

# Joining of Refractory Metals and its Application

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## Abstract

In this work fusion and solid state welding techniques for joining PM refractory metals are addressed. The weldability is discussed and evaluated by electron beam and friction welding experiments. EB welding experiments were performed on rolled sheets made of different Mo-based materials. While a significant decrease in the mechanical properties was found for the weldments of unalloyed molybdenum and the alloy TZM, good weldability is given for Mo41Re. A welding procedure was established and used for the manufacturing of thin walled tubes. Rotary friction welding experiments were carried out on rod material of the Mo-alloys TZM and MHC. Suitable process parameters were determined and successfully transferred for the joining of tubular TZM parts.

## Keywords

Refractory metals, molybdenum, joining, EB-welding, friction welding

## Introduction

Refractory metals such as Mo and W are used in components of several high tech industries due to their high temperature and / or physical properties. One challenge for the industrial application of these specialty materials is the development of dedicated joining technologies. Joining can be achieved by conventional mechanical methods e.g. riveting and fastening as well as by advanced brazing and welding techniques for temperature resistant, strong and tight joints. This study is focused on the joining of refractory metals produced by powder metallurgy (PM) and addresses fusion welding as well as solid state welding techniques.

Refractory metals generally exhibit limited weldability. Issues for fusion welding are the embrittlement of the weldment due to impurities and the grain growth in the heat affected zone. The impurities can be introduced by insufficient weld preparation or by interstitial pick up of O and N during the welding process [5]. Several studies were performed to understand and improve the weldability of refractory metals. For example the mechanical properties of welded TZM were investigated by [1]. Strategies considered for Mo-based materials were alloying with different Re content [4] or additions of Zr, B and C [2, 3]. Some weldability issues of fusion welding can be avoided by solid state joining techniques. Good weld properties can be achieved by diffusion bonding and friction welding. However, industrial application is limited depending on the component size and geometry. Diffusion bonding of refractory alloys should be performed under high vacuum at temperatures above 0.5 of the melting temperature.

The mechanical properties of such joints are comparable to those of the parent material. Using inter-layers the process temperatures can be significantly reduced [6]. Friction welding may be considered for parts with axial symmetry. Lison [8] demonstrated the feasibility of rotary friction welding for Mo and W. Although the process parameters are dependant on the material heat treatment condition, rotational speed and welding pressure are typically high. Therefore high power friction weld facilities and specific fixture tools are pre-requisites. This is in line with [6] who point out that Mo and W have the highest power requirements in friction welding due to their high thermal conductivity and high melting points. For the inertia friction welding of Mo typical specific welding power and minimum critical peripheral velocity are given with  $1.4 \text{ kW/mm}^2$  and  $10 \text{ m/s}$  respectively [6]. One example for the industrial application of rotary friction welding is the manufacturing of target-stem assemblies of X-ray tube applications [7]. Increased power requirements of X-ray anodes resulted in high rotational speeds and high operating temperature. Solid state joints were introduced as they offer higher strength and better creep properties compared to other joining techniques as mechanical attaching or brazing.

## 1 Fusion welding

### 1.1.1 Experimental procedures

In the first part of this study the fusion welding welding of different Mo-based alloys was investigated as this is the preferred technology for joining sheet based structures. In the basic welding experiments the following materials were evaluated: the dispersion strengthened alloy TZM (balance Mo; 0.5% Ti; 0.08% Zr; 0.01-0.04% C), the alloy Mo41Re (Mo balance, 41% Re) and commercially pure Mo (> 99.97% Mo). All sheet material was produced to a thickness of 2 mm and a length of 200 mm via the classical powder metallurgical route by pressing, sintering and hot rolling. The electron beam welding experiments were carried out in a high vacuum chamber using a standard beam gun. The joint geometry was butt joints. The samples to be welded were preheated above the ductile-brittle transition temperature in order to minimize the risk of cracking. A post weld heat treatment at  $1000^\circ\text{C}$  for 2 h under high vacuum was performed for stress relief of the weldments. For purpose of comparison tungsten inert gas (TIG)-welding experiments were performed on the TZM sheet material. In order to prevent embrittlement by interstitial pick-up of O and N, the TIG-welding was performed in the controlled atmosphere of a glove box which was purged with Ar-gas. From the welded and stress relieved sheets flat tensile test pieces with a length of 70mm and a gauge length of 20 mm were machined. The test direction was perpendicular to the weld line. Prior tensile testing the specimens were inspected by dye penetration to give evidence of crack free welds. The tensile tests were carried out at RT on a Zwick testing machine using a transverse testing velocity of  $0.1 \text{ mm/min}$ .

Based on the experiences of this basic study the material Mo41Re was chosen for the manufacturing of thin walled tubes for a heat exchanger system. The sheet material was rolled to a thickness of 0.2 mm and formed to shape. The longitudinal seam was performed by EB-welding using a dedicated fixture device. In total 50 tubes were produced and inspected. The qualification programme included visual inspection, dye penetrant testing, He-leak tight testing and water pressure testing at 200 bar for 1 min. The mechanical properties were evaluated on flat specimens. Using the established welding procedure sheet material was welded in the relevant thickness and joint geometry. Test specimens were taken from both directions (in and parallel to the rolling direction) while the weld line always was perpendicular to the designed test direction. The tensile tests were carried at RT and  $600^\circ\text{C}$ . From both, welded sheets and tubes metallographic sections were taken and investigated by light microscopy. Finally the proof for the

manufacturing of the component was given by technological bending tests. Welded tubes were bent to different radii and inspected by non-destructive testing methods.

### 1.2 Results and discussion

The weldability of the different Mo-based materials was evaluated based on the results of mechanical testing at RT. Fig. 1a presents the ultimate tensile strength (UTS) of weldments in comparison to base material properties. For all investigated materials fusion welding resulted in a decrease of strength in the weldments. However, the extent of the reduction is varying for the different materials.

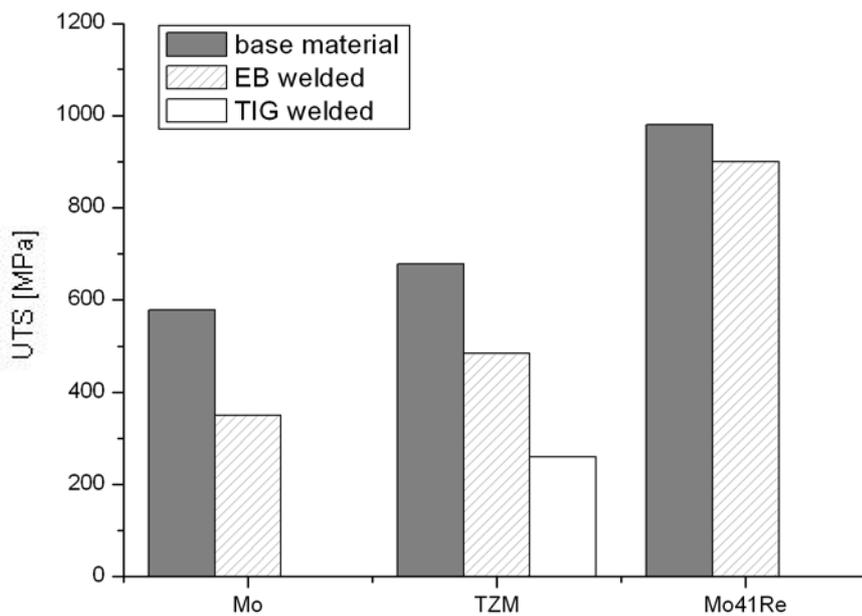


Fig. 1a: Ultimate tensile strength of welded Mo-based materials

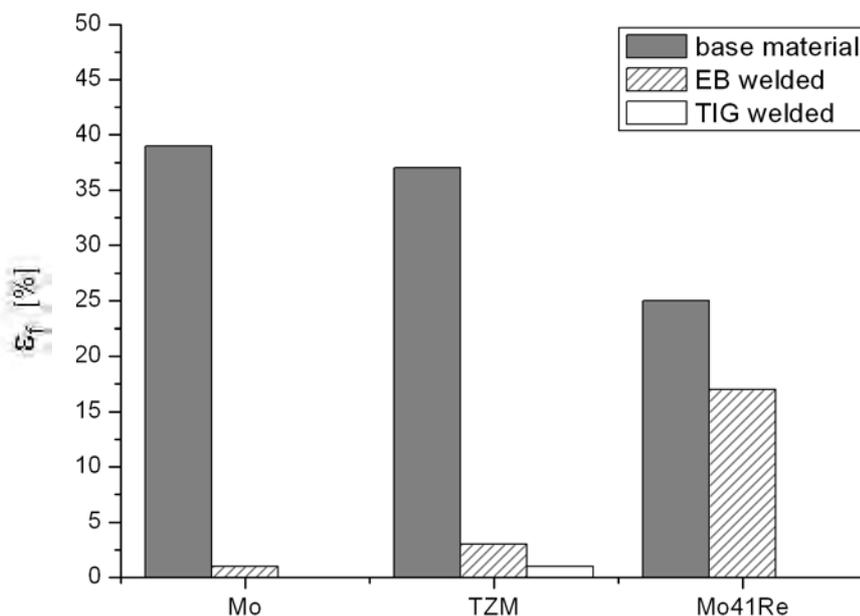


Fig. 1b: Elongation to fracture of welded Mo-based materials

The lowest joint efficiency was determined for unalloyed molybdenum. Due to excessive grain growth in the heat affected and weld zone the tensile strength of the EB - welds was only about 60% of the value for the base material. Improved joint efficiencies can be achieved when EB-welding the alloy TZM. The recrystallisation in the heat affected zone as well as the grain coarsening in the weld zone were significantly reduced compared to welded unalloyed molybdenum. High energy density processes such as EB welding are preferable. TIG welding where the high heat input leads to a broad heat affected zone resulted in low joint efficiencies of only 40%. One specific issue for the fusion welding of TZM is weld porosity. Due to the reactive alloying elements TZM sheet material typically exhibits oxygen levels of 250 ppm. Therefore gas porosity cannot be avoided in the weld zone even by using EB welding under high vacuum. Clearly the best results were obtained on the Mo41Re material where 90% of the strength of the base material can be maintained in the condition as welded. This picture was confirmed by the determination of the elongation to fracture (fig. 1b). The welds of unalloyed molybdenum exhibited brittle behaviour and values of elongation to fracture of less than 1%. Using the TZM material the ductility of the weldments was improved to 3%, still far away from the value of the base material where elongations of 35% are typical. Ductile welds were realised in the Mo41Re material. Although in tensile testing the deformation was localised in the coarse grained weld zone, values of the elongation to fracture were about 15%. Hence for highly stressed components the MoRe material can be recommended.

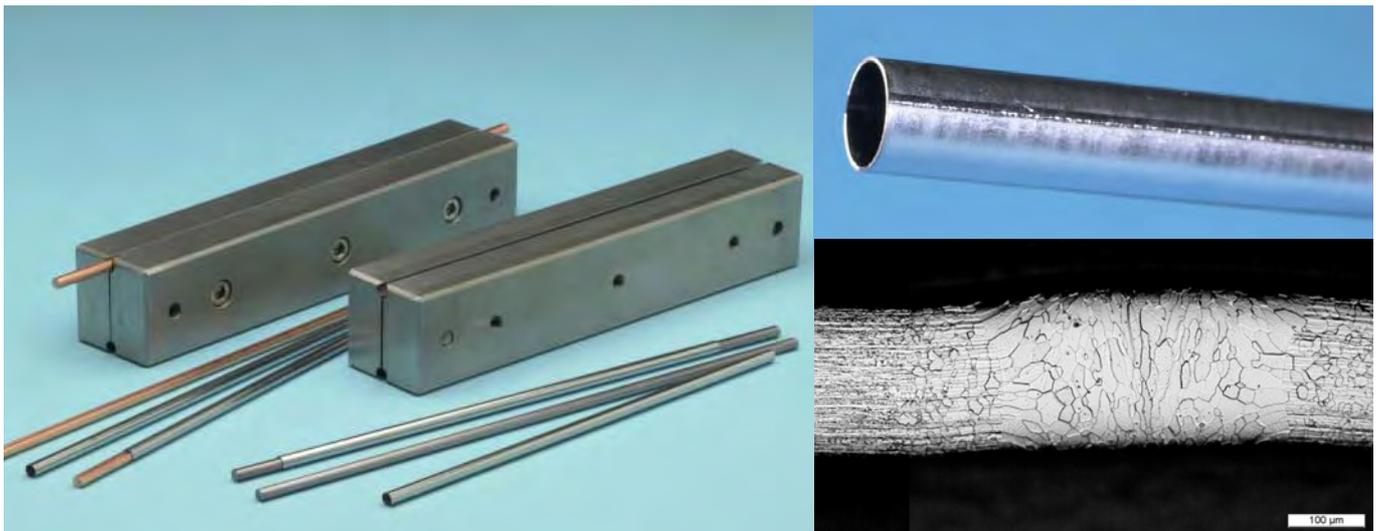


Fig. 2: Manufacturing of thin walled Mo41Re tubes by seam welding

Based on these results the Mo41Re material was chosen for the manufacturing of thin walled tubes of a heat exchanger. The longitudinal seam welding of the tubes using a dedicated welding jig is illustrated in fig. 2. Dye penetrant and X-ray testing of welded tubes gave no indication of weld defects. All welded tubes were He-leak tight and fulfilled the requirement with a leak rate  $< 5 \cdot 10^{-9}$  mbar l/s. The tubes passed successfully the water pressure test. The metallographic investigation revealed sound joints, but some small gas porosity was found below the specified size of 50  $\mu\text{m}$ . The results of the mechanical testing is given in fig. 3. The ultimate tensile strength of welded test pieces is compared to the values of the base material. For the test temperatures RT and 600°C joint efficiencies of the weldments were determined at 70% and 60% respectively. The strength values in rolling direction (RD) and transverse to the rolling direction (TD) were comparable. The technological bending tests demonstrated that bending of welded tubes up to radii of 23 mm is possible without damage.

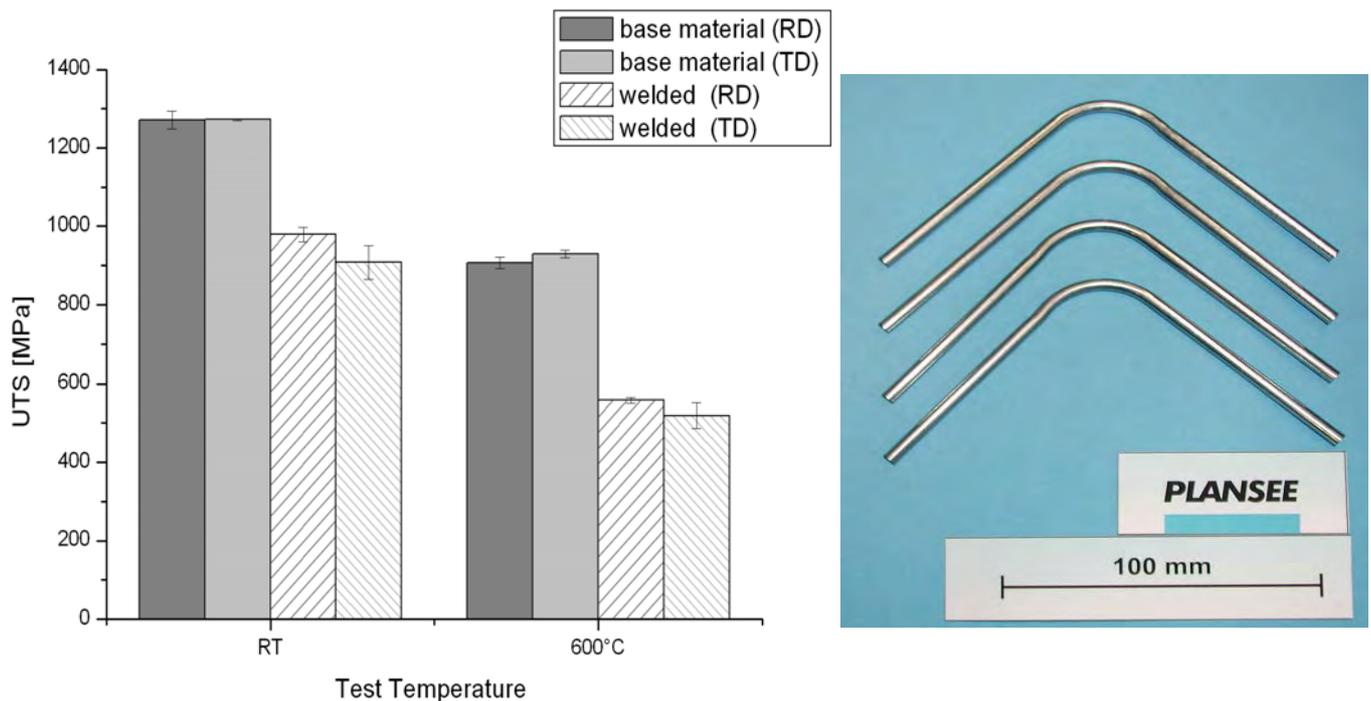


Fig. 3: a) Tensile strength of flat specimens from Mo41Re (#0.2mm) in testing at RT and 600°C; and b) welded tubes after bending test

## 2 Solid state welding

Although in the general standard [9] good friction weldability is attributed to molybdenum and tungsten, recommendations for suitable process parameters and studies on the welding of alloys are limited. In this work the rotary friction welding of the molybdenum alloys TZM and MHC should be investigated and demonstrated for typical joint designs.

### 2.1 Experimental procedures

TZM and MHC are dispersion strengthened molybdenum based alloys which exhibit increased high temperature strength and creep properties. The used material was produced by pressing, sintering and radial forging via the classical powder metallurgical route. The feasibility for friction welding was studied for the material combinations TZM-TZM and MHC-MHC. Symmetric and asymmetric joint designs were investigated using rod diameters of 15 mm and 20 mm. The basic welding experiments were carried out on a continuous motor driven friction welding facility type KUKA RSE4E. In a friction time the parts were rotated with constant speed and heated up at the faying surfaces. In the following forging phase the system was braked and the force was significantly increased resulting in expelling softened material from the joint line to the flash. In the parameter study the rotation speed was varied from 2500 to 6000 rpm. The machine set up allowed maximum friction and forging forces up to 40 kN. A burn-off in the range of 3-6 mm was intended by choosing friction times of 1-5 s and forging times of 5-15 s. These first welding trials were performed under local inert gas shielding with Ar. The welded samples were characterised by non-destructive and destructive testing. By visual inspection the continuous flash, the symmetric geometry of the weld and the visual appearance were evaluated. The dimensional inspection monitored the upset of the welded samples. Metallographic investigations were carried out on welded samples in order to check the integrity and the microstructure of the welds. After optimised welding parameters had been established for the two material combinations and joint designs, samples were friction welded and

round tensile test specimens with a diameter of 8 mm were machined thereof. All test specimens were dye penetrating tested in order to reveal cracks or other surface weld defects. The tensile tests were performed at RT and 1200°C on a Zwick testing machine using a transverse velocity of 1 mm/min.

In a next step the findings of the basic experiments were transferred in order to weld tubular TZM parts. The parts to be welded had an outer diameter of 100 mm and a wall thickness of 12 mm. The weld parameters were adapted according to the increased weld area and the larger diameter. The welding trials were performed on an inertia friction welding facility MTI 250B as this machine offers the required high forging force. This inertia welding machine can exert a maximum forging force of 890 kN. Using dedicated fixtures rotating speeds up to 4000 rpm can be achieved. In the inertia welding the flywheel was speeded up to the pre-determined rotation speed. Then the motor was disengaged and an axial force was applied until the flywheel and the work piece had stopped after a few seconds. A total of 10 pieces were welded using the established weld parameters and after a post weld heat treatment machined to test pieces. The post weld heat treatment was carried in a high vacuum furnace at 1200°C for 2 h. Visual inspection checked the integrity of all 10 welds. In addition 30% of the samples were characterised by dye penetrant and ultra-sonic testing. Finally some parts were subjected to temperatures above 1500°C in order to simulate high temperature in-service conditions. The impact of the heat treatment on the microstructure of the welds was evaluated by a metallographic analysis.

## 2.2 Results and discussion

The basic experiments demonstrated that the investigated molybdenum alloys can be successfully friction welded. Due to their high temperature strength and the good thermal conductivity it is difficult to reach a sufficient level of plastification. Too less plastification can lead to different work hardening of partial areas in the weld zone which may cause delamination or cold cracking. If the input of weld energy is excessive, the upset cannot be adequately controlled. In the parameter study the following parameters were established for the welding of TZM and MHC (table I).

Table I: Optimised parameter for the friction welding of TZM and MHC

Material combination	Joint design	Rotation speed [rpm]	Friction force [N/mm <sup>2</sup> ]	Forge force [N/mm <sup>2</sup> ]
TZM-TZM	D15 / D15	4200	60	100
	D15 / D20	5000	70	100
MHC-MHC	D15 / D15	4500	180	225
	D15 / D20	4500	225	225

The welding experiments clearly demonstrated that for both materials preheating using a high rotation speed (> 4000 rpm) at a low friction force was preferable. Then the friction force was increased to achieve plastification. While good plastification was realised during welding TZM at the rather low friction force of 60-70 N/mm<sup>2</sup>, the friction force for sufficient plastification was significantly higher in case of MHC. In the forging phase the force was further increased to 100 N/mm<sup>2</sup> and 225 N/mm<sup>2</sup> respectively when welding TZM and MHC.

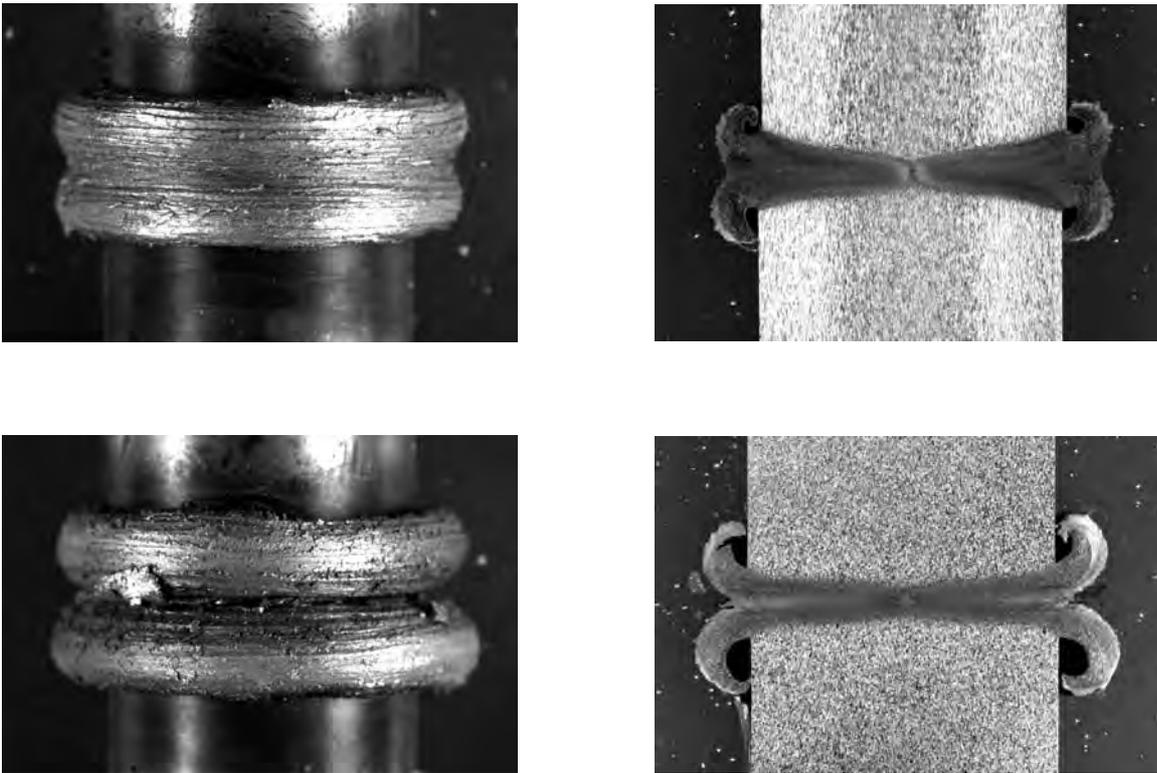


Fig. 4: Photographs and metallographic sections of friction welded TZW and MHC rods (D15 / D15 mm)

In fig. 4 photos and the metallographic sections of friction welded TZW and MHC rods (D15 / D15) are presented. In the welded TZW samples the flash formation is very regular and continuous which indicates a good weldability. In case of the MHC material the flash formation is less smooth. This is in line with the observation that there is a higher risk of delamination due to different work hardening when welding MHC material. Hence the process window was generally found smaller for MHC than for TZW. Using optimised welding parameters the upset can be well controlled for both material combinations. The equal upset on both sides indicates a symmetric heat transfer and a good weld. The welded samples of MHC exhibit a stronger expulsion of flash material and a narrower weld zone. This is attributed to the higher used forging force. The welding of asymmetric joint designs was evaluated to be more critical. The flash formation and the upset was less reproducible. This risk of weld defects was clearly more pronounced in both materials for the joint design D15 / D20. Especially in the weld area near the edge some delaminations were detected. This can be explained by the asymmetric heat transfer and locally insufficient plastification.

In fig. 5a and b the tensile properties of friction welded TZW and MHC samples are given for the test temperatures RT and 1200°C respectively. The values of ultimate tensile strength were determined for both joint designs and compared to the tensile properties of the base material. At RT testing the friction welds of both alloys had lower strength and elongation values than their base material. All friction welded test samples fractured in the weld or heat affected zone. For the MHC material joint efficiencies of about 65% were determined irrespective of the joint design. The test data of friction welded TZW showed more scatter. The ultimate tensile strength values were 520 and 190 MPa for the joint designs D15 / D15 and D15 / D20 respectively. Hence the asymmetric joint design resulted in a significantly lower joint efficiency. The better joint efficiencies of MHC compared to TZW are somewhat in contradiction to the experiences

during the welding tests where the process window was narrow in case of MHC. However, the higher forging force used for welding MHC obviously gives advantages regarding the strength of the welds.

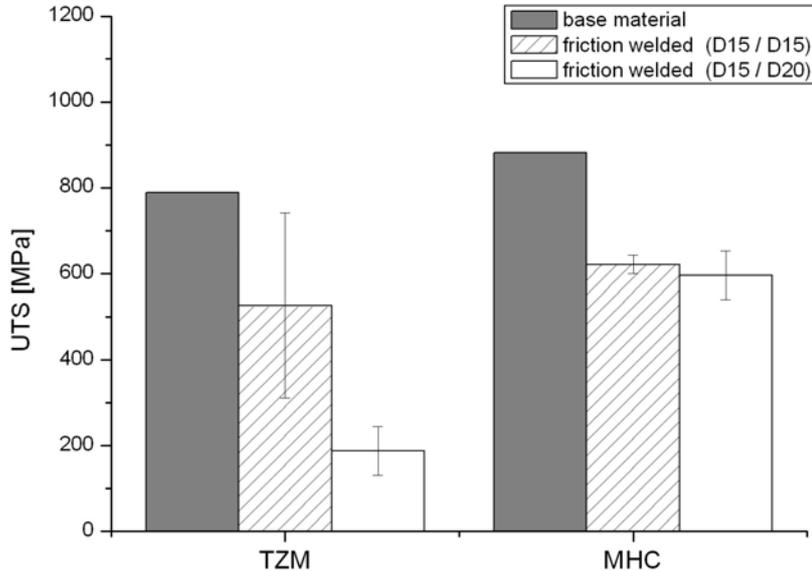


Fig. 5a: Ultimate tensile strength of friction welds tested at RT

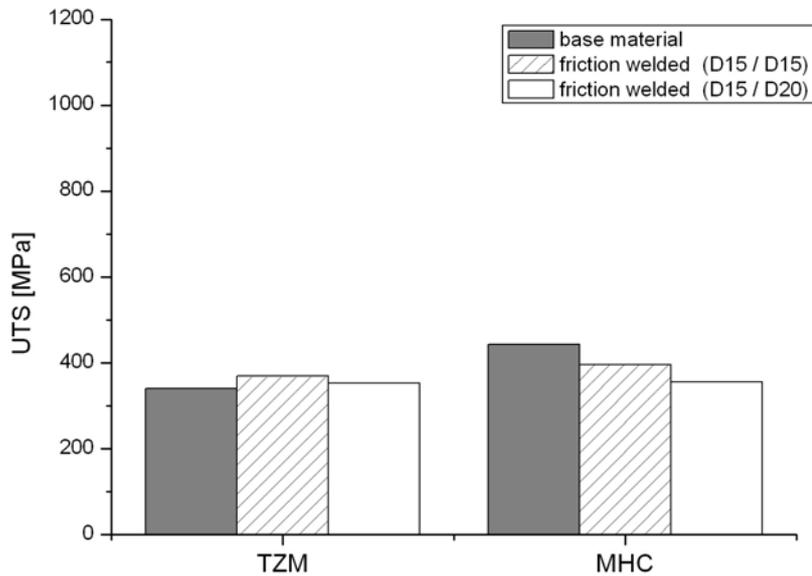


Fig. 5b: Ultimate tensile strength of friction welds tested at 1200°C

In fig. 6 one can see the welding of the tubular TZM parts on the inertia welding machine and the result of one welded part. Visual inspection showed an equal upset and continuous flash formation. The dye penetrant testing gave no indication of cracks, delaminations or other surface defects in the friction welded parts (fig. 7a). Verification of the test method was given by the results on a reference sample which exhibited several weld defects. The ultra-sonic testing confirmed the result of the excellent weld quality. In all tested parts no critical weld defects larger than 3 mm were detected. A typical photo of the ultra-sonic scan is presented in fig. 7b. This part was non-destructively tested after a high temperature treatment and after a section for metallographic analysis had been removed.

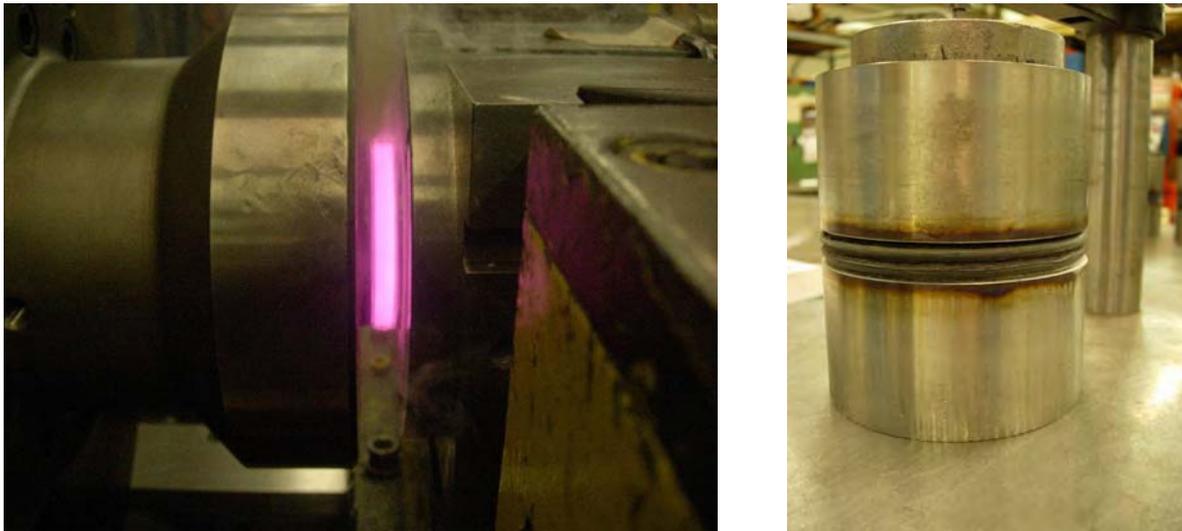


Fig. 6: Welding of tubular TZM parts on the inertia welding machine

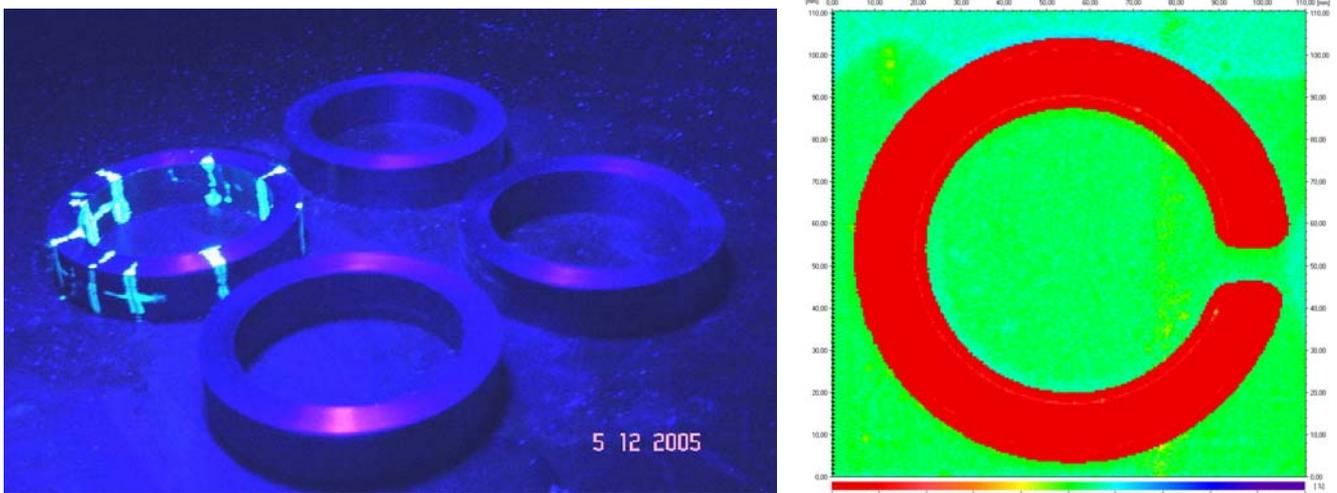


Fig. 7: a) Three friction welded parts (with no defects) and reference part (with multiple weld defects) in the dye penetrant testing and b) ultra sonic scan of a friction welded test piece

Fig. 8a shows the light microscopy photo of an etched metallographic section in the condition as friction welded. One can see three different regimes: the weld zone, the heat affected zones and the parent material. The weld zone has a thickness of about 2 mm and a fine grained microstructure. This microstructure may arise from dynamic recrystallisation during the welding process. This is supported by the metallographic analysis before and after high temperature loading where no significant difference in grain size was determined for this zone (fig. 8b). However, a significant coarse graining by the heat treatment was found in the heat affected zone, adjacent on both sides to the weld zone. It is believed that the friction welding introduces a high degree of deformation in this zone, but the heat input during welding is too low or too short to initiate recrystallisation. Hence there is a high driving force for grain growth in a post weld heat treatment. The high temperature loading obviously resulted in large grains up to several 100  $\mu\text{m}$  in this zone. For high temperature applications this change in the microstructure of the friction weld has to be taken into account.

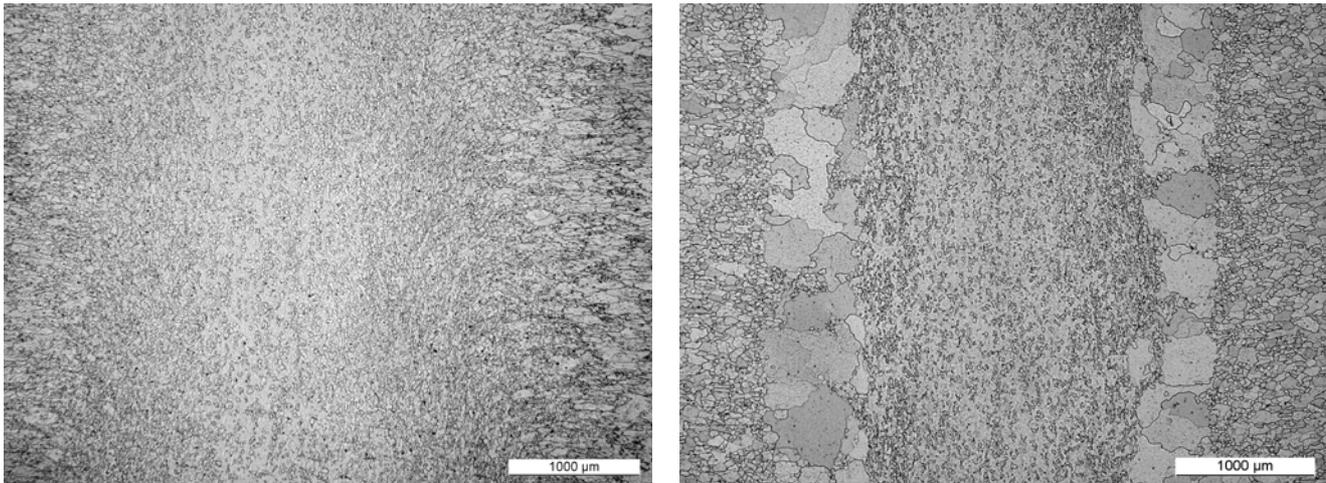


Fig. 8: Metallographic section of a friction weld before (a) and after (b) high temperature heat treatment

### 3 Conclusions

The weldability of different Mo-based materials was discussed and evaluated by EB-welding and rotary friction welding experiments. EB-welded Mo41Re exhibited good joint efficiency and weldability. When fusion welding unalloyed molybdenum or the alloy TZM a significant decrease in strength and ductility cannot be avoided due to grain growth in the weld and heat affected zone. A welding procedure for 0.2 mm thick Mo41Re foil was established and applied for the manufacturing of thin walled tubes. Rotary friction welding experiments were carried out on rod material of the Mo-alloys TZM and MHC. Suitable process parameter were determined and successfully transferred for the joining of tubular TZM parts. The mechanical properties of friction welds were found comparable to the base material when testing at elevated temperatures. However, high temperature loading lead to grain growth in the heat and deformation affected zone of the weldments.

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