Thermal properties of pure tungsten and its alloys for fusion applications

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ABSTRACT

Tungsten is a promising candidate for plasma-facing materials in a fusion reactor, and several research studies have been conducted to improve the mechanical properties of pure W. However, the thermal properties are also important characteristics of plasma-facing materials. In this study, the thermal properties of pure W and its alloys were measured, and the effect of alloying on the thermal properties of pure W was investigated. Potassium-bubble (K-bubble) dispersion, which is one of the major methods utilized to improve the mechanical properties of W, did not affect the thermal diffusivity and conductivity of pure W. On the other hand, the presence of rhenium, which is major alloying element of W, affected the absolute values and the temperature dependence of thermal diffusivity and conductivity. The effect of alloying on specific heat and the anisotropy in thermal diffusivity of pure W and its alloys were also investigated. Measurements of the specific heat showed that K-bubble dispersion and Re addition had insignificant effects on the values obtained for pure W. Anisotropy in thermal diffusivity was not observed, and the effect of anisotropic grain structure and alloying was insignificant.

1. Introduction

Tungsten has attracted much attention as a plasma-facing material (PFM) for in-vessel components such as divertors and blankets in a fusion reactor because of its high melting temperature, high thermal conductivity, low tritium retention, and low sputtering yield. However, its inherent brittleness at low temperature and the embrittlement due to recrystallization and neutron irradiation are major concerns regarding the use of W in fusion reactor applications. To overcome these drawbacks, several efforts have been made to modify W through grain refining, alloying, dispersion of secondary phases, and formation of composites [1–7]. One of the important functions of the divertor in a fusion reactor is the removal of high heat flux. When used as a PFM, W is exposed to high heat loads that are typically ~10 MW/m^2 in the steady state and > 20 MW/m^2 in the non-steady state; the thermal properties of W play an important role in maintaining the integrity of the material itself as well as the reactor component. Although the modifications mentioned above can improve the mechanical properties of W, these changes may also lead to degradation of the thermal properties. For example, it is well known that a higher rolling ratio improves the ductility of a W plate or sheet. However, Zhang et al. measured the thermal conductivity of pure W prepared with different rolling-reduction rates, and lower thermal conductivities were obtained in pure W samples that were subjected to higher and lower rolling-

reduction rates [8]; they reported that the decrease in thermal conductivity was probably due to the presence of large defects such as pores and cracks, as well as microscale defects such as dislocations and boundaries [8].

We evaluated the material properties of W alloys in our previous studies, and we reported that W with a small amount of added rhenium or dispersed potassium bubbles (K-bubbles), or both Re and K-bubbles, showed higher grain structure stability and better mechanical properties than pure W [9,10]. The objective of this study is to evaluate the thermal properties of pure W and its alloys, and to investigate the effect of material modification (i.e., secondary-phase dispersion and Re addition) on the thermal properties of pure W.

2. Experimental

The materials examined in this study were pure W, K-doped W, W–1%Re, W–3%Re, K-doped W–3%Re, and La-doped W–3%Re (the percentages represent the nominal mass ratio of Re in the alloys). Both single-crystalline and polycrystalline materials were used for pure W. The single-crystalline pure W used in this study was prepared by secondary-recrystallization heat treatment of polycrystalline pure W. The dimensions of the examined original plates were ~85 × ~80 × 5 mm, where 5 mm corresponds to the plate thickness. Detailed descriptions of the materials, such as the fabrication process and chemical

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composition, are provided in the literature [9,10]. The W–1%Re and La-doped W–3%Re were fabricated by the same process as other polycrystalline materials. The reduction ratios of these materials were 80 and 60%, respectively. The final heat-treatment temperature of materials except single-crystalline pure W was 900 °C for 20 min. The thermal diffusivity and specific heat of pure W and its alloys were measured in this study to investigate the thermal properties; the thermal conductivity was calculated using the densities of materials obtained from the literature and the measured values of thermal diffusivity and specific heat.

The thermal diffusivity was measured with a laser flash apparatus, LFA-457 MicroFlash (NETZSCH GmbH, Germany), and the values between room temperature (RT) and 1080 °C were measured in an Ar atmosphere; three measurements were obtained at each temperature. Pulse-length correction and non-linear regression for the Cowan fit were applied to correct the raw data. Disk-shaped (10 mm (ϕ) × 2 mm (t)) and plate-shaped (5 mm (l) × 5 mm (l) × 1 mm(t)) samples were prepared for these measurements; the plate-shaped samples were used only for investigation of the anisotropic thermal diffusivity owing to the limitation of the original plate thickness (5 mm). The specific heat was measured by using two differential scanning calorimeters, DSC 204 F1 Phoenix (NETZSCH GmbH, Germany) and STA 449 F3 Jupiter (NETZSCH GmbH, Germany), for the low-temperature (10–100 °C) and high-temperature (100–1400 °C) measurements, respectively, in an Ar atmosphere. The heating rates for the low- and high-temperature measurements were 10 and 20 °C/min, respectively, and disk-shaped (5 mm (ϕ) × 1 mm (t)) samples were used.

The specimens for the measurement of thermal diffusivity and specific heat were cut by electro-discharge machining from the plate materials, and the surface of each specimen was mechanically polished with up to #3000 mesh before the measurements. To analyze the effect of microstructure on the thermal properties, the microstructure of the selected materials was observed using a digital microscope (VHX-200, Keyence Corp., Japan) and a scanning transmission electron microscope (STEM, JEM-ARM200F, JEOL Ltd., Japan).

3. Results

Fig. 1 shows the temperature dependence of the thermal diffusivity in pure W and its doped W–Re alloys (i.e., K-doped W, K-doped W–3% Re, and La-doped W–3%Re), measured using disk-shaped samples (10 mm (ϕ), 2 mm (t)) cut from the RD–TD plane (RD: rolling direction; TD: transverse direction) of the plate materials. The thermal diffusivity of the examined materials decreased with increasing temperature, and this trend was especially prominent in pure W and K-doped W: pure W (both single-crystalline and polycrystalline samples) and K-doped W
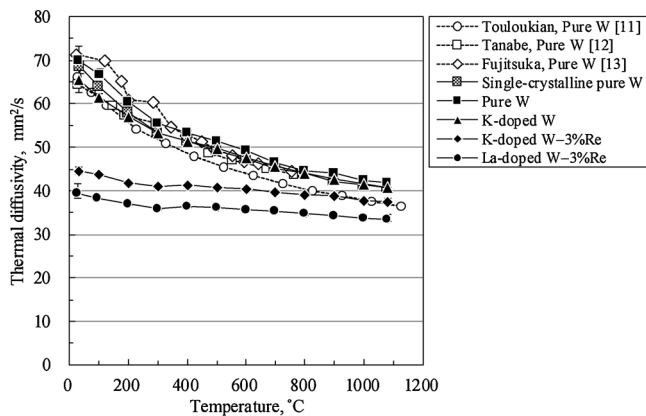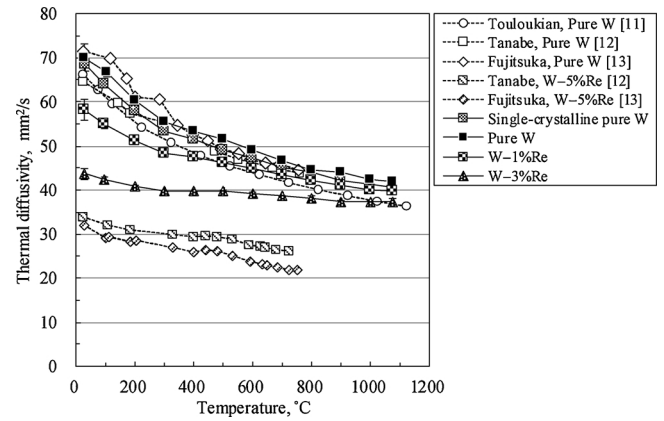
Fig. 2. Measured thermal diffusivity of pure W and non-doped W–Re alloys. The values of thermal diffusivity reported in the literature are also shown in this figure [11–13].

showed almost the same thermal diffusivity in the temperature range of RT–1080 °C; K-doped W–3%Re and La-doped W–3%Re showed lower thermal diffusivity than other samples, especially in the intermediate temperature region (below ~800 °C). The temperature dependence of thermal diffusivity was insignificant in these materials.

Fig. 2 shows the thermal diffusivity of pure W and non-doped W–Re alloys in the temperature range of RT–1080 °C. It can be clearly seen that the thermal diffusivity decreased with increasing Re concentration, especially at temperatures below ~800 °C. W–1%Re and W–3%Re showed values that were approximately 18 and 23% lower than that of pure W at 100 °C, although these differences were less than 10% at temperatures above 900 °C, as shown in Fig. 3.

Fig. 4 shows the results of specific heat measurements for pure W and its alloys. Pure W showed relatively lower specific heat at elevated temperatures (> 700 °C), although the difference in values was less than 3% and there was no significant difference among the examined materials. The reference specific heat increased with increasing temperature, although the measured values decreased at approximately 900 °C. The thermal conductivity of the examined materials was calculated by using the following equation:

$$\lambda = \alpha \cdot c_{\mathrm{p}} \cdot \rho,$$

where α and c_{p} are the thermal diffusivity and the specific heat, respectively. The values of density, ρ, reported in the literature were used to calculate the thermal conductivity at each temperature [14]. The thermal conductivities calculated for pure W and K-doped W were
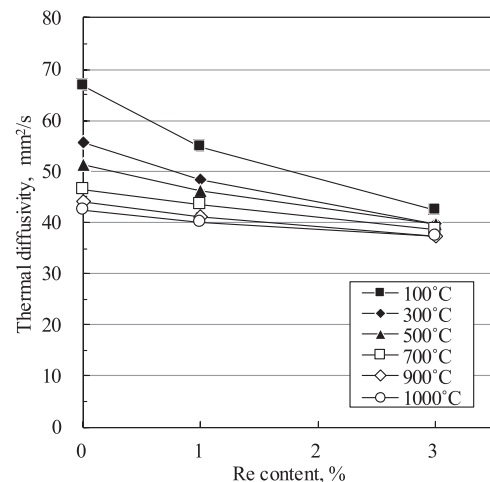
Fig. 1. Measured thermal diffusivity of pure W and its doped W–Re alloys. The values of thermal diffusivity reported in the literature are also shown in this figure [11–13].

Fig. 3. Effect of Re addition on thermal diffusivity of pure W at selected temperatures.
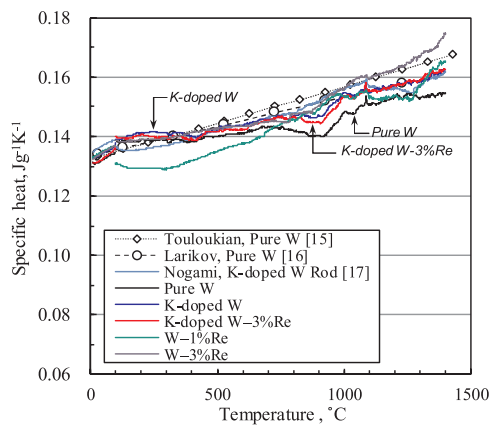
Fig. 4. Specific heat of polycrystalline pure W, K-doped W, W–3%Re, and K-doped W–3%Re in the temperature range from RT to 1400 °C. The values of specific heat reported in the literature are also shown in this figure [15–17].
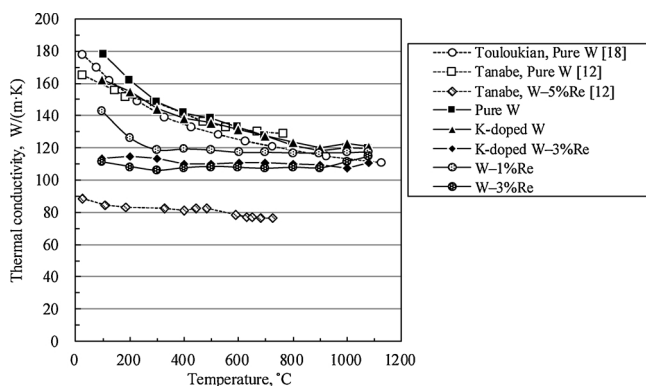

Fig. 5. Thermal conductivity of pure W, K-doped W, W–3%Re, and K-doped W–3%Re. The values of thermal conductivity reported in the literature are also shown in this figure [12,18].

almost the same up to ∼1100 °C, as shown in Fig. 5. K-doped W–3%Re showed lower thermal conductivity than pure W and K-doped W, and the temperature dependence was not obvious when compared with that exhibited by pure W and K-doped W. The thermal conductivity of K-doped W–3%Re was almost constant at ∼110 W/(m K).

It is well known that tungsten plates, sheets, and rods have aniso-tropic grain structures [17,19–21]. The degree of anisotropy in the grain structure depends on the fabrication method and process, re-duction rate, etc., and as shown in Fig. 6, the materials examined in this study also exhibited anisotropic grain structures. To evaluate the effect of anisotropic grain structures on the thermal properties of the ex-amined materials, the thermal diffusivity was measured using plate-shaped samples (5 mm (l) × 5 mm (l) × 1 mm (t)) cut along three planes in the plate materials (ND–TD, ND–RD, and RD–TD planes in Fig. 6; ND: normal direction).

The measured aspect ratio of the grains ranged from ∼1.3 to 5.5, as shown in Table 1. The materials with the highest (pure W and K-doped W) and intermediate (W–1%Re) grain aspect ratios were examined in detail, and the results are shown in Fig. 7. There were no obvious dif-ferences in the thermal diffusivity and no clear trend for dependence of the thermal diffusivity of each material on the temperature and mea-sured plane. In addition, anisotropic thermal diffusivity was not ob-served. It should also be noted that the thermal diffusivity values shown in Fig. 7 are lower than those in Figs. 1 and 2.

4. Discussion

The values of thermal diffusivity of pure W reported in the literature

are also plotted in Figs. 1 and 2. The thermal diffusivities measured in this study for single-crystalline pure W, polycrystalline pure W, and K-doped W are in the scattering range of the reference values, and there was no obvious difference between the measured thermal diffusivities of single-crystalline and polycrystalline pure W. Microstructure ob-servation showed that the average grain size of polycrystalline pure W was 52 μm, and sub-grain boundaries as well as dislocations were ob-served, as shown in Figs. 6 and 8. The single-crystalline pure W was prepared by secondary-recrystallization heat treatment, and this ma-terial was almost free of boundaries and dislocations. Therefore, these results suggest that the boundaries, including both grain and sub-grain boundaries, and defects such as dislocations in the examined poly-crystalline pure W did not have large effects on the thermal diffusivity.

Changes in the thermal diffusivity, such as decreased values and reduced temperature dependence, were observed only in the case of Re-added alloys. As shown in Figs. 1 and 2, W–1%Re showed lower thermal diffusivity than pure W (both measured and reference values). The thermal diffusivity of the alloy with 3% Re was lower than those of pure W and W–1%Re and higher than that of W–5%Re. The solubility limit of Re is > 20% at temperatures above ∼300 °C [22] because Re atoms in these Re-added alloys may have been in the solid-solution state. Since the thermal diffusivity of W–Re alloys has been reported to be below that of pure W [12,13], the solute Re atoms may act as scattering centers of electrons and phonons and contribute to heat transfer. Tanabe et al. reported the thermal diffusivity and thermal conductivity of pure W and W–Re, and they calculated the contribution of electrons and phonons to thermal conductivity [12]. Their results showed that both electrons and phonons contribute to the thermal conductivity of pure W and W–Re alloys, although the electrons made higher contributions than phonons in W–Re alloys. The scattering of electrons by solute Re may have been one of the major reasons that the thermal diffusivity of Re-added W was lower than that of pure W. Al-though this assumption agrees with the measured values of electrical resistivity reported by Tanno et al. [23], the contribution of other sources, such as phonons, to changes in W–Re alloys cannot be excluded in this study. The difference between the temperature dependence of pure W and Re-added W may have been caused by the changes in the magnitude of scattering by solute Re and the kinetics of heat transfer.

The other possible factor of the decrease in thermal diffusivity of W was the presence of dispersed secondary phases such as K-bubbles and La_2O_3 in K-doped W, K-doped W–3%Re, and La-doped W–3%Re. The difference between the thermal diffusivity of pure W and K-doped W was small, and these materials showed almost the same thermal diffu-sivity values in the examined temperature range because K-bubbles did not appear to have a significant effect on the thermal diffusivity of pure W in this study. In W with dispersed K-bubbles, only small amounts of elements such as Al, K, and Si were added to form and disperse K-bubbles, and the result of chemical analysis showed that 30 ppm (by mass) of K and less than 20 ppm (by mass) of Al and Si were present on the final plate material. One of the expected roles of K-bubbles was to help multiply dislocations during fabrication processes such as rolling and forging, and to form a finer grain structure than that in pure W. The average grain size of the examined K-doped W was 20 μm, which is ∼60% smaller than the grains in pure W, although this difference is not too big and the results may not have a large influence on the thermal diffusivity of pure W. In addition, the densities of pure W and K-doped W were 99.0 and 99.1%, respectively, which are almost the same. Therefore, it was possible that the elements added to disperse K-bubbles and the K-bubbles themselves did not have a significant effect on the thermal diffusivity and thermal conductivity of W. For the case of K-doped W–3%Re, the decrease in thermal diffusivity was mainly caused by the presence of 3% Re; therefore, the thermal diffusivity of this material was almost the same as that of W–3%Re.

La-doped W–3%Re showed lower thermal diffusivity than W–3%Re, suggesting factors other than the presence of 3% Re contributed to the decrease. La-doped W–3%Re contained 1.07 mass% of La, 0.178 mass%
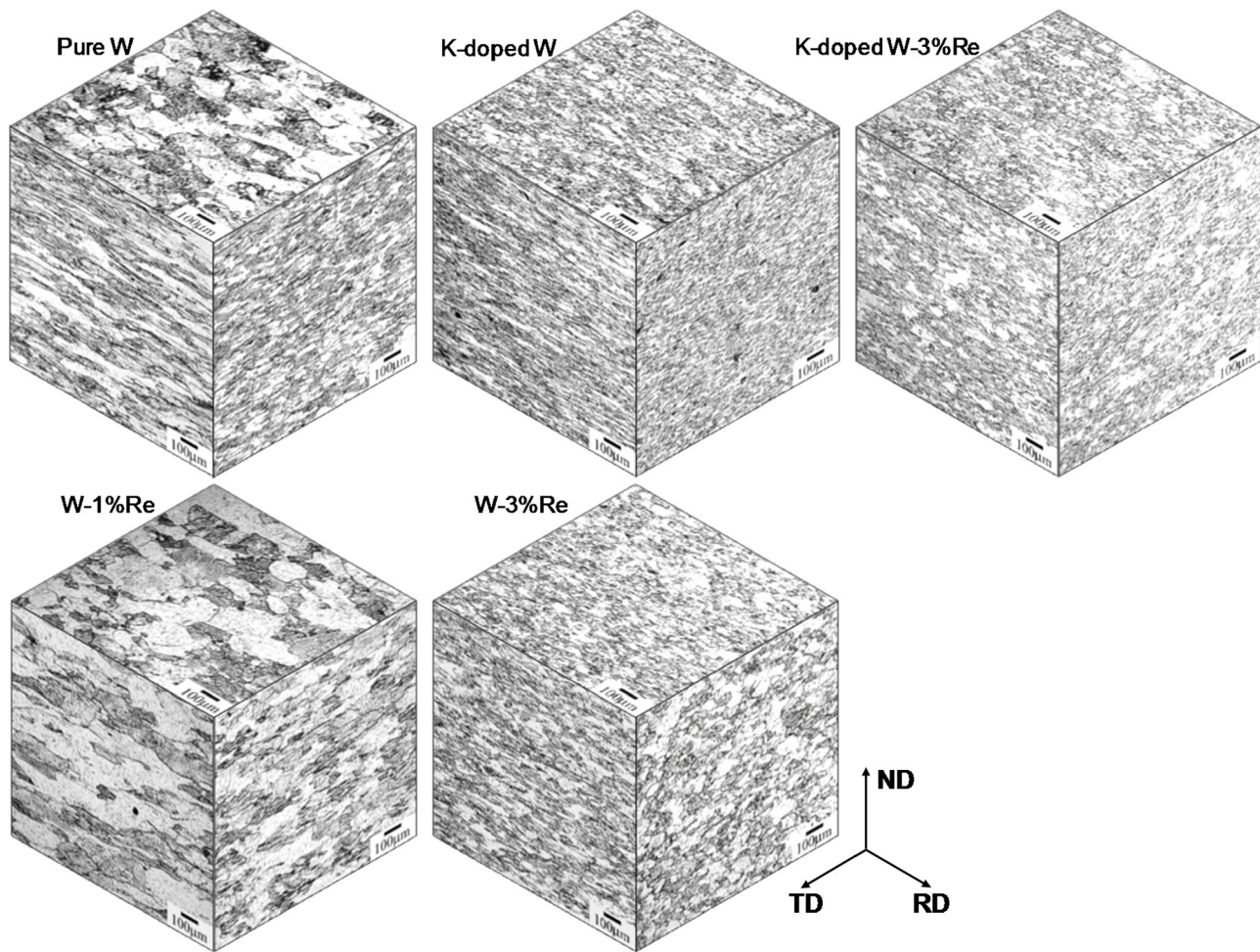
Fig. 6. Grain structure of examined materials; the normal direction (ND), rolling direction (RD), and transverse direction (TD) are shown. The grain structure of La-doped W–3%Re was not obtained owing to the difficulties encountered during electro-polishing and etching.

Table 1
Grain size of materials on the ND–RD plane, measured from the linear intercept. RD and ND correspond to the horizontal and vertical axes, respectively, of the measurement lines.

Material	Grain size, μm		Grain aspect ratio (RD/ND)
	RD	ND	
Pure W	109	20	5.5
K-doped W	38	7	5.4
K-doped W–3%Re	37	20	1.9
W–1%Re	113	57	2.0
W–3%Re	35	27	1.3

of O, and 2.9 mass% of Re. It was highly possible that La and O, which did not form La_2O_3, were present in the matrix, and they prevented heat transfer, resulting in a decrease in the thermal diffusivity.

The measurement of specific heat showed almost the same value in all of the examined materials, and the effects of Re addition and K-bubble dispersion were insignificant. The recovery of dislocations in W would have begun around 800–900 °C [24] because one of the possible causes was the recovery of the residual strain induced by hot-rolling or mechanical polishing. The magnitude of the decrease in specific heat at approximately 900 °C was relatively small and would not have had any obvious effect on the thermal conductivity.

The temperature dependence of thermal conductivity in pure W and its alloys showed almost the same trend as that displayed by thermal diffusivity, which remained almost constant in the measurement-temperature range. Since there was no observable difference between the measured and reference thermal conductivity, it can be concluded that K-bubble dispersion did not affect the thermal properties of W. The thermal conductivity of K-doped W–3%Re was lower than that of pure W and higher than that of W–5%Re. The difference between the thermal conductivity of K-doped W–3%Re and other samples (pure W and K-doped W) may have been caused by the addition of 3 mass% Re for the reason mentioned above. The thermal conductivity of W–3%Re was almost the same as that of K-doped W–3%Re, while W–1%Re showed higher thermal conductivity than W–3%Re and K-doped W–3%Re, which reflect the trend shown by the thermal diffusivity.

The anisotropy in thermal diffusivity was insignificant in materials with both high and intermediate aspect ratios of grains. The anisotropic electrical resistivity of pure W, which is related to thermal conductivity, was reported to depend on the crystal orientation [25], whereas poly-crystalline pure W consists of grains with different orientations, which may have resulted in the isotropic thermal diffusivity observed in this study. The lower thermal diffusivity in Fig. 7 compared with the results shown in Figs. 1 and 2 may have been caused by the different dimensions of the specimens. The lower volume and thickness of the smaller specimens caused shorter half-times in the thermal-diffusivity measurements, which was one of the major causes of the decrease in thermal diffusivity values. Therefore, the results of small specimens (5 mm (l) × 5 mm (l) × 1 mm (t)) were used only for comparing the thermal diffusivity on each plane of the materials.
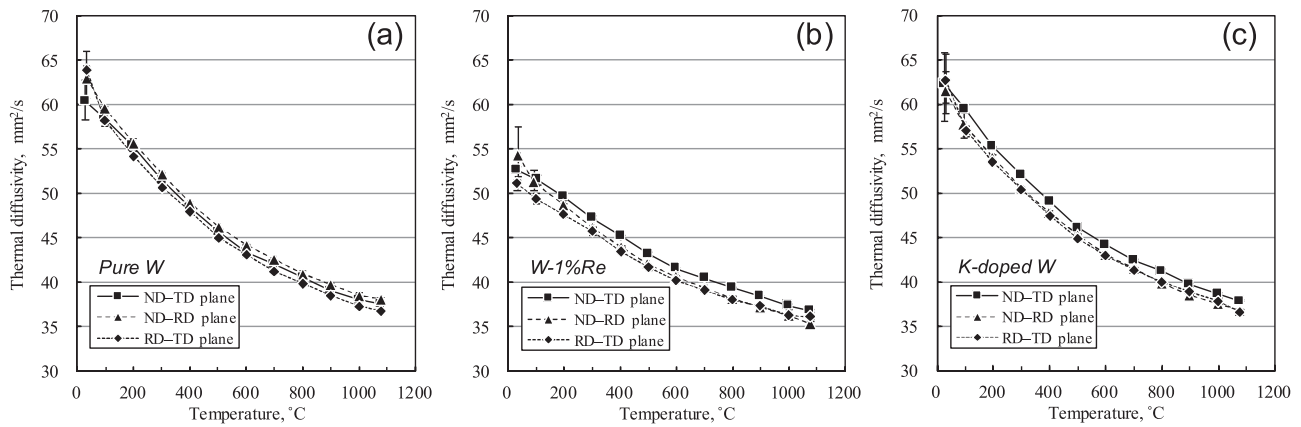
Fig. 7. Thermal diffusivity of (a) pure W, (b) W–1%Re, and (c) K-doped W on each plane of the plate materials.
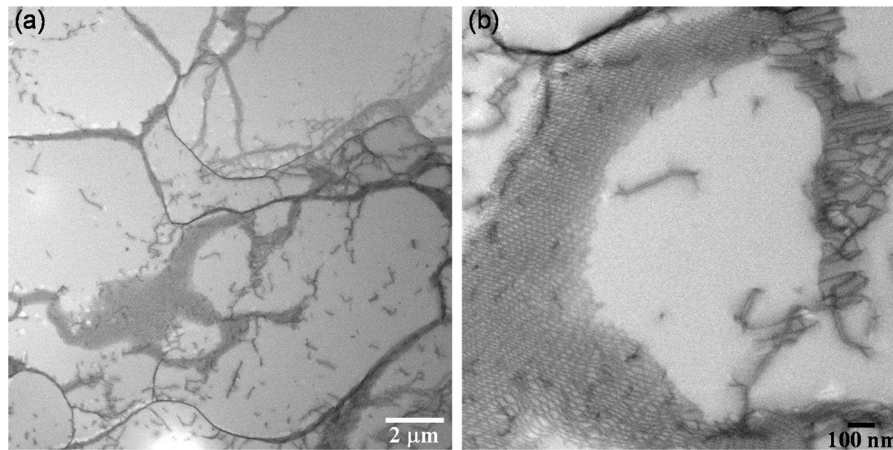

Fig. 8. Scanning transmission electron microscope (STEM) images of pure W examined in this study at (a) low and (b) high magnification. The black lines show the dislocations and boundaries.

5. Summary

The thermal properties of pure W and W alloys were investigated in this study. The obtained results are summarized as follows:

- The thermal diffusivities of single-crystalline and polycrystalline pure W were almost the same, and the difference in microstructure (i.e., grain boundaries and dislocation densities) of the examined pure W samples did not affect the thermal diffusivity.
- K-bubble dispersion did not have an obvious effect on the thermal properties of pure W, whereas the addition of 3 mass% of Re changed the thermal diffusivity and its dependence on temperature. La-doped W–3%Re showed the lowest thermal diffusivity among the examined materials in this study, although the temperature dependence was the same as that exhibited by W–3%Re and K-doped W–3%Re.
- The thermal diffusivity of W decreased with increasing Re concentration and became less sensitive to temperature changes. La-doped W–3%Re exhibited the lowest thermal diffusivity probably because it had a higher concentration of La and O in the matrix than the other examined materials, which caused an additional decrease in the thermal diffusivity.
- Anisotropy in thermal diffusivity was not observed in pure W, K-doped W, and W–1%Re, suggesting that the anisotropic grain structure in the examined materials did not have an obvious effect on the thermal diffusivity.

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