PD-W BIMETALLIC SYSTEMS: EROSION BEHAVIOR AND HYDROGEN PERMEABILITY

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Erosion and hydrogen behavior are examined in the dense and high porous W-films on Pd. The obtained results are discussed from the viewpoint of plasma facing diffusion system creation for hydrogen isotope recycling control or tritium extraction.
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1. INTRODUCTION

To create the effective plasma facing W-Pd diffusion systems for hydrogen recycling control and possible tritium extraction, it was necessary to increase as much as possible the hydrogen permeability of erosion resistant W-layer of such two-layer diffusion systems.

One of the ways for this is the use of very porous W. So in this work the technology was developed to produce Pd-W systems with high porous tungsten coatings, and the investigations of erosion behavior and hydrogen permeability of such systems were carried out.

2. EXPERIMENTAL AND RESULTS

The experimental setup used in the *erosion experiments* was the device DSM-1 [1] with steady state mirror Penning discharge, which was ignited at magnetic field 0.05 T and at work gas pressure about 0.2 Pa. Plasma characteristics measurements with help of multigrid and single Langmuir probes (central and peripheral discharge regions, accordingly) have shown that nitrogen ion energy for maximum of distribution function are about 0.8 U for the whole range of discharge voltage U from 0.6 keV to 2 keV. The samples for studies (surface morphology and structure are shown in Figures 1 and 2) were W films vacuum-plasma deposited (VPD) or chemical vapor deposited (VPD) on palladium/nickel foils of 20...25 mm diameter (for erosion experiments) or on the 99.98 % pure Pd tubes of 6 mm diameter, 0.25 mm thickness and 190 mm length (for permeation studies). Porosity of VPD tungsten coatings was changed over the range from 1...3 % to 45 %. The last was produced in argon atmosphere at the pressure of more than 10 Pa. Only dense CVD W-coatings were used, as the impurity concentration in coatings strongly increased at the regime of porous CVD W-film producing.

Erosion coefficient values were measured by the weight loss method reviewed in details in [1]. It is seen in Figures 3-5 that erosion rate of porous tungsten films is similar to that for dense VPD/CVD tungsten films and that W erosion rate weakly depends on hydrogen admixture to nitrogen up to 40...50 % concentration.

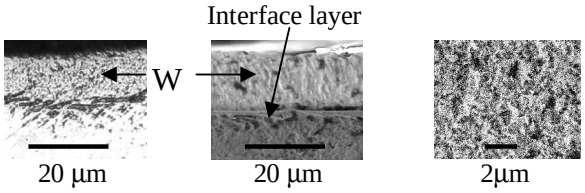

Fig. 1. The metallographic microscopic sections: a) after and b) before etching; (c) surface morphology of 15μm VPD W film deposited at Ar pressure of 10.4 Pa

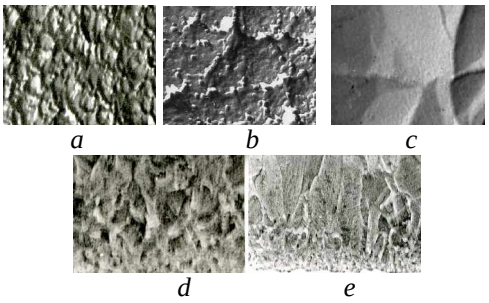

Fig.2. Surface morphology (x6000) of CVD W-films deposited at the pressure of 2.6 Pa and at the temperatures: a) 673 K, b) 773 K, c) 1073 K, and microstructures (x1000) of the ones deposited at the temperatures: d) 773 K , e) 1073 K

The scheme and methods of *hydrogen permeation experiments* were similar to the used in the previous works [2]. The dependencies of specific hydrogen flow (permeation rate) j through Pd membrane with VPD W-films on inlet hydrogen pressure p and on membrane temperature T are shown in Fig. 6 and Fig. 7. The similar dependencies have been measured for CVD W-Pd systems. From the data of temperature dependencies of hydrogen permeation (Fig.7), the activation energy values E of hydrogen permeability were calculated to be for two-layer Pd-W systems with high tungsten porosity E = 15.44 kJ/mol (for 9 μm and 14 μm W film thickness).

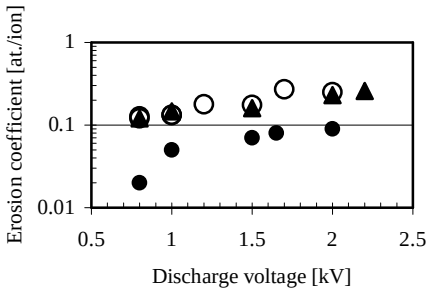

Fig. 3. Erosion coefficient dependence on nitrogen ion energy for Pd-W systems: ▲- denseVPD W film, ○ – VPD W film of the 45% porosity, – - experimental data for hydrogen saturated W films [2]

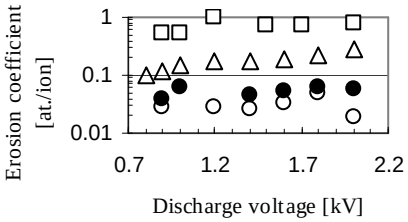

Fig.4.CVD W-coating erosion coefficient energy dependencies for different ions: Ar^+ (o), N^+ (r), He^+ (–), H^+ (○)

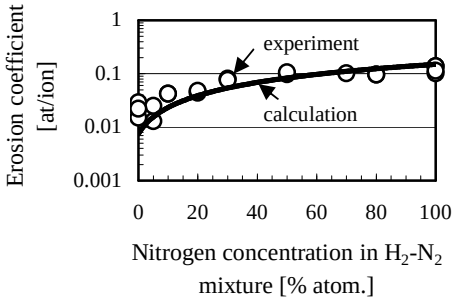

Fig.5. Tungsten erosion coefficient versus nitrogen concentration in hydrogen: experimental data for 1.2 keV ion irradiation (○), curve is the calculated data

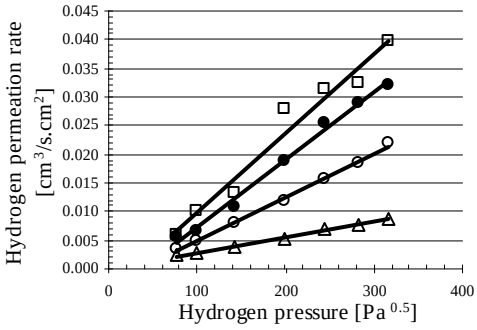

Fig. 6. Hydrogen permeation rate at 973K temperature versus pressure: bare palladium (o); high porous 14 μm and 9 μm VPD W film on Pd (●, š ,); 4 μm dense VPD W film on Pd (r)

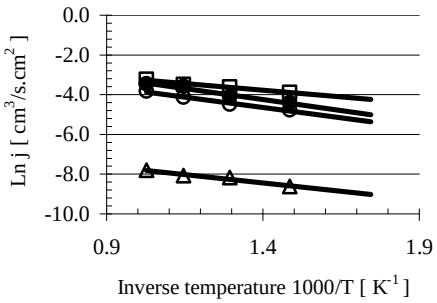

Fig. 7. Temperature dependencies of specific hydrogen flow through two-layer Pd-W systems at 10^5 Pa H_2 pressure: bare palladium (o); high porous 14 μm and 9 μm W film on Pd (●, š); 4 μm dense W film on Pd (r)

This value of activation energy is higher that for bare palladium (11 kJ/mol) and higher than for dense VPD/CVD W-films (11...13 kJ/mol). But, in any case, the values of activation energy of hydrogen permeation through W-Pd bimetallic systems are much lower than the one (131.67 kJ/mol) for the bulk W under gas-driven permeation experiments reviewed in [3].

3. DISCUSSION

The absence of an influence of W porosity on its erosion behavior (Fig. 3) is very encouraging result for creation of plasma facing diffusion system. To explain such erosion behavior one can to suppose the strong influence of redeposition processes, when large number of sputtered tungsten atoms deposits on nearest surfaces of W film pores.

It is seen in Fig. 5, that the sputtering weakly depends on hydrogen admixture to nitrogen up to the 40% concentration. Such dependence can be easily explained on the base of mass dependence of erosion rate, according to equation [1]: $\alpha = \alpha(H^+)\cdot I_H/I + \alpha(N^+)\cdot I_N/I$, where $\alpha(H^+)$ and $\alpha(N^+)$ are the erosion coefficients under hydrogen and nitrogen ion bombardment, respectively; I is the total ion current; I_H and I_N are hydrogen and nitrogen ion currents, respectively. So, hydrogen addition to the main working gas (nitrogen) could not be the reason of the reported in [2] and above mentioned effect of an erosion coefficient decrease for hydrogen saturated W. And the suggested in [2] physical mechanism to explain this effect is, most likely, hydrogen selective sputtering.

Hydrogen permeation flow through W-Pd membranes is near to $j(P) \sim P^{0.5}$ for both dense and porous W-films (see Fig. 6). But the unusual film thickness dependence of hydrogen flow is observed: hydrogen flow through 14 μm tungsten layer is higher than through W film of 9 μm thickness. Investigations of substrate-film interface have shown that transition layer forms of W solid solution in Pd in Pd-W samples produced at more than 873 K (see Fig.1). The additional heating can change transition layer state (width, composition, etc.) and can lead to increase of its role in hydrogen permeation process. The special investigations of the influence of long time baking of W-films on hydrogen permeation rate have shown its essential (up to one order of magnitude) increase with baking time increase [Fig. 8]. The activation energy values increase, too.

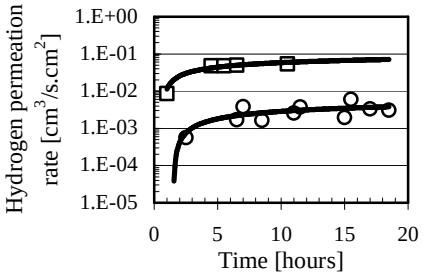

Fig.8. Hydrogen permeation rate measured at 973K temperature and at hydrogen pressure of 10^5 Pa versus time of baking at 973K temperature: o - 4 μm VPD W- film on Pd, š - 300 μm CVD W-film on Pd

So the diffusion in transition layer on the Pd-W interface could be the limiting stage. If so, one can explain high hydrogen permeation with film thickness and heating time increase as "interface opening".

If to analyze the results on Pd-W system erosion behavior together with the data on hydrogen permeation, it is came into clear that high porous tungsten films are very convenient material for plasma facing diffusion system creation. Being of high erosion resistant, such films can provide high hydrogen flows through them. These properties could be useful not only for hydrogen recycling control but for tungsten erosion decrease and tritium extraction in future fusion systems. The obtained results allowed starting the creation of working model of plasma facing diffusion system. In the nearest future such system will be installed in the Uragan-3M stellarator in order to test in the edge plasma conditions.

4. CONCLUSIONS

There was not observed the influence of porosity of W films on their erosion behavior.

Hydrogen admixture to nitrogen (up to 40% atom.) does not essentially influence on the W erosion rate. This confirms the hydrogen selective sputtering as the main physical mechanism explaining the early observed effect of W erosion coefficient decrease in hydrogen saturated Pd-W system.

Hydrogen permeation rate through Pd-W systems with high-porous W-films and activation energy of hydrogen permeability for such systems are higher than for Pd-W two-layer systems with dense W coatings. These results could be explained if to suppose the diffusion in transition layer on the Pd-W interface as the limiting stage of Pd-W two-layer system permeation process.

ACKNOWLEDGEMENTS

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PD-W БИМЕТАЛЛИЧЕСКИЕ СИСТЕМЫ: ЭРОЗИОННОЕ ПОВЕДЕНИЕ И ВОДОРОДОПРОНИЦАЕМОСТЬ

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Исследуется эрозия и поведение водорода в плотных и высокопористых W-пленках на Pd. Полученные результаты обсуждаются с точки зрения создания обращенной в плазму диффузионной системы для контроля рециклинга изотопов водорода и выделения трития.

PD-W БІМЕТАЛІЧНІ СИСТЕМИ: ЕРОЗІЙНА ПОВЕДІНКА І ВОДНЕВА ПРОНИКНІСТЬ

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Досліджується ерозія і поведінка водню у щільних і високопористих W-плівках на Pd. Одержані результати обговорюються з погляду створення звернутої до плазми дифузійної системи для контролю рециклінгу ізотопів водню і виділення тритію.