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Analysis of tensile and fracture toughness results on irradiated molybdenum alloys, TZM and Mo–5%Re

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Abstract

Due to their good resistance at high temperature, good thermal conductivity and swelling resistance, molybdenum alloys are amongst the candidates for divertor structural materials. However, little is known about their tensile and fracture toughness behaviour, in particular after irradiation-induced embrittlement. This paper aims to investigate the tensile and fracture toughness properties of two molybdenum alloys, namely TZM and Mo–5%Re. Tensile and compact tension specimens were irradiated in the BR2 reactor at 40°C and 450°C up to a fast neutron fluence of 3.5×10^{20} n/cm^2 (0.2 dpa). Tests were performed on both precracked and notched specimens. Results show a drastic decrease of the ductility due to irradiation, but only a slight decrease of the fracture toughness in the lower shelf domain. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

The structural materials and plasma facing materials (PFMs) are important elements of a fusion reactor.

In the European long-term technology programme [1], structural materials for the blankets, the first wall and the shielding are currently investigated: austenitic stainless steel, SiC/SiC ceramic composites, V-alloys, Ti-alloys and Cr-alloys, reduced activation ferritic martensitic (RAFM) steels and oxide dispersion strengthened (ODS) steels. However, these materials do not have the necessary refractory properties to be used in the armour.

The three categories of PFMs under consideration in the fusion scientific community [2–4] are non-metallic materials (graphite, carbon fibre composite), low Z metallic element beryllium, and high Z metallic elements (tungsten, molybdenum). The PFMs will be submitted to tremendous heat and radiation flux and disruptive loading. The choice of the PFMs will be dictated by various criteria [3] such as tritium permeation, plasma–PFMs interactions, erosion resistance, thermal fatigue, swelling and embrittlement, creep properties, thermal ageing, neutron activation, biological hazards, fabricability and cost.

The most interesting properties of refractory metals are the high melting point, high mechanical resistance at high temperature, low thermal expansion coefficient and good thermal conductivity, which induce an excellent dimensional stability and a good resistance to thermal shock. In [4], the tensile properties of 10 refractory materials were studied: sintered and wrought Mo and W, phases dispersed Mo base alloys: $Z6$ ($Mo{-}ZrO_2$) and TZM and solid solution alloys, Mo–5%Re, Mo–41%Re, W–5%Re, W–26%Re. The study concludes that TZM and Mo–41%Re are the most resistant materials with good ductility.

The tensile properties for candidate divertor armour materials Mo- and W-alloys are now well established. However, the assessment of the effect of radiation damage on the tensile, fracture toughness and fatigue properties still requires a large effort. In this context, the tensile and toughness properties of two materials TZM and Mo–5%Re are investigated in this study.

2. Materials and irradiation condition

Two molybdenum alloys, TZM and Mo–5%Re, were supplied in stress-relieved condition by Plansee AG, Reutte, Austria.

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The TZM is a dispersed and single-phase alloy with a body centred cubic crystalline structure. The dispersed phase consists of Ti and Zr carbides, which are distributed inside the grains [4,5]. The presence of a dispersed phase prevents grain size increase at high temperature [4,5]. It increase the recrystallisation temperature, improves the tensile strength and decreases the ductility [5].

The Mo–5%Re is a substitution solid solution; the structure remains single phase with a body cubic centred crystalline structure [4,5]. The addition of rhenium increases the ductility and the recrystallisation temperature [5].

The chemical compositions of both alloys are given in [6]. The chemical analysis was performed at KFA-Jülich. The low level of impurities and the good stability of Mo make that these two alloys are characterised by a low activation due to neutron irradiation. A tensile specimen irradiated in MOST1 (see below) up to a fluence of 3.50×10^{20} n/cm^2 ($E > 1$ MeV) gives a dose rate of 3 μSv/h at a distance of 300 mm after being stored during five years.

The elastic mechanical properties are given in [5,6]. The Poisson ratio is 0.32 and the Young's modulus, E is 320 GPa at room temperature.

The material was produced in form of rods [7]. Specimens were machined by Plansee AG. For both materials, metallographic examinations perpendicular and parallel to the axis of the rod show elongated grains parallel to the rod axis [7].

Both materials were irradiated in two conditions in the BR2 materials testing reactor located at SCK-CEN, Mol (Belgium). Two irradiation experiments were performed: MOST1 and MOST2 [8]. The main parameters of the irradiation experiments are summarised in Table 1. Although a NaK liquid metal bath was used for the evacuation of the heat produced by the gamma rays and to homogenise the temperature, a homogeneous irradiation temperature was not obtained along the rig. Although not representative of fusion irradiation condition, this experiment allows to assess the radiation embrittlement and the effect of the irradiation temperature on the mechanical properties.

3. Experiments

The tensile properties of the TZM and Mo–5%Re are measured with small tensile specimens. Specimens have a diameter of 3.1 mm, a length of reduced section of 15 mm and a total length of 40 mm.

For high temperature testing, specimens are kept inside a furnace for 30 min before testing: in the soaking period and during the test, the temperature is controlled within ±2°C of the nominal value. Tests are performed in displacement control at a constant crosshead speed of 0.2 mm/min. A self-align tensile line is used to prevent bending. Load versus crosshead displacement records are analysed to derive the tensile properties. Extraneous elastic displacements due to the machine compliance are subtracted in order to adequately derive the tensile properties. The test matrix given in [6] contains a total of 28 tensile tests in the non-irradiated and irradiated conditions for temperature varying from 25°C to 450°C.

The fracture toughness of TZM and Mo–5%Re material is measured with small standard Disk-Shaped Compact Tension specimens DC(T). The orientation of the DC(T)s is R–C [7]. The design is based on the ASTM E399 recommendations. The specimen has a nominal width of 16 mm and a nominal thickness of 8 mm. The geometry and size were chosen to allow a large number of specimens to be irradiated in the MOST1 and MOST2 experiments. Notches for the application of a clip gauge were machined at SCK-CEN on irradiated and non-irradiated specimens.

Although very brittle, both materials display small non-linearties in the load versus Load Line Displacement (LLD) record at high temperature in the non-irradiated condition. Therefore, tests were not analysed according to the ASTM E399, which was developed in a Linear Elastic Fracture Mechanics (LEFM) context. Instead, tests were conducted according to the more general ASTM E1820 standard allowing for non-linear behaviour.

The experimental test matrix was selected to allow characterisation of the fracture toughness of the two materials in the non-irradiated and irradiated conditions for temperatures varying from 25°C to 450°C. The test matrix initially planned is given in [6].

Prior to fracture toughness testing, a crack of about 2 mm ahead of the notch is generated by fatigue. The obtained precracked specimen is mounted on a dedicated tension testing grips of a universal tensile machine. A clip gauge is mounted on the specimen to measure the LLD. The set-up is heated and kept at the desired temperature ±2°C for at least 30 min prior to and during the test. The specimen is then loaded in

Table 1
Irradiation conditions in the BR2 reactor [8]

	Fluence $E > 1$ MeV (n/cm^2)	Damage (dpa)	Coolant	Irrad. temp. min/max (°C)
MOST1	3.50×10^{20}	0.35	Water	40
MOST2	2.88×10^{20}	0.29	NaK	370/475

displacement control at a constant crosshead speed of 0.2 mm/min up to unstable rupture. The specimen dimensions, crack length and load versus LLD record are used to determine the fracture toughness.

Preliminary tests showed that accomplishing the experimental matrix would have been particularly difficult. The main encountered difficulties were:

1. The molybdenum material is particularly brittle and therefore it is very likely that the specimen breaks while being mounted into the tension line.
2. Initiation of fatigue precracking requires high load values but propagation can be very fast. As the material is very brittle, there is a high probability to break the specimen during fatigue.
3. For conventional materials, fatigue crack growth can be visually monitored on the polished external surface of the specimen. Due to the low ductility of this material, fatigue crack growth is not visible even when using a microscope. It is thus difficult to monitor the crack growth with conventional techniques.
4. The fracture surface texture of the fatigue crack growth is not distinguishable from that of unstable crack growth. It is thus not straightforward to determine the initial crack length.
5. The maximum operational temperature capacity for clip gauges and extensometers is generally limited to 200°C. Consequently, conventional extensometers cannot be used.
6. Due to the small size of the specimen, it is not practical to measure the LLD as the available space is not sufficient.
7. The MOST1 and MOST2 specimen are activated. Therefore, test are performed remotely in hot-cell using manipulators.

A lot of specimens were broken during fatigue. However, we achieved to precrack unirradiated specimens of the Mo–5%Re material on a resonant testing machine. The stress intensity factor during fatigue is limited to 14 MPa $\sqrt{\mathrm{m}}$, which is outside ASTM E1820 requirements ($K_{\mathrm{fatigue}} \leqslant 10$ MPa $\sqrt{\mathrm{m}}$). As crack growth cannot be monitored with conventional optical techniques, an alternative method was used. This method is based on the change of the resonant frequently of the system with crack growth.

To distinguish fatigue precrack and unstable crack, specimens were heat tinted at 300°C during 4 h. This thermal ageing cannot have an important effect on the material properties as the material is not irradiated and the recrystallisation temperature of molybdenum is very high. This was demonstrated in [9] by performing impact three-point bend tests on annealed (100 h at 600°C) and non-irradiated condition.

As the TZM material in the non-irradiated condition could not be precracked, specimens were tested without crack. The test matrix was changed in order to study the effect of precracked versus not precracked specimens on the fracture toughness of the Mo–5%Re in the non-irradiated condition.

The analysis of the load displacment trace is performed according to the ASTM E1820 standard. No side-grooves were machined as no ductile crack growth was expected. The stress intensity factor (K) and (J)-integral (J) equation are given in [6]. K and J are a function of load, specimen thickness, specimen width, crack length and plastic area under the load versus load line displacement (LLD).

The crack mouth opening displacement (CMOD) is measured at the external surface of the specimen using a clip gauge. This position is slightly different from the LLD. Therefore, relations given in [6] are used to derive the LLD from the CMOD.

In order to instrument the test at high temperatures, different clip gauges were used. The most satisfactory clip gauge is based on bonded strain gauges. It was qualified and developed at SCK-CEN for temperatures ranging from −150°C to 300°C [10]. All tests in non-irradiated and irradiated conditions were instrumented. More details on the instrumentation are given in [6].

4. Results

4.1. Tensile test results

The tensile test results in the non-irradiated condition are reported in [6] and shown in Figs. 1 and 2. Tests were performed on a 100 kN electromechanical universal testing machine.

The tensile tests in the irradiated condition were performed on a 20 kN servo hydraulic universal testing machine located in hot cell 11 of the Laboratory of High and Medium Activity (LHMA) at SCK-CEN. Results from the MOST1 and MOST2 irradiation experiments are reported in [6] and shown in Figs. 3 and 4. For both irradiated materials, the analysis of the load versus

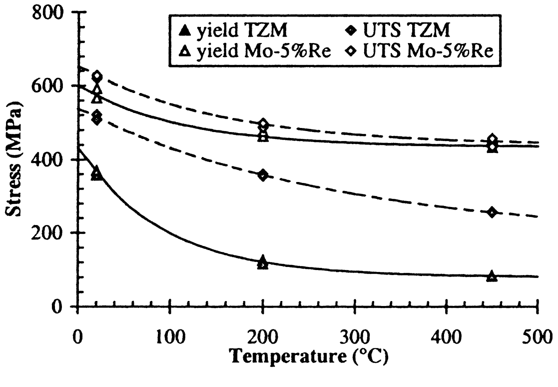

Fig. 1. Yield and ultimate tensile strength (UTS) for the TZM and Mo–5%Re in non-irradiated condition.

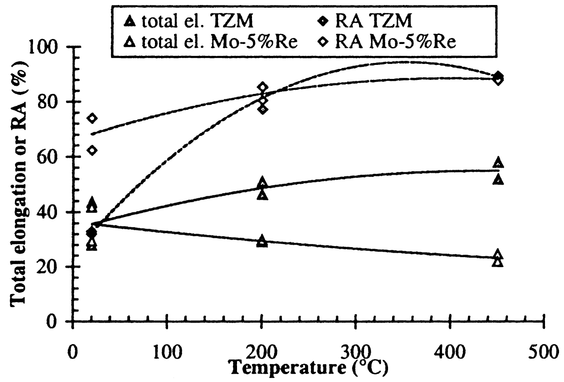

Fig. 2. Reduction of area and total elongation for the TZM and Mo–5%Re in non-irradiated condition.

displacement record show that yield point and maximum point collapse into a single point. Specimens tested at 200°C on the TZM material irradiated in MOST1 did not break in the gauge length section. This can be attributed to the brittleness of the material and to its sensitivity to stress concentrations. Tests at room temperature foreseen for TZM MOST1 were not performed. Instead, they were tested at 450°C. However, as the test temperature is much higher than the irradiation temperature, the test can introduce an annealing of some defects induced by the neutron irradiation.

4.2. Fracture toughness test results

Fracture toughness tests on the non-irradiated material with DC(T) specimens were performed on a 100 kN electromechanical universal testing machine. Results on precracked DC(T) from the Mo–5%Re material are given in [6] and shown in Fig. 5. The crack length is measured using the nine-point averaging method. It should also be noted that no ductile crack growth is observed. All tests except one performed at 450°C display a linear load displacement record. Four specimens displayed significant pop-in events. A pop-in is a small unstable crack growth followed by a crack arrest. The significance of a pop-in is established in accordance with ASTM E1820–96 appendix A4.

For the TZM and Mo–5%Re material in the non-irradiated condition, fracture toughness tests were also performed on non-precracked specimen with a notch tip radius of about 0.4 mm. Indeed, a correlation between precracked versus not precracked fracture toughness

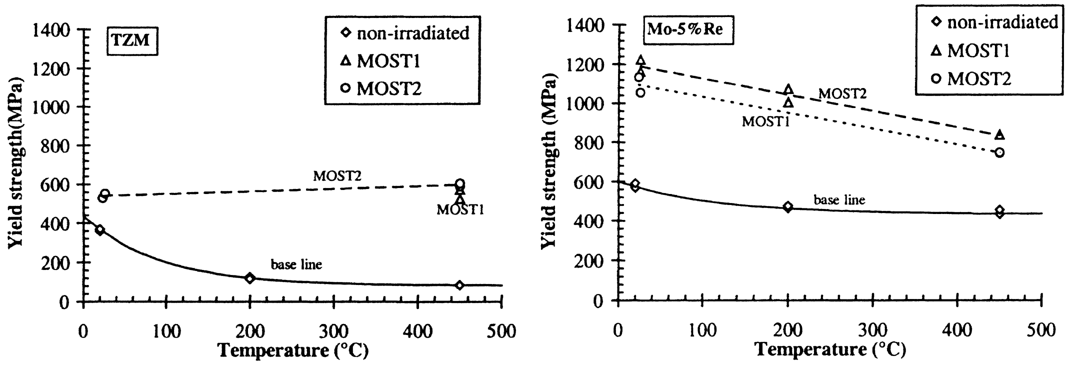

Fig. 3. Yield strength of the TZM and Mo–5%Re material.

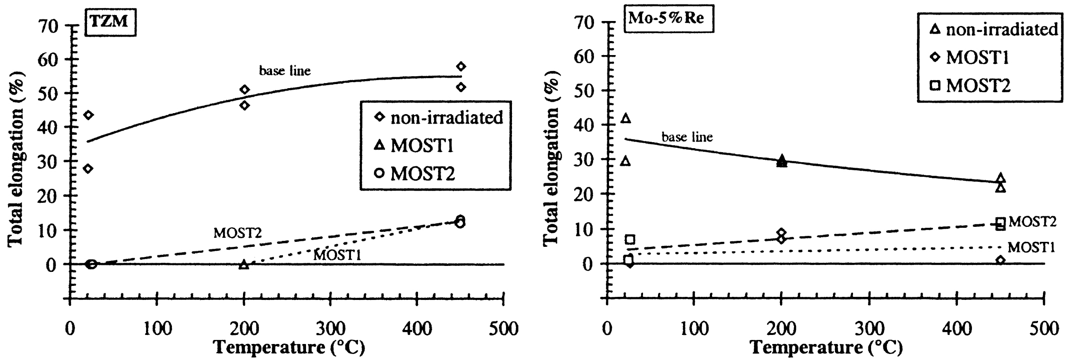

Fig. 4. Total elongation of the TZM and Mo–5%Re material.

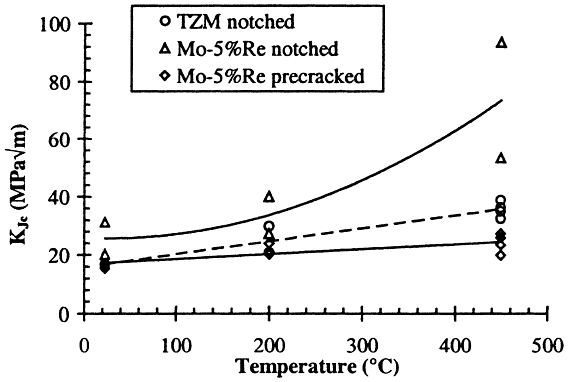

Fig. 5. Fracture toughness in the non-irradiated condition.

exists for brittle materials [7]. It has been established that a large notch tip radius leads to an over-estimated fracture toughness. A correlation between precracked and notched specimens has been established empirically [7]:

$$K_{\mathrm{c,notched}} = K_{\mathrm{c,precracked}}\left(1 + \sqrt{r/r_0}\right), \tag{1}$$

where r is the radius of the notch and r_0 a fitting parameter.

Fracture toughness tests on notched specimen are reported in [6] and shown in Fig 5. Neither pop-in nor ductile crack growth is observed.

Fracture toughness tests on irradiated specimens were performed on the same machine used for irradiated tensile tests. Specimens were precracked before irradiation by CRM-Liège. Results for the MOST1 and MOST2 experiments are given in [6] and shown in Fig. 6. No ductile crack growth was observed.

5. Discussion

In the non-irradiated condition the yield strength of Mo–5%Re is higher than for the TZM material. The TZM material has a higher strain-hardening capacity than Mo–5%Re. The ductility evaluated by the total elongation at rupture and by the reduction of area is good for both materials. An increasing strain at rupture with decreasing temperature is found for the Mo–5%Re material, an opposite behaviour is found for TZM material.

The effect of irradiation on the tensile properties of both materials is a strengthening of the material (see Fig. 3) and a reduction of ductility (see Fig. 4). From the

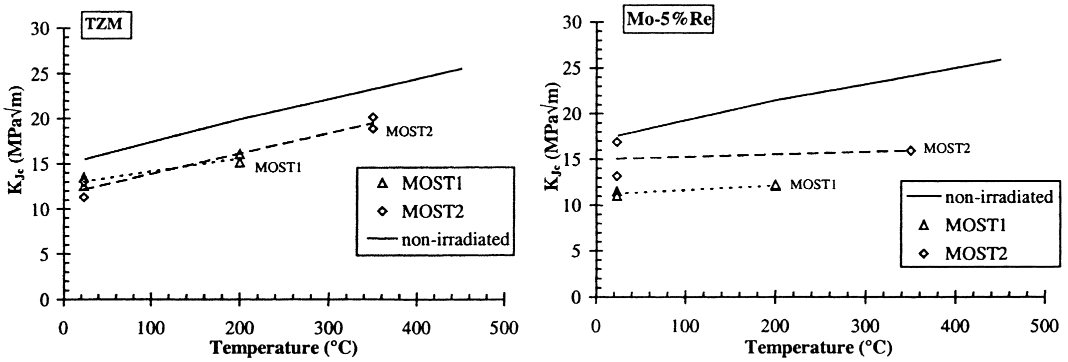

Fig. 6. Effect of the irradiation on the fracture toughness of the TZM and Mo–5%Re material.

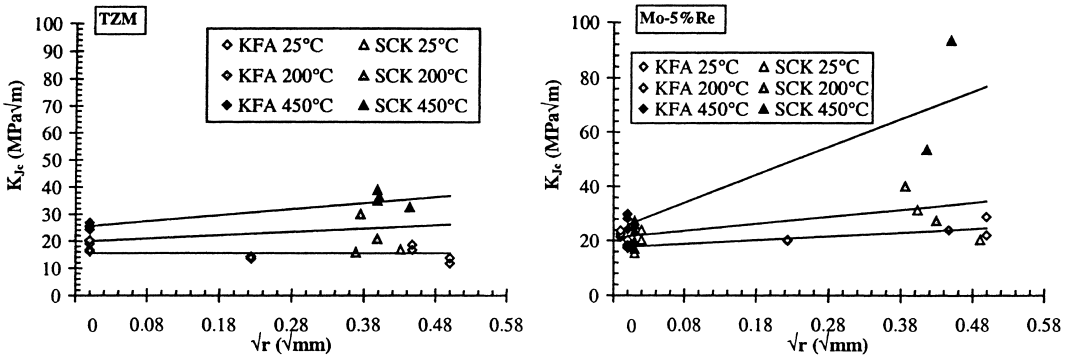

Fig. 7. Fracture toughness of the TZM and Mo–5%Re material in the non-irradiated condition.

ductility point of view, Mo–5%Re retains a higher ductility after irradiation, especially at low temperature (see Fig. 4). This is particularly important for start-up and shut down-phases.

Specimen in MOST1 and MOST2 received approximately the same fluence. The tensile results from MOST1 experiment shows a slightly higher irradiation embrittlement for Mo–5%Re (see Figs. 3 and 4) as compared to MOST2. There is a decreasing irradiation hardening with increasing irradiation temperature. This effect, also observed in [11] on Mo–5%Re and Mo–5%Re, is not very large for irradiation temperatures in the range 50°C to 500°C.

Fig. 5 shows an increasing fracture toughness with increasing temperature. This figure allows the comparison of the fracture toughness of notched versus precracked specimens for the Mo–5%Re in the non-irradiated condition. The notched specimens give a systematic higher apparent fracture toughness as compared to the precracked specimens. This is expected as the crack induces a more severe stress concentration. Another difference is that no pop-in's are observed for notched specimens. A possible explanation is that the notched specimens are loaded to higher loads and therefore crack arrest is more unlikely. Fig. 5 also shows a better fracture toughness of Mo–5%Re as compared to the TZM material.

The same materials and geometry were tested by KFA [7]. The analysis was based on the ASTM-E399 standard. The test matrix given in [6] contains 39 tests in the non-irradiated condition.

The effect of the notch radius on the fracture toughness is shown in Fig. 7. This effect can be rationalised using Eq. (1). The overestimated fracture toughness of notched specimen is more pronounced at high temperature. The scatter of fracture toughness data is more pronounced at high temperatures. This might be due to the fact that tests were performed closer to the brittle-to-ductile transition.

To discuss radiation embrittlement, the evolution of fracture toughness as a function of temperature is depicted in Fig. 6. The fracture toughness values of the notched specimens are normalised using Eq. (1). These figures show that the effect of irradiation is a small decrease of the fracture toughness. Fig. 6 also shows a beneficial effect of irradiation temperature on the fracture toughness of the Mo–5%Re material.

In the non-irradiated and irradiated conditions, no ductile crack growth is observed. Therefore all results were obtained in the lower shelf regime and the ductile-to-brittle transition in term of fracture toughness is higher than 450°C. Consequently, no definitive conclusion can be drawn on the eventual shift of the ductile-to-brittle transition.

6. Conclusions

Tensile and fracture toughness properties of two molybdenum alloys, namely, TZM and Mo–5%Re have been investigated in the non-irradiated and irradiated conditions. The main findings are:

- TZM and Mo–5%Re materials show good tensile strength and ductility in the non-irradiated condition in the range 25°C to 450°C.
- There is a drastic diminution of the ductility due to irradiation for both materials. However Mo–5%Re retains a better ductility.
- Fracture toughness slightly due to neutron irradiation.
- The lower irradiation temperature of MOST1 induces a higher reduction of ductility and of fracture toughness for the Mo–5%Re.
- Fracture toughness tests on notched and precracked specimen can be rationalised using Eq. (1).
- Fracture toughness increases with temperature in nonirradiated and irradiated conditions.
- No ductile crack growth was observed. Therefore, the range 25°C to 450°C is inside the lower shelf domain.
- Higher test temperatures would be needed to obtain information on the shift of the ductile-to-brittle transition.

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