Thermal Shock Behavior Under Deuterium Plasma Exposure of Tungsten–Tantalum Alloys

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Abstract

Tungsten–tantalum (W-Ta) alloys for the application to fusion reactor divertor were developed to overcome the issues of W related to the low temperature brittleness, embrittlement by recrystallization, and embrittlement by neutron irradiation. In the present study, the response to cyclic thermal shock tests as well as the fundamental mechanical properties of W-3%Ta and pure W were investigated, which simulated a transient heat load event in the actual divertor environment. Because it was concerned that the effect of deuterium (D) plasma exposure might be much more pronounced in the Ta-alloyed materials, the thermal shock tests were performed under the background steady state D-plasma exposure. Based on the present study, W-3%Ta might be a competitive alloy system from the viewpoint of thermo-mechanical response in the actual divertor environment as well as the fundamental mechanical properties and resistance to recrystallization.

Keywords:

Fusion reactor, Tungsten, Alloying, Tantalum, Mechanical properties, Thermal shock, Deuterium

1. Introduction

Divertor, which has a removal function of waste materials from fusion plasma, is one of the most critical components to realize nuclear fusion reactors with ability of power generation, safety, and economy. Plasma facing materials (PFMs) of the divertor will be suffered by the steady state and transient heat loads. As a result, degradation of divertors could be induced due to the melting, cracking, and deformation of PFMs [1].

Pure tungsten (W) is a primary candidate for the PFMs of divertor of ITER and DEMO because of high melting point, high thermal conductivity, and the other several physical properties. However, low temperature brittleness [2] and embrittlement caused by recrystallization [3] and neutron irradiation [4] are intrinsic issues of W under fusion reactor environment. According to Geach *et al*. [5], it was firstly clarified that the solid solution alloying by rhenium (Re) could improve the ductility of W. The previous studies also reviled that the mechanical properties not only before but after the neutron irradiation were improved due to the alloying by 3%Re [6, 7]. However, the irradiation-induced embrittlement of Re-alloyed W materials may become more significant compared to non-alloyed materials above certain neutron fluence because of the higher amount of solid transmutation products, which could be due to the irradiation enhanced precipitation and formation of Re-rich clusters [8]. Therefore, complete solid solution elements in W, e.g., tantalum (Ta), vanadium (V), niobium (Nb), and molybdenum (Mo), could be an alternative candidate of alloying element to suppress those issues.

In the framework of our recent research, W-Ta alloys for the PFMs were developed. As well as the fundamental mechanical properties, thermo-mechanical response under heat load environment was investigated, which is expected in the actual PFMs. In this paper, the fundamental mechanical properties and degradation behavior under cyclic thermal shock tests of W-3%Ta alloy and pure W, which simulated a transient heat load event like edge localized modes (ELMs), are reported. Because hydrogen-induced embrittlement is an issue of Ta, it was concerned that the effect of deuterium (D) plasma exposure might be much more pronounced in the W-Ta alloys. Therefore, the thermal shock tests in the present study was performed under the background steady state D-plasma exposure.

2. Experimental

W-3%Ta alloy and pure W plates with a thickness of 5 mm, supplied by A.L.M.T. corp. in Japan, were examined, which were fabricated by powder metallurgy and hot rolling followed by stress relief heat treatment. The fabrication route of both materials was the same except the addition of 3%Ta. The fundamental mechanical properties of the pure W plate were described in open literature [7, 9, 10].

To characterize the both materials, observation of grain structure, measurement of grain

size, and evaluation of recrystallization behavior under isochronal heat treatments for 1 h up to 2300 °C were performed. The recrystallization behavior was evaluated by the Vickers hardness measurement (load: 1.96 N, dwell time: 15 s). These evaluations were conducted on the L × S surface, where the L, T, and S directions correspond to the rolling direction, normal to L direction, and perpendicular to both L and T directions (direction along plate thickness), respectively.

To evaluate the effect of alloying by 3%Ta on the fundamental mechanical properties in the as-received condition, tensile and Charpy impact tests of both materials in the as-received condition were performed. Tensile tests at temperatures up to 1300 °C were carried out using plate specimens with 5 mm in gauge length, 1.2 mm in gauge width, and 0.5 mm in thickness. The loading axis was aligned along the L and T directions. To obtain the microstructure after heat treatment at each tensile test temperature, temperature duration for tensile test were 1 h. Charpy impact tests along the L-S direction at temperatures up to 1000 °C were carried out using a test apparatus at Karlsruhe Institute of Technology, which were conducted under the EU standard [11]. As for the nomenclature of L-S direction, the first letter (L) and the second letter (S) correspond to the direction perpendicular to the expected crack plane and the expected direction of crack growth, respectively. A KLST-type specimen was used, which has 27 mm in length, 3 mm in width, 4 mm in height, 1 mm in notch depth, and 0.1mm notch root radius.

In addition, tensile tests of W-3%Ta alloy after D-exposure were performed to investigate the effect of D-exposure on the tensile properties. The D-exposure experiments were carried out using a vacuum apparatus with a high temperature furnace at Shizuoka University. The detailed structure is shown in ref. 12. The specimens were put into a metal chamber and heated up to 600 °C by an electric furnace under the pressure range of 10^{-5}–10^{-3} Pa. After the temperature reached 600 °C, D_2 gas at 0.8 or 80 kPa was introduced into the chamber and maintained for 6 or 24 h, respectively. Then, the chamber was air-cooled to room temperature.

Cyclic thermal shock tests by pulsed irradiation of Nd:YAG laser with background of a steady state D-plasma exposure was performed using a linear plasma device, PSI-2, at Forschungszentrum Jülich [13]. Specimens with the dimension of 10 mm in L direction, 10 mm in T direction, and 4 mm in S direction were machined from the plates and L × T surface as heat-loaded surface was mechanically polished to mirror state. Base temperature, number of laser pulse, laser power density, pulse duration, and D-plasma fluence were 700 °C, 1×10^3–1×10^5, 0.38 GW/m^2, 0.5–1.0 ms, and 3.2×10^{24}–4.1×10^{25} m^{-2}, respectively. Because the repetition frequency of laser irradiation was 10 Hz for the tests to 1×10^4 and 1×10^5 pulses and 0.5 Hz for the tests to 1×10^3 pulses, the D-plasma fluence of the tests to 1×10^3 pulses (6.6×10^{24} m^{-2}) was close to that of the tests to 1×10^4 pulses (3.2×10^{24} m^{-2}). As post-mortem analyses to evaluate the degradation caused by the thermal shock tests, the observation of the heat loaded surface using a scanning electron microscope (SEM) and profilometry to measure the arithmetic mean surface

roughness, R_a, of the heat loaded surface were carried out.

3. Results and discussion

As shown in Fig. 1, elongated grains along rolling (L) direction were observed in both materials. Alloying by 3%Ta induced grain refining; the grain sizes of pure W and W-3%Ta along L and S directions in the as-received condition were 106 and 28 μm and 53 and 13 μm, respectively, which might be induced by the inhibiting the motion of grain boundaries by solute Ta. According to the previous studies [6, 7, 9, 10, 14], it was expected that the grain refining could lead to strengthening and toughening of W. In addition to the grain refining, the alloying by 3%Ta induced approximately 10% increase in Vickers hardness in the as-received condition and approximately 400 °C increase in recrystallization temperature due to isochronal heat treatment for 1 h, which might be attributed to the solid solution strengthening and hindering the motion of grain boundaries and dislocations by solute Ta, respectively (see Fig. 2 [7, 10]). The recrystallization temperature was defined as the temperature, where a steep reduction of Vickers hardness occurred, after the microstructural recovery stage with a slight reduction in the hardness. Since the recrystallization temperature of W-3%Re was 1500 °C, which was produced by the same fabrication route [6, 7, 10], the effect of solute Ta on the resistance to recrystallization was higher compared to the solute Re.

As shown in Fig. 3 (a) [7, 9, 10], the increase in ultimate tensile strength (UTS) due to the alloying by 3%Ta was observed at all temperatures for both L and T directions, which might be induced by the grain refinement and solid solution strengthening by Ta. In contrast to the UTS, the total elongation (TE) along L direction of W-3%Ta was similar to that of pure W below 1100 °C, as shown in Fig. 3 (b) [7, 9, 10]. The increase in UTS and no change of TE below 1100 °C might indicate a higher fracture toughness of W-3%Ta compared to the pure W. Above 1100 °C, TE along L direction of pure W was significantly higher than that of W-3%Ta, which might be attributed to the recrystallization of pure W. Thus, this could not show the improved mechanical property of pure W. In contrast to L direction, W-3%Ta along T direction below 500 °C showed higher TE compared to pure W, which could indicate the improved low temperature ductility of W-3%Ta. The difference of UTS and TE between L and T directions was clear below 300–600 °C, which might be attributed to the microstructural anisotropy in the as-received condition caused by the unidirectional hot-rolling. Because the cleavage fracture was dominant at such low temperatures and the cleavage fracture stress was dependent on the grain structure (e.g., grain size), it was considered that the difference in the cleavage fracture stress along each direction might induce the anisotropy of tensile properties at relatively low temperatures [7, 9, 10].

As for the Charpy impact properties, hot-rolled W materials in the present study showed

better performance compared to the forged round-blank materials in the previous studies, regardless of Ta concentration, as shown in Fig. 4 [7, 9, 10, 15]. This meant that the mechanical properties could be improved even if the major chemical composition was the same. As for the materials examined in the present study, the upper shelf energy (USE) and DBTT were slightly improved due to the alloying by 3%Ta, which might be induced by the grain refinment and solid solution strengthening by Ta. This tendency was consistent with the above-mentioned tensile property. Since the DBTT of W-3%Re was 100 °C lower compared to pure W, the effect of solute Ta on DBTT was not significantly inferior to the solute Re [6, 7, 10]. Based on these evaluations, W-3%Ta might be one of the competitive alloy systems from the viewpoint of fundamental mechanical properties. Further investigations on the effect of Ta concentration, microstructural anisotropy, and microstructural change due to high temperature heat treatment (microstructural recovery, recrystallization, and grain growth) are planned as future works to optimize the W-Ta material.

In addition to the improved fundamental mechanical properties, the degradation of W-3%Ta caused by the thermal shock tests under the background steady state D-plasma exposure was suppressed compared to pure W. As shown in Fig. 5, after the laser irradiation to 1×10^3 pulses with D-plasma fluence of 6.6×10^{24} m^{-2}, very small change of surface morphology was observed in pure W, while W-3%Ta showed no change. In this test condition, difference in surface degradation was not quantitatively distinguished by surface roughness measurement (R_a = 0.4 μm for both materials). After the laser irradiation to 1×10^4 pulses with D-plasma fluence of 3.2×10^{24} m^{-2}, surface morphology change and increase in surface roughness (R_a = 0.5 μm) occurred in pure W, while W-3%Ta showed very small change of surface morphology, which resulted in the no increase in R_a value. After the laser irradiation to 1×10^5 pulses with D-plasma fluence of 4.1×10^{25} m^{-2}, not only surface morphology change but cracking were clearly observed in both materials. As well as the other test conditions, the surface degradation was suppressed in W-3%Ta compared to pure W. The R_a values of W-3%Ta and pure W were 4.64 and 16.1 μm, respectively. The area showing surface degradation was also suppressed in W-3%Ta compared to pure W. The diameter of degraded area of pure W was close to the laser beam diameter (approximately 3 mm).

Specimens irradiated to 1×10^5 pulses showed an isotropic circular shape surface degradation despite the anisotropic microstructure in the as-received condition. According to the previous study [16], where similar thermal shock tests were performed, it was expected that the thermal shock tests could lead to a temperature increase of around 700 °C during each pulse. This meant that both materials, especially pure W, might be brought to a certain level of recrystallization if considering the base temperature (700 °C), total heat-loaded time (approximately 3 h), and Vickers hardness change by 1 h heat treatment (see Fig. 2). Because the recrystallization could induce isotropic microstructure with equiaxial grains, the isotropic circular

shape surface degradation was likely. In addition, anisotropy of tensile strength and elongation was not observed above the base temperature of thermal shock tests (see Figs. 3 (a) and (b)). Thus, it was expected that the isotropic circular shape surface degradation might be caused by no anisotropy of mechanical properties and recrystallization-induced isotropic microstructure.

Although it was concerned that the effect of D-plasma exposure might be much more pronounced in the W-3%Ta, no such phenomena under the thermal shock tests were observed within the post-mortem analyses of the present study. Fig. 6 shows the surface roughness, R_a, of pure W and W-3%Ta in the present study and the previously reported surface roughness, R_a, of five kinds of W materials by Nogami *et al*. [17] and four kinds of W materials by Pintsuk *et al*. [18], which were obtained by the thermal shock tests to 1×10^3 pulses with no D-plasma exposure using an electron beam irradiation device, JUDITH 1, at Forschungszentrum Jülich [19]. If comparing among the pure W materials obtained by the thermal shock tests to 1×10^3 pulses (shown by black color in Fig. 6), the effect of D-plasma exposure could not be clearly observed. According to the thermal desorption experiments by Schmid *et al*. [20], it was possible that D could be desorbed from both pure W and W-Ta alloy below 700 °C. Therefore, one of the possible reasons for no clear effect of D-plasma exposure was the desorption of D during the thermal shock tests above 700 °C. In addition, negligible changes of tensile strength and elongation due to the D-exposure were observed by the tensile tests at 200 and 300 °C, as shown in Figs. 3 (c) and (d). Therefore, another possible reason for no clear effect of D-plasma exposure was that the effects of D on the mechanical properties of pure W and W-3%Ta were negligible. Therefore, the difference in the evolution of surface degradation between pure W and W-3%Ta could be mainly attributed to their mechanical properties in the as-received and recrystallized conditions. Based on these evaluations, W-3%Ta might be one of the promising alloy systems, even if the thermo-mechanical response under heat load environment in the actual PFMs was considered. Further investigation of such thermo-mechanical response and the optimization of W-Ta material based on those evaluations are planned as future works.

4. Conclusion

The degradation under cyclic thermal shock tests with background steady state D-plasma exposure of W-3%Ta alloy and pure W and their fundamental mechanical properties were investigated. The main results were summarized as follows:

1) W-3%Ta showed smaller grains, higher recrystallization temperature, higher strength, slight improvement of USE and DBTT compared to pure W.
2) Degradation of W-3%Ta by the thermal shock tests under the background steady state D-plasma exposure was suppressed compared to pure W. Although it was concerned that the

effect of D-plasma exposure might be much more pronounced in the W-3%Ta, it was not clearly observed in the present study.

3) Based on the present study, W-3%Ta might be a competitive alloy system from the viewpoint of thermo-mechanical response under heat load environment in the actual PFMs as well as the fundamental mechanical properties and resistance to recrystallization.

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Fig. 1 Metallographic images (L × S surface) of pure W and W-3%Ta in as-received condition.

Double column figure

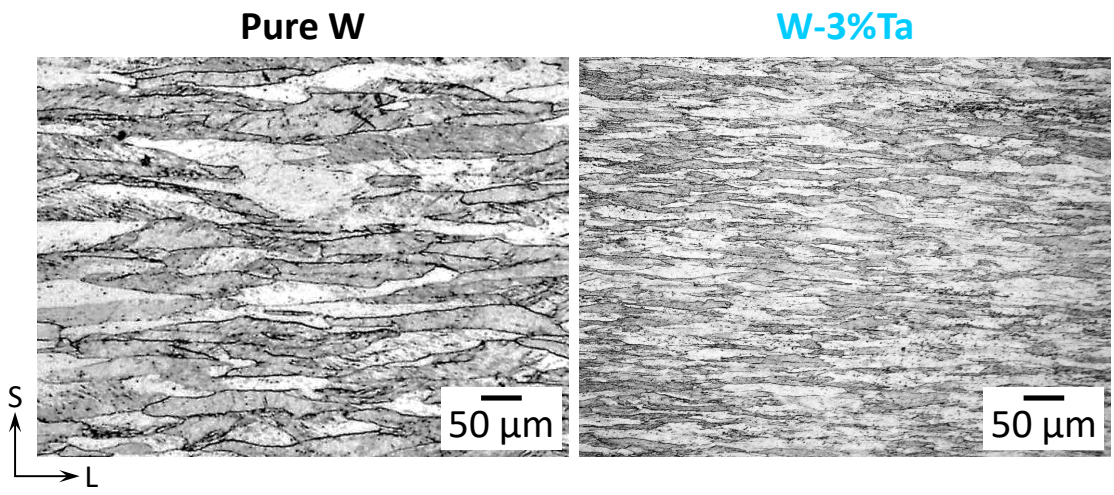

Fig. 2 Heat treatment temperature dependences of Vickers hardness measured on L × S surfaces of pure W [7, 10] and W-3%Ta. Heat treatment time was 1 h.

Single column figure

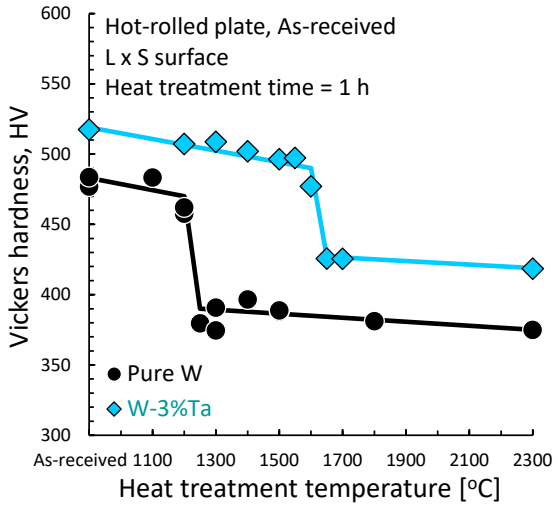

Fig. 3 Test temperature dependences of (left) UTS and (right) TE by tensile tests along L and T directions of pure W [7, 9, 10] and W-3%Ta (a, b) in as-received condition and (c, d) after D-exposure (Low-D: 0.8 kPa × 6 h, High-D: 80 kPa × 24 h).

Double column figure

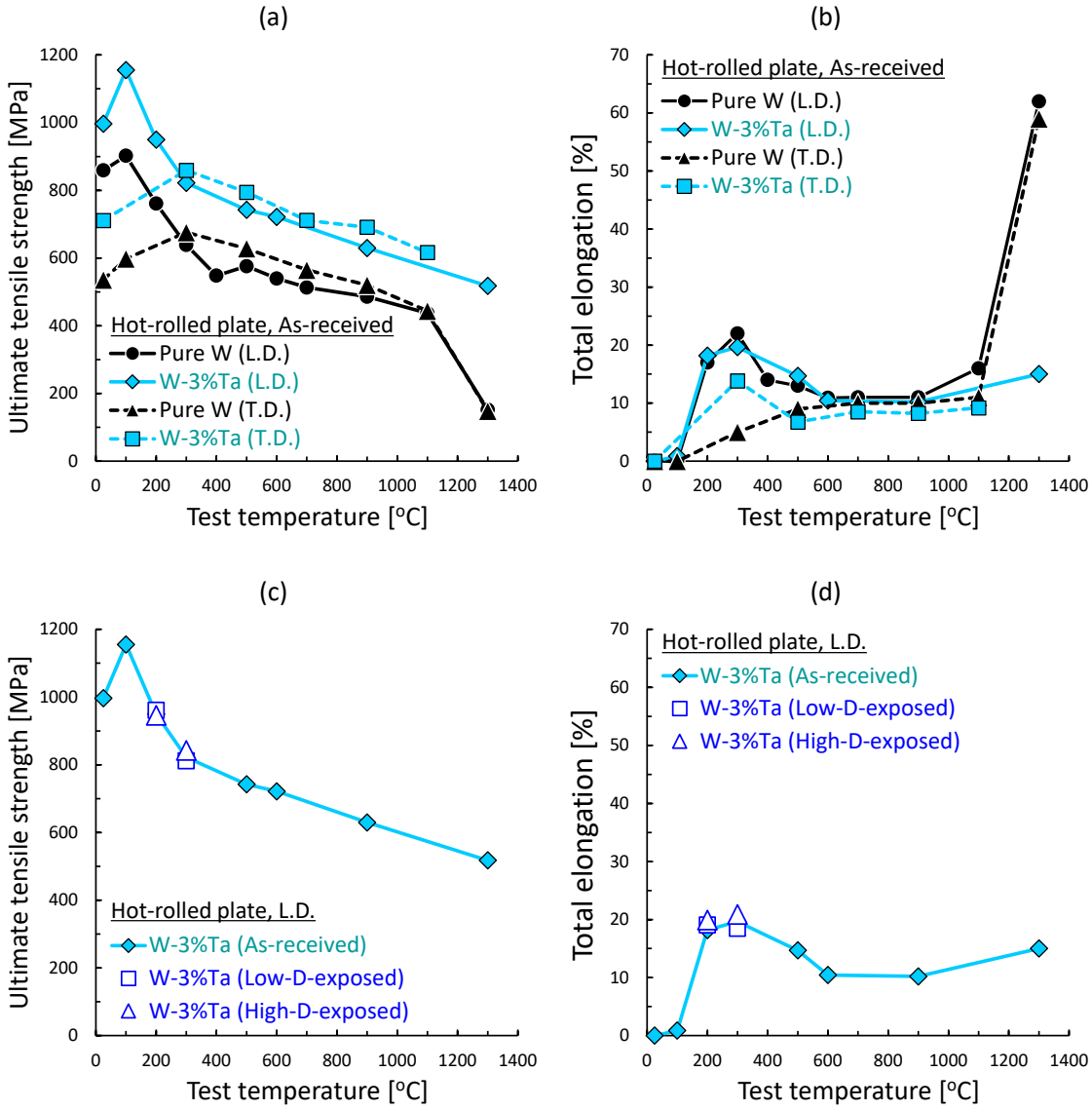

Fig. 4 Test temperature dependences of absorbed energy from Charpy impact tests along L-S direction of pure W [7, 9, 10] and W-3%Ta in as-received condition. Previously reported data on pure W, W-1%Ta, and W-5%Ta forged round-blanks by Rieth *et al*. [15] are also plotted.

Single column figure

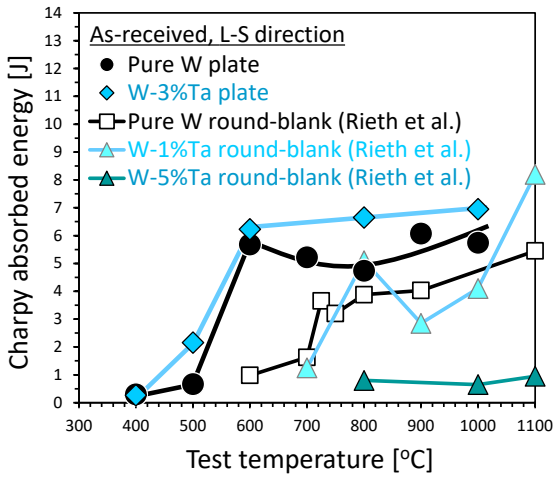

Fig. 5 Surface images (L × T surface) obtained by an SEM of pure W and W-3%Ta after thermal shock tests with background steady state D-plasma exposure. Arithmetic mean surface roughness, R_a, after the tests are shown in each image.

Double column figure

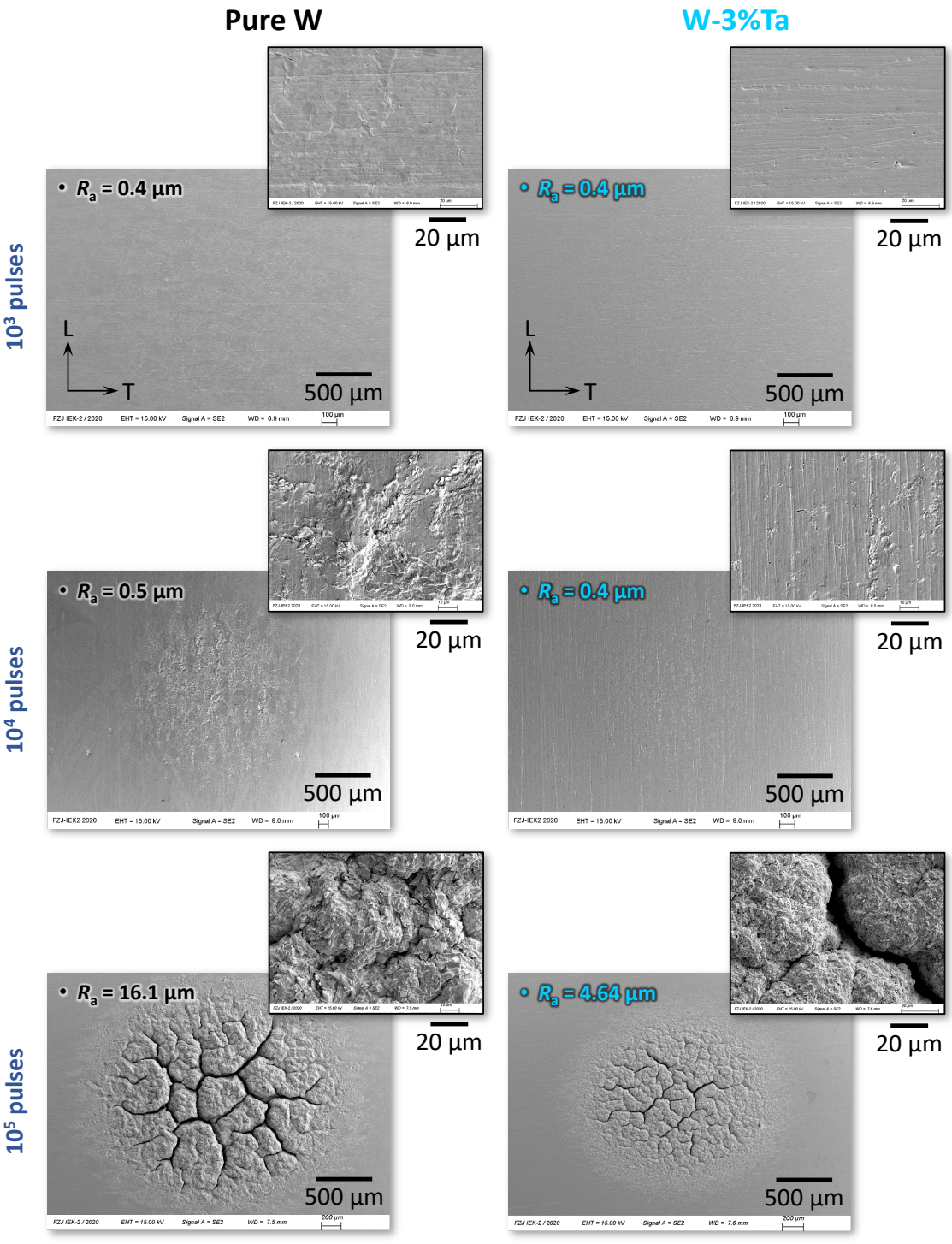

Fig. 6 Arithmetic mean surface roughness, R_a, produced by thermal shock tests with background steady state D-plasma exposure of pure W and W-3%Ta. Previously reported surface roughness, R_a, of five kinds of W materials by Nogami *et al*. [17] and four kinds of W materials by Pintsuk *et al*. [18] are also plotted.

Single column figure