealed after welding	As-welded
EB-Mo (40C)	Base metal	—	—	-75°C	—	—	—
	EBW	55mA, 50kV, 2540mm/min	1 × 10 ⁻⁴ Torr	220°C	180°C	—	—
		45mA, 50kV, 1000mm/min		130°C	—	130°C	10°C
		80mA, 40kV, 1000mm/min		60°C	—	—	—
	GTAW(Ar)	150A, 11V, 100mm/min	Ar	180°C	—	130°C	90°C
170A, 11V, 100mm/min		220°C		—	—	90°C	
GTAW (Ar+Air)	150A, 11V, 100mm/min	Ar+0.26%Air	220°C	—	—	—	
		Ar+0.59%Air	310°C	—	—	—	
PM-TZM	Base metal	—	—	—	—	—	—
	EBW	45mA, 50kV, 1000mm/min	1 × 10 ⁻⁴ Torr	270°C*	—	—	—
	GTAW (Ar)	170A, 11V, 100mm/min	Ar	130°C	—	—	—
	GTAW (Ar+Air)	170A, 11V, 100mm/min	Ar+0.03%Air	180°C	—	—	—
			Ar+0.13%Air	180°C	—	—	—
Ar+0.26%Air	270°C	—	—	—			
AM-TZM	Base metal	—	—	-96°C	—	—	—
	EBW	55mA, 50kV, 2540mm/min	1 × 10 ⁻⁴ Torr	270°C	—	—	—
		45mA, 50kV, 1000mm/min		180°C	—	—	—
		80mA, 40kV, 1000mm/min		130°C	—	—	—

* Porous weld bead

Sheet thickness: 1.5mm, Bending radius: 0.8mm, Supporting radius: 0.8mm
Distance between bending supports: 20mm, Deflection rate: 1mm/min

0.59% air it was increased to 310°C for EB-Mo. For PM-TZM, only 0.03 – 0.13% air increased it from 130°C to 180°C and at 0.26% air it was increased to 270°C. Purity of welding atmosphere should be kept as pure as possible.

- (5) Postheating of welds after welding for stress relief was beneficial to weld bend ductility.
- (6) Rough surface of weld bead was detrimental to weld bend ductility.

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