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(別紙様式 2)
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論文の要旨

Summary (Abstract) of doctoral thesis contents

Reduced activation ferritic steel alloys (RAFSs) such as F82H (Fe-8Cr-2W) are the candidate materials for the first wall of DEMO reactors. In this PhD thesis work, one of the technical issues related to hydrogen isotopes permeation through the first wall has been studied.

For the blankets employing self-cooled breeder, the first wall is supposed to be subjected to bi-directional hydrogen isotopes permeation: in one direction by edge plasma-driven permeation (PDP) of deuterium as well as tritium into blankets, and in the other direction by bred tritium gas-driven permeation (GDP) into the edge plasma. It is important to note that deuterium PDP will hinder the recovery efficiency of tritium from the breeder and will probably necessitate isotopes separation as well. On the other hand, tritium GDP, essentially the same effect as gas-puff for fueling except that it is uncontrollable, will lead to an unwanted increase of the particle recycling in the first wall region, which could even affect core confinement performance as well as isotopes mixture imbalance.

Tungsten has been proposed as a candidate plasma-facing material for the divertor of the International Thermonuclear Experimental Reactor (ITER). For a DEMO reactor, surface coatings made of tungsten are necessary to protect the plasma-facing wall made of RAFSs such as F82H. The characterization of hydrogen isotopes PDP and GDP through tungsten coated F82H is of crucial importance to evaluate major reactors design issues including tritium retention, breeding feasibility and first wall particle recycling. There is a number of experimental data of hydrogen isotopes transport and retention behavior in bulk polycrystalline tungsten. However, only limited experimental investigations aimed at understanding that for tungsten coatings. In particular, there have been no experimental studies reported on hydrogen isotopes PDP through tungsten coated RAFSs, which is important to the plasma-wall interaction studies for a DEMO reactor. In this PhD thesis research, hydrogen isotopes PDP and GDP through F82H coated with two different types of tungsten coatings, i.e., vacuum plasma-sprayed tungsten (VPS-W) and sputter-deposited tungsten (SP-W) have been studied. The purposes of this PhD thesis research are: (1) to demonstrate experimentally the hydrogen isotopes (H/D) bi-directional permeation predicted for the first wall of a fusion power reactor, (2) to investigate tungsten coatings effects on GDP and PDP behavior, (3) to investigate the isotopic effects on hydrogen transport, and (4) to establish a fundamental database on hydrogen and deuterium transport parameters for designing the first wall of fusion power reactors.

Hydrogen isotopes (H/D) PDP and GDP through F82H has been investigated in a steady-state laboratory-scale plasma device: VEHICLE-1 under some of the

reactor-relevant conditions. For PDP experiments, the plasma density is of the order of 10^{10} cm^{-3} and the electron temperature is ~5.5 eV measured by a Langmuir probe in front of the sample. The particle bombarding energy is set at -100 V by a negative DC bias applied on the membrane. Taking into account the ion species mix and surface particle reflection, the net implantation flux is estimated to be ~1 × 10^{16} atoms cm^{-2} s^{-1}, relevant to the first wall environment of DEMO reactors. For GDP experiments, the upstream gas pressure is in the range of 10^3-10^5 Pa measured by an absolute pressure gauge. The samples made of F82H and α-Fe are prepared in the same dimensions as those commercially available conflat flanges with an outer diameter of 70 mm, except that a circular area of 35 mm in diameter inside the knife-edge is machined down to thicknesses of 0.5-2 mm. A resistive heater is set beneath the membrane and the sample temperature varies from 150 ℃ to 550 ℃.

The hydrogen transport parameters data taken for α-Fe have been found to be in good agreement with the literature data, which validates the experimental setup in VEHICLE-1 for the evaluation of other first wall candidate materials. Both the GDP and PDP data taken for F82H show thickness dependence, suggesting that hydrogen permeation through F82H is diffusion-limited. The hydrogen and deuterium transport parameters such as permeability, diffusivity, solubility and surface recombination coefficient have been successfully measured for F82H. In particular, the surface recombination coefficient of deuterium on F82H, which is essential to correctly predict the hydrogen isotopes permeation flux through the first wall, has been experimentally measured for the first time. Isotopic effects on hydrogen transport parameters have been discussed. It has been found that the permeability and diffusivity show isotopic mass effect which is predicted by the classical rate theory relating hydrogen diffusion to the atomic jumping frequency. The solubility, however, is governed by the heat of solution, exhibits no isotopic mass effect.

Simultaneous bi-directional hydrogen isotopes (H/D) permeation has been demonstrated experimentally for the first time under controlled experimental conditions. For bi-directional permeation experiments, the thickness of F82H membrane is 0.5 mm and the membrane temperature is set at ~500 ℃. The electron temperature is raised up to ~10 eV for the improved sensitivity of H_α spectroscopy. Experiments indicate that gas-driven hydrogen permeation takes place in the counter direction of plasma-driven deuterium permeation, which then results in an unwanted increase in edge plasma density. Plasma-driven deuterium flows into hydrogen gas has been detected as well. Both the D_2 and HD partial pressures increase after an extended accumulation period of ~2 h for PDP deuterium pressure build-up, meaning that PDP deuterium flows into the hydrogen gas side.

Hydrogen isotopes GDP and PDP experiments have been performed for VPS-W and SP-W coated F82H. This is the first report of experimental studies on hydrogen PDP through tungsten coated RAFSs. The VPS-W coatings are deposited at the temperature of ~600 ℃. The average size of tungsten powder particles is ~25 μm. The thickness of VPS-W coatings used in this work varies from 40 μm to 200 μm. VPS-W coatings have an inhomogeneous microstructure, i.e., a mixture of disorganized areas composed of large unmelted tungsten particles, fine randomly melted tungsten and pores. The average density is evaluated to be ~90% of bulk polycrystalline tungsten. The coatings have been found to be porous and have an open system of connected pores. The observed hydrogen GDP flux through VPS-W coated F82H is reduced compared to that through bare F82H. The main effect of VPS-W coatings on hydrogen permeation is to reduce the incoming flux at the W/F82H interface owing to pore diffusion in the coatings and to reduce the effective surface area for hydrogen dissolution in the substrate. PDP experiments indicate that VPS-W coatings reduce PDP fluxes by more than one order of magnitude relative to that for bare F82H from 300 ℃ to 550 ℃. Reduced PDP fluxes are attributed to the complex microstructure and a substantial surface-connected porosity of the coatings.

SP-W coatings are prepared by argon ion sputtering at the temperature of ~300 ℃ and gas pressure of ~0.19 Pa. The density of SP-W coatings is evaluated to be ~99.5% of bulk polycrystalline tungsten. The thickness of SP-W coatings used in this work varies from 0.5 μm to 4 μm. The surface of SP-W coatings is well organized with an average crystal size of ~100 nm. Columnar tungsten grains are observed from the cross-section view and the W/F82H interface is sharp without voids, cracks or other visible defects. Hydrogen isotopes GDP fluxes are reduced significantly by SP-W coatings. The hydrogen and deuterium transport parameters for SP-W coatings have been evaluated by the gas permeation technique in the temperature range from 200 ℃ to 550 ℃ employing a diffusion-limited permeation model. The permeability is comparable to the literature data of bulk polycrystalline tungsten. However, the effective solubility and diffusivity are different from that of bulk polycrystalline tungsten by several orders of magnitude, which is attributed to the presence of trapping sites resulting from the characteristic microstructure of SP-W coatings. The trapping effects on hydrogen migration have been discussed. For PDP experiments, it has been found that SP-W coatings tend to enhance PDP compared to that through bare F82H which is attributed to the surface recombination characteristics of SP-W. Static deuterium retention has been measured by the thermal desorption spectroscopy (TDS) after steady-state plasma exposure. Increased deuterium retention in SP-W coated F82H has been observed. The deuterium depth concentration profile by the secondary ion mass spectrometry (SIMS) shows a good agreement with the retention data and exhibits a sign of “uphill diffusion” of deuterium in the W/F82H bi-layer structure.

As a proof-of-principle experiment, first wall particle flux measurements in the QUEST spherical tokamak at Kyushu University have been conducted using a SP-W coated F82H permeation probe. The thicknesses of tungsten coatings and F82H substrate are 0.5 μm and 0.25 mm, respectively. The permeation area is 35 mm in diameter. The permeation probe is installed inside the closed port at the mid-plane and the membrane is ~15 cm recessed from the radiation shield. A resistive heater is set behind the membrane to keep the sample temperature in the range of 300-350 ℃. The hydrogen PDP data have been taken for the probe exposed to a steady-state wall conditioning discharge. The plasma is produced by the 2.45 GHz ECR system with an input power of 7.5 kW. The steady-state hydrogen permeation flux for the SP-W coated F82H membrane has been evaluated by taking into account the effects of the radiation shield and the present data have been compared with our previous results for bare F82H. A much longer PDP breakthrough time and higher steady-state permeation flux have been observed for SP-W coated F82H. It is concluded, therefore, SP-W coatings enhance hydrogen PDP not only in the VEHICLE-1 linear plasma device but also in the QUEST spherical tokamak.

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博士論文審査結果の要旨

Summary of the results of the doctoral thesis screening

磁場閉じ込め核融合発電炉は、プラズマ周辺部に三重水素増殖と熱交換のためブランケットと呼ばれる装置を備えている。プラズマ側の重水素とブランケット側の三重水素は、鉄系低放射化材料を用いた第一壁で分離される。しかし熱交換効率の要請から高温で維持する必要があるため、水素同位体が鉄系第一壁を容易に透過し、ブランケットで増殖した三重水素の回収効率の低下が予想される。XU YUE 氏は、タングステンが水素透過の障壁となり、プラズマ照射に対して低損耗特性を持つことに着目し、核融合炉材料科学の視点から世界で初めてタングステン膜を成膜した低放射化材料 F82H 鋼の双方向水素透過特性を詳細に調べ、第一壁材としての有効性を明らかにした。

本論文のイントロダクションでは､ブランケットと核融合プラズマに対向する第一壁は、熱交換効率のため約 500℃の高温で使用し、かつ熱機械歪低減のために第一壁厚が約 5mm にする必要があることが述べられている。この条件により、第一壁を介した重水素と三重水素の双方向透過が容易に起こり、第一壁の両面からは異なる機構で第一壁金属内部への水素同位体の溶解と拡散が起こるうることが述べられている。すなわち、プラズマに対向する第一壁側では、数十 eV のエネルギーを持った重水素原子／重水素正イオンが第一壁中へ埋め込まれ、プラズマ誘起透過（Plasma Driven Permeability, 以下 PDP）により第一壁内をブランケット側へ拡散する。逆に、ブランケット側からは気相である三重水素分子が第一壁表面で解離した後に、気体誘起透過（Gas Driven Permeability, 以下 GDP）によりブランケット側からプラズマ側へ向かって拡散する。

上記の過程では､水素同位体のイオンインプランテーション､表面再結合､固体内拡散､固体中の水素トラップを含む種々の物理機構が重複しているため、まず基準データとして F82H 試料片面からの GDP と PDP のそれぞれに対し、水素透過の実験結果を解析し、その後に一面から GDP､他面から PDP による双方向水素透過が起きることを実証している。また、実際に計測した α 鉄と F82H に対する物理量を、先行研究の実験データと比較することで、本実験手法の検証がなされている。

続いて、双方向水素透過低減のために考案した複合材料、すなわち F82H 基板に真空プラズマスプレー (Vacuum Plasma Spray, VPS) 法でタングステン被膜を形成した試料を用い、第一壁の水素透過挙動に対する被膜の影響に関して、発電炉を模擬する条件下での結果と議論が記述されている｡試料温度 120 から 500℃の範囲で GDP と PDP による水素透過の比較において､VPS 膜側からの PDP 透過は２桁程度減少するが、同被膜が多孔質であるため基板側からの GDP 束を抑制することは出来ない事を見出している。

さらに、VPS タングステン膜の問題点の解消するために作製した、より稠密なスパッタ (Sputter-deposition, SP)法によるタングステン膜を F82H 鋼に成膜した試料の実験結果について述べている。この SP 膜により基板側からの GDP 束を約 1 桁減らすことに成功し、SP タングステン膜と F82H 中の水素透過パラメーターを得ている。また、SP 膜側からの PDP では、被膜が無い F82H 試料と比較して水素透過が 1 桁増大する事を実験的に明らかにした。くわえて、この現象が表面再結合特性に起因することを見出した。これらは本研究で初めて得られた実験成果である。この SP タングステン膜付き F82H 試料は、九州大学の球

状トカマク型装置（QUEST）中のプラズマ放電下においても、実験室での結果と同様、PDP束が増大することを検証している。

これらの結果をもとに、第一壁の双方向水素透過束低減のため F82H 鋼に VPS 膜と SP 膜の重畳成膜を行うことで各々の膜の長所を活かした第一壁材料を検討し提案している。

本論文は、将来の核融合炉第一壁材料として、タングステン膜を有する F82H 鋼内における双方向水素透過に関する水素透過パラメーターのデータベースを初めて構築し、タングステン膜の水素透過に関する詳細な特性を解明した。これらは、核融合炉設計に資する重要かつ有用な成果である。

以上の結果から、本審査委員会は本論文が博士学位論文として十分な価値を有し、合格に値するものであると判定した。