Effect of Rhenium and Niobium on Weld Metal Ductility of Molybdenum by EB Welding†

–Weldability of Molybdenum and Its Alloy Sheet (Report III) –

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Abstract

Effect of rhenium and partly niobium on the ductility of the welds of electron-beam melted pure molybdenum and arc-melted TZM alloy by electron-beam welding have been investigated by using a 3-points simple bend test. It was made clear that only rhenium was a beneficial alloying element to improve the ductility of weld metal of pure molybdenum. Bend DBTT of the pure Mo welds decreased to 20°C and –20°C for 3.3% and 13.5%Re in weld metal, respectively in comparison with 60°C of pure Mo welds. However, there was no obvious improvement of ductility of TZM welds even with much Re content of 18.5% in weld metal. Niobium showed almost no improvement of ductility of pure Mo welds in the range of 1.3% to 2.7% in weld metal.

KEY WORDS: (Weldability) (Ductility) (Molybdenum) (Filler Metal) (Rhenium) (Niobium) (Electron Beam Welding)

1. Introduction

It is well known that molybdenum (Mo) welds shows the embrittlement at room temperature due mainly to the harmful effect of small amount of oxygen and nitrogen[1–3].

Therefore, in case of welding of Mo metal, it is necessary to select the high-purity Mo metal as a base metal, of which oxygen and/or nitrogen contents are as small as possible.

At the same time, as a viewpoint of welding method, it is also required to prevent these gas contaminations in the weld metal from welding atmosphere and in addition to this, to prevent coarsening in grain size of weld metal and heat-affected zone (HAZ) due to the welding thermal cycle.

To satisfy above requirement, it is considered that the combination of electron-beam melted high-purity molybdenum (EB-Mo) and high vacuum electron-beam welding is the most desirable one as the combination of welding material and welding method, respectively in current situation.

On the basis of the above idea, in a previous report[4], the bend ductility of the welds of EB-Mo, which was the highest-purity molybdenum sheet available in commercial industrial level, by using high vacuum electron-beam welding (EBW) was investigated. Nevertheless, the bend ductile-to-brittle transition temperature (DBTT) of EB welds was over room temperature of about 60°C in as-welded condition, although it was lower than that of GTA welds with high-purity argon gas shielding.

These results suggested that it was difficult to obtain the ductile welds only by the purification of Mo metal by eliminating the harmful interstitial elements in commercial industrial level. On the other hand, alloying of some elements to Mo metal is considered to be an another possible means to lower the DBTT of Mo welds below room temperature. Among commercial Mo alloys, TZM (Mo-0.4%Ti-0.1%Zr) is the most popular Mo alloy. However, the bend DBTT of EB welds of TZM was about 130°C in as-welded condition which was higher than that of EB-Mo weld[4].

As alloying elements to improve the weld ductility of Mo metal, rhenium is considered to be the most beneficial element and there are several works[5–9] for its effect. There are, however, only a few reports about rhenium effect on the ductility of Mo welds[10–11]. So effective amount of rhenium addition to Mo welds still remains uncertain.

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Unfortunately in this moment it is very difficult to obtain enough large Mo-Re alloy plate for welding and mechanical testing after welding. Therefore in this experiment, at first, in order to overcome the brittleness of Mo weld metal, pure Re wires in two different diameters were inserted to EB-Mo weld bead as a filler metal to get the Re containing weld metal. And thereafter, effect of Re addition on the ductility of weld metal in relatively small amount of Re content in molybdenum was investigeted by means of a simple three points bending test[4].

Moreover, TZM alloy as a welding base metal and pure niobium (Nb) wire as a welding filler metal which is a strong carbide former and has complete solid solubility in molybdenum were also used for comparison.

2. Experimental Procedures

2.1 Materials used

Mainly high-purity EB-Mo with low carbon content (40 ppm) and partly arc-melted TZM alloy (AM-TZM) were used as welding base metals. Sheet thickness of these materials was 1.5 mm. Chemical compositions were shown in **Table 1**.

Table 1 Chemical compositions of EB-Mo and AM-TZM alloy

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.

As a filler metal, 99.97% pure rhenium and niobium wires of 0.25 and 0.5 mm in diameter were used.

2.2 Welding method

A square groove of 0.5 mm in width and 0.3 or 0.5 mm in depth was machined on a specimen surface and a filler wire was laid in a groove as shown in **Fig. 1**. A specimen

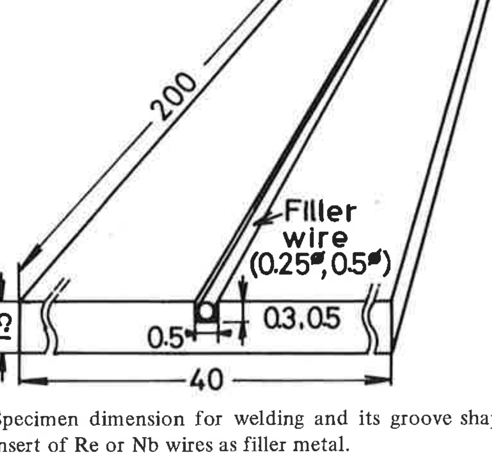


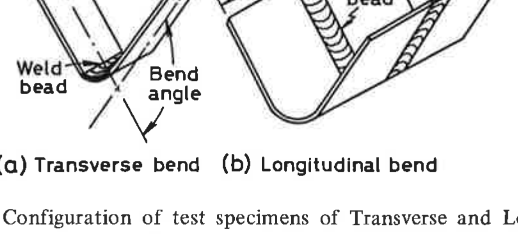
Fig. 1 Specimen dimension for welding and its groove shape for insert of Re or Nb wires as filler metal.

sheet and a filler wire were pickeled with chromic acid[4] prior to set up them for welding.

And then melt-run by electron-beam has been carried out along the groove with a filler wire under a welding condition of 40 kV in accelerating voltage, 80 mA in beam current and 900 mm/min in travel speed with 1×10^{-4} torr atmospheric pressure.

2.3 Bend test

A bend test specimen was cut off from a welded sheet transversely to a welding direction to dimensions of approximately 5 mm in width and 40 mm in length for transverse bend test and partly of 40 mm square for longitudinal bend test as shown in **Fig. 2**. Each surface of



(a) Transverse bend (b) Longitudinal bend

Fig. 2 Configuration of test specimens of Transverse and Longitudinal bend tests.

specimen was mechanically polished to 1.3 mm in thickness with #1500 emery paper. No heat treatment of test specimen for stress-relief after welding was employed.

Bend test of Mo welds was evaluated by means of the simple three points bend test[4], in which the specimen rested on two 0.8 mm dia. tungsten rollers placed 20 mm apart and was bent at a strain rate of 1 mm/min by a Instron type tensile tester having a bending punch with 0.8 mm dia. circular tip. Bending strain was applied transversely to a welding direction (transverse bend test) so that the weld bead was mainly deformed with its top surface in tension. And partly longitudinal bend test were also employed. Atmospheric temperatures during bending test was varied from −100 to 300°C.

Bend DBTT was defined as the temperature at which a full-bend of 120° bend angle was accomplished without fracture in the test specimen.

After bend test, the specimen was fractographically investigated in aid of scanning-electron microscope (SEM).

3. Experimental Results and Discussions

3.1 Effect of filler metal on macrostructure and hardness of weld metal

3.1.1 Amounts of rhenium and niobium added in weld metal

Chemical analysis results of Re and Nb in weld metal are shown in **Table 2**. These values were nearly equal to

Table 2 Result of chemical analysis of Re or Nb contents in weld metals of EB-Mo and AM-TZM

Base metal	Filler metal	Re or Nb in weld metal
EB-Mo	0.25Re	3.33 (wt%)
	0.50Re	13.46
	0.25Nb	1.27
	0.50Nb	2.71
AM-TZM	0.50Re	18.30

those estimated from the ditution of filler metal by base metal in weld bead. In case of TZM, Re content was more than that of EB-Mo weld, because the fusion zone of TZM welds was narrower than that of EB-Mo weld even with the same welding condition.

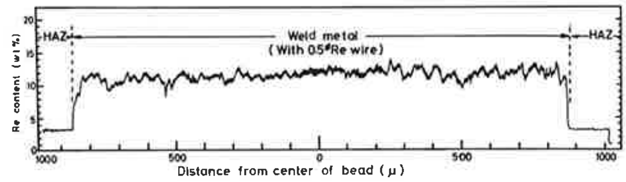


Fig. 3 EPMA results of distribution of rhenium in weld metal of EB-Mo with 0.5%Re filler.

Figure 3 shows the distribution of Re in weld metal measured with EPMA across the weld bead added with 0.5 mm dia. Re filler wire. Re content was almost uniform throughout weld bead except for the microsegregation associated with solidification, though comparably large changes in Re content became more remarkable near fusion boundaries which were caused by the ripple segregation. Similar distributions of Re or Nb were also observed in another weld beads.

3.1.2 Macrostructures

Figures 4 and **5** show the macrostructures of weld bead in horizontal section near top surface and cross-section for EB-Mo without and with Re and Nb filler metals and AM-TZM without and with Re filler metals, respectively.

There is no obvious difference in macrostructures of weld metal associated with filler additions for each base metal, though grain sizes in both weld metal and HAZ of TZM welds were considerably smaller than those of EB-Mo welds.

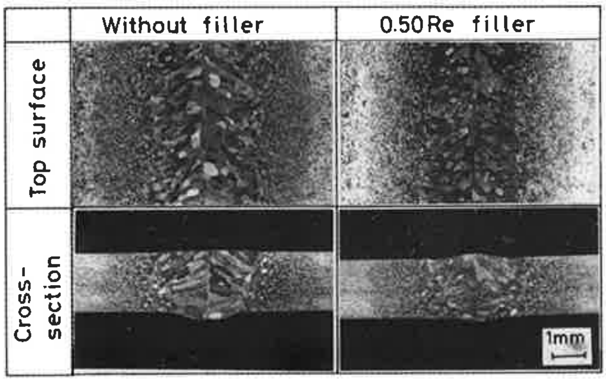


Fig. 5 Macrostructures on top surface and crosssection of AM-TZM welds without and with Re filler metal.

3.1.3 Hardness

Figure 6(a), **(b)** and **(c)** shows the hardness distributions across the base metal, HAZ and weld metal on cross-section of the welds for EB-Mo without and with Re or Nb fillers and AM-TZM without and with Re filler, respectively.

The decrease in hardness of about 20 VHN in maximum was observed in EB-Mo welds metals with addition of Re fillers, though hardness values near the center of weld metal were not so varied in comparison with those without Re filler.

Nb filler addition increased much the hardness of weld metal, though it was not observed with 0.25 mm dia. Nb filler.

In case of AM-TZM welds, Re filler addition, however, remarkably increased the hardness of weld metal up to about 280 VHN, almost equal to base-metal hardness.

The decrease in hardness observed in Re-added EB-Mo weld metals is considered to be due to the alloy softening by Re addition. According to J.R. Stephens *et al*[9], the decrease in hardness in recrystalized pure Mo was observed up to about 11 wt%Re addition, and maximum

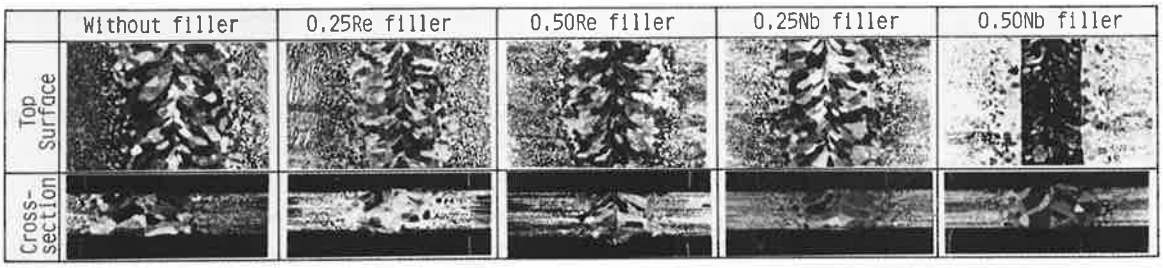


Fig. 4 Macrostructures on top surface and crosssection of EB-Mo welds without and with Re or Nb filler metals.

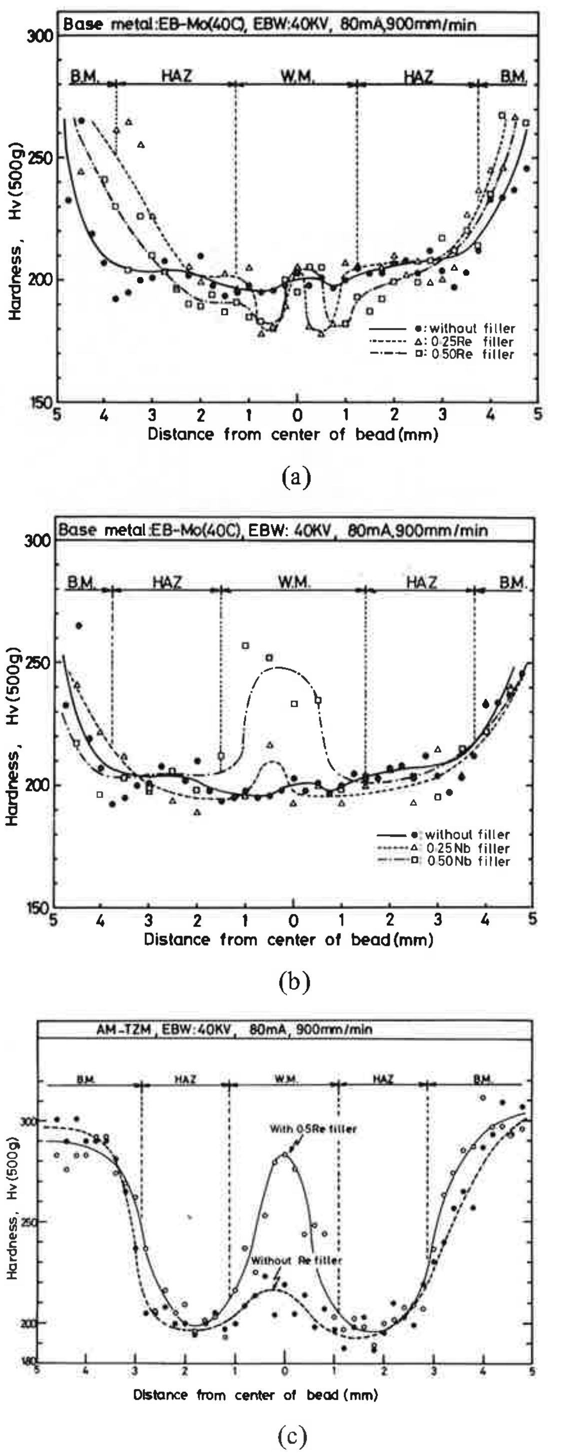


Fig. 6 Hardness distributions on crosssection of welds: (a) EB-Mo welds with Re filler, (b) EB-Mo welds with Nb filler and (c) AM-TZM welds with Re filler.

decrease of about 20 VHN from that of pure Mo appeared at about 4 wt%Re at room temperature and more addition progressively increased the hardness. Hardness increase in Re-added TZM weld metal seems to be due to the solusion hardening[9] or reaction of Re with Ti and/or Zr of alloying elements, though these were not decided in this paper.

As to Nb filler additions, M. Semchyshen reported[12] that the hardness of as-casted pure Mo with 130 ppm carbon was progressively increased with the addition of Nb by 20 and 70 VHN at 1.3 and 5.8 wt%Nb, respectively, in comparison with that of pure Mo. This seems to be almost comparable with present work inspite of the difference in each carbon content.

3.2 Bend Test Results

Effect of filler metal addition on bend ductility was shown in **Figs. 7** and **8** for EB-Mo and TZM base metals,

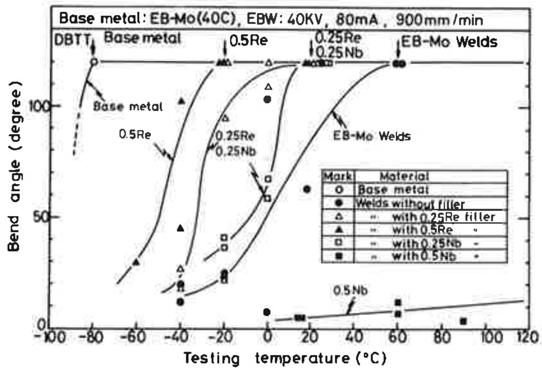


Fig. 7 Effect of Re or Nb filler metals on bend angle of EB-Mo welds for various testing temperatures evaluated by Trans-bend test.

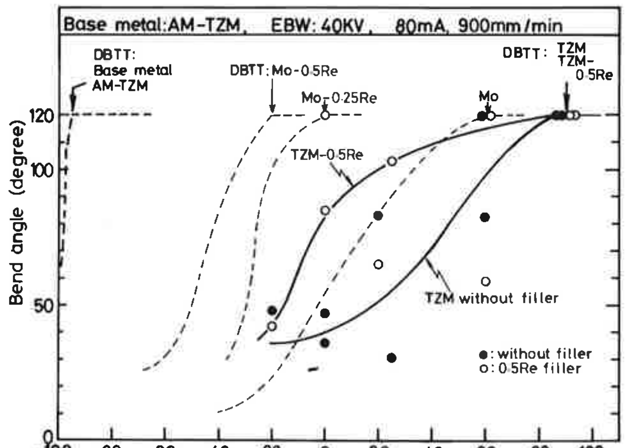


Fig. 8 Effect of Re filler metal on bend angle of AM-TZM welds for various testing temperatures evaluated by Trans-bend test

respectively, in which the relation between bend angle and test temperature was shown.

In case of EB-Mo as shown in Fig. 7, DBTT of welds without filler was about 60°C.

Re filler addition, however, improved much this DBTT and it was decreased to about 20°C at 3.3%Re and about

−20°C at 13.5%Re contents in weld metal. Moreover, these Re-contained weld metals showed comparably large bend angle even at lower temperature than their DBTT. For example, 100 degrees bend angle without fracture was accomplished in the weld bead with 3.3%Re and 13.5%Re at −20°C and −40°C, respectively.

According to Igata *et al*[10], the ductility of pure Mo weld metal by EB welding was improved by the addition of more than 5%Re, but not of 0.5%Re. Hiraoka *et al*[11] also reported the improvement of the ductility of pure Mo welds by EB welding by the addition of more than 1%Re, but not of 0.5%.

Therefore, it is considered that only 1 to 5%Re addition was required to improve the room temperature ductility of pure Mo welds.

With regards to Nb addition, DBTT was slightly decreased to 20°C at 1.3%Nb, though their bend angles were not improved below room temperature. More addition of niobium, however, showed the remarkable increase in DBTT up to 270°C at 2.7%Nb. It seems that niobium is not so beneficial element to improve the DBTT of pure Mo welds.

On the other hand, in case of TZM, as shown in Fig. 8, its DBTT was 90 degrees without filler, slightly higher than pure EB-Mo welds. There was, however, no decrease in DBTT by addition of much Re, 18.5% in weld metal and only a slight improvement in bend angle at 0°C was observed by such Re content. However, this slight improvement seems to be not due directly to Re effect but mainly to the change in fracture position from weld metal to HAZ as will be mensioned in latter.

Fracture position observed on the bend specimens was summarized on **Table 3** for EB-Mo and TZM welds with and without filler wires.

Table 3 Summary of DBTT, fracture position and fracture mode for EB-Mo and AM-TZM welds after Trans-bend test

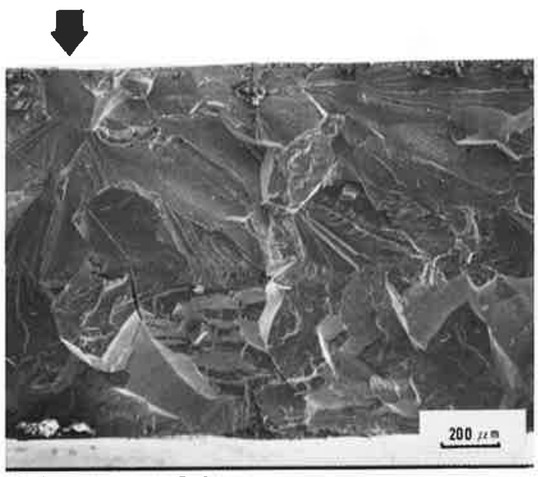
Base metal	Filler metal	DBTT(°C)	Fracture position	Fracture mode of crack initiation
EB-Mo	Non	60	HAZ(+WM)	IG*
	0.25Re	20	WM(+HAZ)	IG
	0.50Re	-20	WM(+HAZ)	IG
	0.25Nb	20	WM	IG
	0.50Nb	270	HAZ(+WM)	IG
AM-TZM	Non	90	WM	IG
	0.50Re	90	HAZ	C**

* Intergranular fracture
** Cleavage fracture

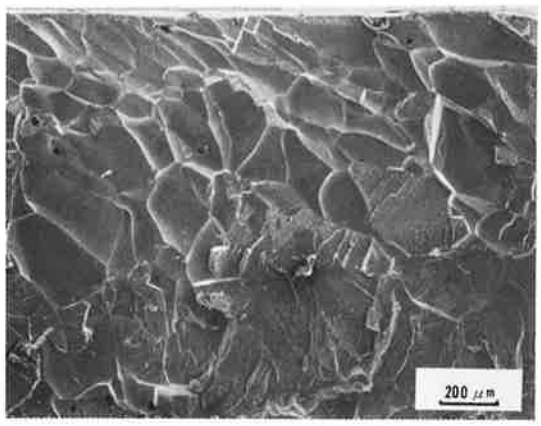
Generally, fracture position was associated with the most soft part of the welds, because it, at first, yielded and bending strain was concentrated there. For example, in case of EB-Mo welds without filler, weld metal and HAZ were much softer than base metal. So fracture was occurred in weld metal and HAZ, though fracture position was mainly HAZ and partly weld metal near fusion boundary.

As hardness of Re-contained weld metal was lower than those of HAZ and base metal, fracture was occurred in weld metal in this case. On the contrary, 2.7%Nb-contained weld metal was much harder than HAZ. In this case, fracture position was restricted mainly to HAZ near fusion boundary as same as that of Re-contained TZM welds.

Figure 9(a) and **(b)** shows the typical fracture surface



(a) without filler metal



(b) with 0.5Re filler metal

Fig. 9 SEM fractograph of EB-Mo welds with or without Re filler after Trans-bend test (brack arrow shows origin of fracture)

of EB-Mo welds without Re and with 13.5%Re, respectively. Fracture mode in Re-free EB-Mo welds in Fig. 9(a) was mainly transgranular cleavage with partly intergranular, but from river pattern, its origin was grain boundary of coarsened grains in HAZ near fusion boundary. However, in spite of improved ductility in weld metal of Re-contained welds, its fracture mode was mainly intergranular with partly cleavage, and its origin was also grain boundary of coarsened columnar structure in weld metal as shown in Fig. 9(b).

From these results, it seems that rhenium does not strengthen the grain boundary in such a small content up to about 10–15%, but it relatively lowers the strength of the grain due to alloy softening effect of rhenium and this may decrease the build-up of the dislocation in grain boundaries. In case of much Re content such as 50%, which was not investigated in this paper, it was reported that scavenge effect became dominant[10] and in addition, deformation twin became to be easy to occur[10].

Figure 10(a) and **(b)** shows typical fracture surfaces of

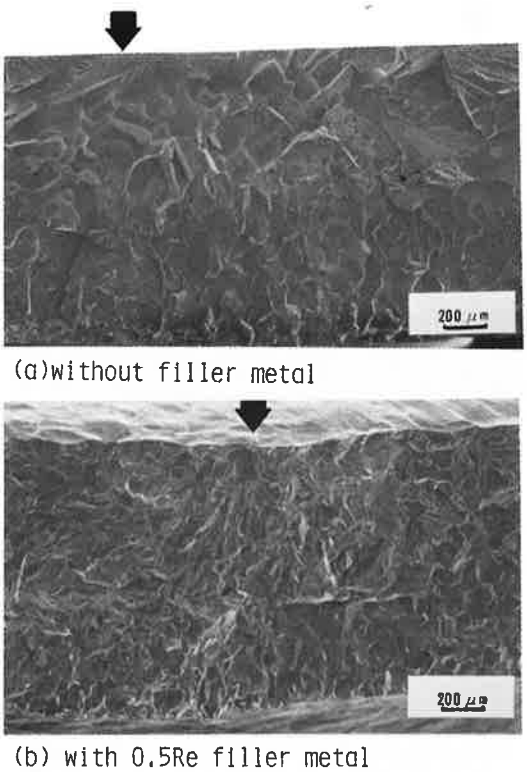


Fig. 10 SEM fractograph of AM-TZM welds without or with Re filler after Trans-bend test.

TZM welds without and with Re filler wires, respectively. As shown in Fig. 10(a), fracture position of TZM welds without filler was weld metal and its origin was intergranular of columnar structure. However, with Re filler (Fig. 10(b)), fracture position was changed to HAZ due to higher hardness of weld metal than that of HAZ (Fig. 6(c)). And its fracture origin was transgranular cleavage of very small facet in comparison with EB-Mo (Fig. 9). This is corresponding to its small grain size in HAZ in Fig. 4. Therefore, in TZM welds, it is considered that HAZ is more ductile than weld metal. So this is the reason why the TZM welds with Re filler showed a slight improvement of ductility.

In addition, in order to examine the real effect of Re addition on the ductility of the TZM weld metal, longitudinal bend test was carried out by using the test specimen as shown in Fig. 2(b). In this case, fracture origins were located in weld metal independent of Re filler and testing temperature. However, there was no evidence to show the improvement of ductility of TZM weld metal by Re addition.

Therefore it is considered that rhenium has no beneficial effect to increase the ductility of TZM weld metal at a content of 18.5%.

4. Conclusions

Effect of rhenium and niobium on the ductility of the welds of electron-beam melted pure molybdenum and arc-melted TZM alloy by electron-beam welding have been investigated by using a 3-points simple bend test.

Main conclusions obtained are as follows;

(1) It has been made clear that rhenium was a beneficial alloying element to improve the ductility of weld metal of pure molybdenum. Ductile-to-brittle transition temperature of the pure Mo welds decreased to about 20°C and –20°C for 3.3% and 13.5%Re contents in weld metal, respectively, in comparison with 60°C of pure molybdenum welds. Taking into the consideration of the decrease in hardness of weld metal by the Re addition, this beneficial effect of rhenium was considered due to the alloy softening effect of rhenium to Mo metal.

(2) Niobium addition showed almost no improvement of ductility of pure Mo welds in the range of 1.3% to 2.7% in weld metal and tended to increase the hardness of weld metal.

(3) Re addition on TZM weld metal, however, showed no improvement of ductility of the welds even with much Re content of 18.5% in weld metal and tended to increase the hardness of weld metal.

(4) There was no obvious change in macrostructure of weld metals of pure molybdenum by the addition of rhenium up to 13.5% or niobium up to 2.7% and of TZM alloy by the addition of rhenium up to 18.5%.

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