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論文内容要約

The plasma facing materials (PFMs) which are used on plasma facing surface of the plasma facing components (PFCs) such as divertor and blanket will be subjected to high heat load and high energy neutron irradiation. Tungsten (W) is the most attractive material for PFM because of its high melting temperature, thermal conductivity, and sputtering resistance. The results of the cyclic heat load experiment using mock-ups of W divertor for ITER (International Thermonuclear Experimental Reactor) to evaluate their performance had been reported. These results showed the crack formation and loss of cooling capability of W during cyclic heat load. Heat removal is an important function of divertor, because, damage by the crack formation and the resulting loss of cooling ability should be suppressed to improve the lifetime and reliability of divertor. To avoid the loss of cooling ability by crack formation and propagation, improvement of the mechanical property in W is desired. The crack formation and propagation behavior will be affected by recrystallization of the grain structure in W. The recrystallization decreases the mechanical properties of W and this phenomenon is called recrystallization embrittlement. The recrystallization embrittlement will accelerate the crack formation and propagation during cyclic heat load. In order to improve the lifetime and reliability of W divertor, recrystallization embrittlement should be suppressed. During the operation of a fusion reactor, W will be exposed to not only high heat load but also neutron irradiation. The neutron irradiation changes the mechanical property, increases yield stress and ultimate tensile strength (UTS), and decreases elongation. This phenomenon is called irradiation embrittlement. Thus, irradiation embrittlement will also accelerate the crack formation and propagation in W as well as recrystallization embrittlement. The objective of this study is the development of the W alloy for PFCs in fusion reactor which has resistance to recrystallization embrittlement and irradiation embrittlement.

In this study, W alloys were designed based on experimental results and data from literatures. To design W alloys, investigation of the neutron irradiation effects of pure W and its alloys was performed. It was because of that the method to suppress irradiation embrittlement and the irradiation effects on W alloys are not well understood. The neutron irradiation was conducted in the Joyo in Japan Atomic Energy Agency-Oarai, Ibaraki, Japan and the High Flux Isotope Reactor (HFIR) in Oak Ridge National Laboratory (ORNL), TN, USA. Irradiation temperature and displacement damage (dpa) were in the range of 531–800 °C and 0.42–0.98 dpa, respectively. Following the irradiation, microstructure observation using transmission electron microscope (TEM) and Vickers hardness measurement were conducted. The neutron-irradiation behavior of pure W and W alloys was summarized based on these results and our previous data, and W alloys for PFC were designed and fabricated by powder metallurgy and hot rolling. Materials fabricated and examined in this study were pure W and its alloys (e.g. potassium (K) -doped W and K-doped W-3% Rhenium (Re)).

In order to evaluate the anisotropy of grain structure and the recrystallization behavior, the grain structure observation and Vickers hardness measurement were conducted using as-received and heat treated specimens. Heat treatment temperature and dwell time were 1100–2000 °C and 1 h, respectively. To evaluate the mechanical property and its anisotropy, tensile tests was conducted using as-received specimen at R.T.–1800 °C. Dwell time at test temperature was 1 h. Strain rate was 10^{-3} /s. Tensile specimens were cut out from three directions (i.e. X, Y, and Z) in plate material. To investigate the thermal property, thermal diffusivity and specific heat were measured. Thermal diffusivity was measured by laser flash method at R.T.–1100 °C. Specific heat was measured using differential scanning calorimetry (DSC) at R.T.–1400 °C.

To evaluate the applicability of designed W alloys under heat load conditions relevant to a fusion reactor, thermo-mechanical analysis using finite element method was performed. In the finite element analysis (FEA), the temperature and stress values and their distributions in W monoblock during a heat load were calculated. The FE-model was based on the dimensions of monoblock for ITER, which consists of W, OFHC-Cu, and CuCrZr as PFM, buffer layer material, and cooling tube material,

respectively. The 1/4 model of the monoblock was used for this analysis. The material property data were obtained from the experimental results of this study and the literatures. To consider the anisotropic strength of W materials in FEA, Hill yield criterion was used in this study. The thermo-mechanical analysis was carried out using ANSYS ver. 15.0. All materials were defined as perfect elastro-plastic solids in this work. The axial symmetry surface of the X and Y axes in 1/4 model were fixed. The heat load conditions were 20 MW/m^2 heat load with a dwell time of 10 s at the top surface of the monoblock and the cooling duration was 20 s. The cooling water temperature and its pressure was 25 °C and 2 MPa, respectively.

Conventional methods to improve the mechanical property of metals and alloys in W are the grain structure refining, strain hardening, solid solution strengthening, and dispersion strengthening. Some of which methods are also effective for the improvement of the resistance to recrystallization embrittlement. In the case of W, Re is considered as the most effective solid solution element because it improves not only mechanical properties such as tensile strength and ductile-brittle transition temperature (DBTT) but also resistance to recrystallization. From the viewpoint of dispersion strengthening, oxide particle, carbide particle, and K-bubble improves mechanical properties and recrystallization resistance of W. Based on these knowledge, the effect of neutron irradiation on W-Re alloys, Lanthanum (La) oxide dispersed W (i.e. La-doped W), and K-bubble dispersed W (i.e. K-doped W) were investigated.

The formation of irradiation-induced defect cluster such as void and irradiation hardening were suppressed in W-3–10%Re alloys compared with pure W after neutron irradiation to ~0.5 dpa. In W-26%Re, irradiation-induced precipitates were formed and irradiation hardening was not suppressed. Suppression of the void formation in W-Re alloys might be attributed to the change in mobility of interstitial atoms in W. It was reported that the mobility of W-Re dumbbell is higher than W-W dumbbell. However, with increasing Re content, Re-rich cluster formation and precipitation will occur, thus, suppression of the irradiation hardening was not observed in W-26%Re. In addition, effects of the grain size on irradiation hardening in pure W and W-Re alloys were investigated. Irradiation hardening in pure W and W-Re alloys with finer grain structure was smaller than that with coarser grain structure. It was considered that the sink density affected these results. Grain boundary and dislocation are sink of irradiation-induced defects, because sink density in material with finer grain structure is larger than that in material with coarser grain structure. Therefore, addition of 3–10%Re and grain structure refining are considered as the effective method to suppress the irradiation embrittlement of W.

In order to investigate the effect of dispersion strengthening on irradiation behavior of W, irradiation effects of La-doped W and K-doped W were studied. The stability of the dispersed second phases such as La oxide and K-bubble and the damage structures were investigated by neutron irradiation to ~0.5 dpa at 531–754 °C. Irradiation hardening in La-doped W and K-doped W showed almost the same behavior compared to pure W. The average size and number density of irradiation induced defect clusters such as void and dislocation loop in La-doped W and K-doped W were almost the same level compared to pure W. The second phases were stable after neutron irradiation in this study. Thus, the effect of second phase dispersion such as La oxide and K-bubble on microstructure development and hardening was not significantly observed under neutron irradiation conditions in this work.

In addition to the irradiation effects of W-Re alloys, La-doped W, and K-doped W, the effect of the difference in neutron energy spectrum on irradiation effect of W was investigated based on the results which were obtained from samples irradiated in the fast test reactor Joyo and the mixed spectrum reactor HFIR. In the pure W, irradiation-induced precipitates were observed after neutron irradiation in HFIR to 0.90 and 0.98 dpa at 500 and 800 °C, respectively. On the other hand, irradiation-induced precipitates were not observed in pure W irradiated to 0.96 and 1.54 dpa at 538 and 750 °C in Joyo, respectively. Due to the formation of irradiation-induced precipitates, irradiation hardening of pure W irradiated in HFIR was larger than that in Joyo. The reason of the differences in microstructure development and irradiation hardening of pure W between HFIR and Joyo irradiated samples was considered to be caused difference of the content of the transmutation products such as Re and osmium (Os). In the HFIR, thermal neutron flux is higher than that in Joyo, because, transmutation from W to Re and Re to Os occurred with higher tendency in HFIR compared to Joyo. The composition change due to transmutation in pure W was estimated using the FISPACT code. The results of the calculation showed that the compositions of pure W changed to W–9%Re–5%Os due to neutron irradiation to 1 dpa in HFIR, and this composition change is larger than that in the case of Joyo irradiation. Therefore, these results suggested that the large composition change during neutron irradiation caused irradiation-induced precipitation in pure W irradiated in HFIR. In W-Re alloys, the size and number density of the precipitates in W-Re alloys were smaller and lower, respectively, than those of pure W after irradiation. The formation of voids and dislocations was suppressed in W-Re alloys as compared with pure W. The existence of Re before irradiation caused the formation of densely populated fine clusters in the W-Re alloys as compared with pure W. The irradiation hardening of W-Re alloys irradiated in HFIR was larger than that in Joyo. The large and dense precipitates are considered to have caused the difference of the magnitude of irradiation hardening.

Based on the experimental results in this study and previously reported results, K-doped W-3%Re plates were fabricated by powder metallurgy and hot rolling in industrial scale as an irradiation and recrystallization resistant W alloy. Other oxide and carbide particle were also considered as the candidate of the dispersed second phase. However, from the view point of the stability of second phases under the fusion reactor operation conditions such as high temperature and high level damage by neutron irradiation, K-bubble was selected. Re was selected to improve the mechanical properties, recrystallization resistance, and irradiation embrittlement. On the other hand, addition of Re decreases thermal conductivity of pure W. One of the major function of divertor is heat removal, because, high thermal conductivity is required. In this study, the addition of Re was selected as 3%, and trade-off relation between thermal conductivity and other properties such as mechanical properties and resistances to recrystallization and irradiation in this material was investigated by FEA. In order to evaluate the combination effects of Re addition and K-bubble dispersion on W, powder metallurgy processed and hot rolled pure W, W-1%Re, W-3%Re, and K-doped W were also fabricated as reference material.

The results of grain structure observation and Vickers hardness measurement of as-received and heat treated materials showed the higher stability of grain structure in K-doped W-3%Re than that in others. In as-received samples, layered grain structure formed by hot rolling were observed. With increasing heat treatment temperature, layered grain structure was changed into isotropic grain structure, and growth of grain was also observed in pure W. On the other hand, W-3%Re, K-doped W, and K-doped W-3%Re kept their grain structure at higher temperature than pure W and W-1%Re. This was because dispersed K-bubble as obstacles and strain field formed by solute Re suppress grain boundary, sub-grain boundary, and dislocation motions at high temperature region. Based on the results of average grain size, grain aspect ratio, and Vickers hardness changes after heat treatment, the temperature where recrystallization starts of the examined materials were estimated. The temperature where recrystallization starts of pure W, K-doped W, and K-doped W-3%Re were estimated as ~1100, ~1300, and ~1800 °C. Higher recrystallization resistance in K-doped W-3%Re by K-bubble dispersion and Re addition were confirmed.

As for the result of tensile tests, highest UTS in X-direction which is parallel to the rolling direction was observed at R.T. On the other hand, the highest UTS was observed at 300–700 °C in Y and Z-directions, and anisotropic tensile strength was observed in pure W, K-doped W, and K-doped W-3%Re. The effects of K-bubble dispersion and 3% Re addition on anisotropic tensile strength and its temperature dependence were not clearly observed. The ductility was also improved by K-bubble dispersion and 3% Re addition.

Based on the results of thermal diffusivity and specific heat measurements, thermal conductivity was calculated. The effect of K-bubble dispersion on thermal conductivity of pure W was not clearly observed. On the other hand, K-doped W-3%Re showed lower thermal conductivity compared to pure W and K-doped W especially at below 800 °C. This was considered to be caused by the scattering of conduction electron by solute Re atoms. The effect of anisotropic grain structure on thermal diffusivity was also investigated, and anisotropy in thermal diffusivity was not clearly observed.

The results of FEA showed that the maximum temperature during heat load of 20 MW/m^2 for 10 s was increased from ~2200 °C to ~2400 °C by 3% Re addition. Although, the temperature area exceeding that where the recrystallization starts in K-doped W-3%Re was smaller than that in pure W and K-doped W. These results suggested that the effect of 3% Re on recrystallization resistance was larger than that on thermal conductivity. As for the thermal stress, the effects of K-bubble dispersion and 3% Re addition on thermal stress and its distribution were not clearly observed. The effect of anisotropic strength in W materials on the result of thermal stress was also investigated in this study. This results suggested that the considering anisotropic strength is necessary to evaluate the magnitude of the thermal stress.

The recrystallization embrittlement decreases UTS at low temperature region. Therefore, temperature level and its distribution in monoblock should be considered. In this study, the area exceeding that where the recrystallization starts which formed by heat load of 20 MW/m^2 during different heat load condition such as 10 and 15 MW/m^2 for 10 s was evaluated using the results of FEA. In the case of pure W, recrystallization embrittled region was formed during 10 MW/m^2 heat load. Therefore, recrystallization embrittlement may occur in pure W during a fusion reactor operation. On the other hand, recrystallization embtittled region was not formed in K-doped W-3%Re during 10 and 15 MW/m^2 heat load. Therefore, it was confirmed that the K-doped W-3%Re will show higher resistance to recrystallization embrittlement. From the view point of irradiation embrittlement, K-doped W-3%Re will show higher resistance to irradiation embrittlement than pure W, because Re will suppress the irradiation hardening, and fine grain structure formed by Re addition and dispersed K-bubble suppress the irradiation hardening. The effect of 3% Re addition on the composition change due to the transmutation in W was considered to be small because of the predicted thermal neutron flux and the composition change due to transmutation were small in DEMO.

In this study, to improve the lifetime and reliability of plasma facing component such as divertor, development of the W material which shows higher resistance to recrystallization and irradiation embrittlement was performed. Result of this study can be summarized as follows:

1. To suppress the irradiation embrittlement of W, it was suggested that addition of 3–10% Re and grain structure refining are effective. Dispersion of the second phase such as La oxide and K-bubble does not affect irradiation hardening behavior in pure W, and dispersion strengthened W may show better mechanical property and resistance to recrystallization under neutron irradiation condition. Based on these experimental results and previous reported data, K-doped W-3%Re was designed and fabricated in this work.
2. The pure W and its alloys examined in this work showed anisotropic grain structure. The anisotropic grain structure caused anisotropic tensile strength and fracture behavior in pure W and its alloys especially at low temperature.
3. The temperature where recrystallization starts was increased by K-bubble dispersion and 3% Re addition. The temperature where recrystallization starts of pure W, K-doped W, and K-doped W-3%Re were estimated as ~1100, ~1300, and ~1800 °C, respectively. K-doped W-3%Re showed ~700 °C higher the temperature where recrystallization starts than that of pure W. Tensile strength was also increased by K-bubble dispersion and 3% Re addition. The magnitude of increase in UTS was 45 and 65 % at most for K-doped W and K-doped W-3%Re, respectively compared to pure W. The temperature region where material shows elongation was expanded by K-bubble dispersion and 3% Re addition. These results showed better recrystallization resistance and tensile properties in K-doped W-3%Re than those in pure W under non-irradiation condition.
4. The results of FEA showed the decrease in recrystallized area in monoblock due to 20 MW/m^2 heat load by K-bubble dispersion and 3% Re addition. The depth of recrystallized area in the mono-block made by K-doped W-3%Re was ~50% smaller than that in pure W after heat load of 20 MW/m^2 for 10 s. As for the trade-off relation between thermal conductivity and recrystallization resistance due to 3% Re addition, improvement of recrystallization resistance was more effective than decrease in thermal conductivity. K-doped W-3%Re will show higher resistance to irradiation embrittlement than pure W due to the suppression of irradiation hardening by 3% Re.