
ROLE OF TUNGSTEN IN CURRENT GENERATION AT CATALYTIC DECOMPOSITION OF WATER

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Current generation, which occurs at the catalytic decomposition of water $\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^-$ on electrodes either fabricated of polycrystalline tungsten (W) or fabricated of *n*-Si or Cr_3Si and covered with a W-admixture electrolytically deposited on those electrodes from the aqueous solution of a tungsten salt, has been studied. The catalytic activity of tungsten with respect to the water decomposition was found to be a little lower in comparison with the activity of other transition metals; nevertheless, the inhibited rate of W oxidation can be used in aqueous electrochemical elements.

1. Introduction

One of the natural sources to obtain energy is the generation of a current between two electrodes with different electron work functions (or with different electrochemical potentials) which are embedded into distilled water [1–7]. The current generation is a result of the decomposition of water molecules following the reaction $\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^-$ due to the catalytic action of electrodes (or at least one of them) and the existing difference of electron potentials between electrodes; the latter being maintained by the energy which is released at electrodes, when ions discharge on them.

The energy obtained in the course of the decomposition reaction $\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^-$ consists of two components. One of them is associated with the current generation arising when the external electric circuit between the electrodes is closed, and the other with the generation of hydrogen at the cathode electrode. The released hydrogen can be further used as fuel. The theoretical results of work [4] testify that the energy obtained due to the current generation is much larger

(by a factor of tens) than that which can be obtained making use of released hydrogen.

In works [1–7], to obtain the most effective current generation, we studied various electrodes. The anode electrode made of ytterbium (Yb), the electron work function for which depends on the degree of electrode oxidation and ranges from 3.0 to 3.7 eV [4], has appeared to be the most effective one. Moreover, Yb is a good catalyst of water decomposition at various degrees of its oxidation.

A wide collection of different materials were studied in works [1–7] as cathode electrodes; in particular, transition metals, which are known [8] to be effective catalysts in redox processes, as well as silicon and various silicides after their surfaces having been subjected to physical and chemical modifications. It turned out that metals, silicon, and silicides are more effective at water decomposition, if their surfaces are made structured (by grinding or chemical etching), or they are deposited as micro- (nano-) crystalline films onto definite substrates. The shortcoming of film electrodes is their relatively fast oxidation in the course of exploitation and storage in the air environment, which can result in the deterioration of cathode electrode operational characteristics [3–7].

In this work, we studied the most refractory and weakly oxidized transition metal – tungsten (W) – as an electrode. An electrode made of polycrystalline W was a cathode in combination with a Yb electrode, and an anode in combination with a Pt electrode. At the same time, tungsten island films obtained on Si- and Cr_3Si -substrates by electrochemical deposition of W from a tungsten salt served mainly as cathodes, making use of Yb electrodes as anodes.

2. Experimental Technique

The technique of experimental researches of the current generation between different electrodes embedded into an aqueous medium was described by us in details in our previous works (see, e.g., works [1–3]). A pair of corresponding electrodes was fixed with the help of the same holder and simultaneously immersed into distilled water. The electrode-to-electrode distance was in the range $L = 1.5 \div 2$ cm, and the electrode area was $S = 1 \div 2$ cm². In 1 to 2 min after the electrode immersion, an electronic voltmeter was used to measure a stationary voltage ΔV_c between the electrodes; afterwards, after the external electric circuit having been short-circuited, an microammeter was used to measure the time dependences of the current $J(t)$ generated in water by H^+ and OH^- ions.

In the experiments with different electrodes, the values of the parameters L , S , and ΔV_c were different. So, in order to compare the influence of electrode materials on the rate of water decomposition at electrodes, we plotted the time dependences of the effective conductivity of the electrochemical system $\sigma_{\text{eff}} = AJ(t)$, where the coefficient $A = L/(\Delta V_c S)$. Besides the current $J(t)$ caused by the difference ΔV_c , we also measured the currents, when an additional source of voltage $V_0 = 9.7$ V (a battery) was included into the external circuit, being connected by its positive (+) or negative (–) pole, respectively, with the anode electrode. This enabled us to draw new conclusions about the role of the voltage amplitude between electrodes in the formation of the $AJ(t)$ quantity. In the latter case ($V_0 \neq 0$), while determining σ_{eff} , the values of $\pm V_0 + \Delta V_c$ rather than ΔV_c were taken into consideration.

A Pt electrode is characterized by a relatively stable value of the electron work function $\varphi_{\text{Pt}} = 5.32$ eV in comparison with that for electrodes made of other materials. Therefore, we used it to determine the electron work function φ_{el} of other counter electrodes with respect to this one, by measuring the currents $J_1(t \approx 0)$ and $J_2(t \approx 0)$ corresponding to the inclusion of the voltage $\pm V_0$, respectively, into the electric circuit and making use of the formula $\varphi_{\text{el}} = (5.32 - \Delta\varphi)$ eV, where [1–3]

$$\Delta\varphi = qV_0 \frac{J_1 - J_2}{J_1 + J_2},$$

and q is the electron charge. The Table contains the data determined in such a way for the work function φ

of electrodes used in this work in their various physical and chemical states.

It is worth noting that, in this work, the current amplitudes and their time dependences did not vary, when the natural illumination changed from twilight to bright sun light. This means that the photochemical processes played no substantial role in our present experiments, as it was observed in specially arranged researches fulfilled in work [5].

3. Experimental Results and Their Discussions

Our research of tungsten as an electrode in an electrochemical system was stimulated by its physical and chemical properties. First, tungsten is a transition metal (its external electron configuration is $4f^{14}5d^46s^2$) with the unfilled electron shells $4f$ and $5d$. This fact must predetermine its catalytic activity in redox processes; in particular, at the water decomposition $H_2O \rightarrow H^+ + OH^-$. Second, tungsten is the most refractory metal ($T_m = 3410$ °C) and, chemically, is rather stable. It hardly interacts with water, oxygen, and hydrogen. An appreciable oxidation of tungsten in the air environment to the WO_3 oxide is observed only at temperatures $T \geq 450$ °C.

At the first stage of our researches, we studied the electrode pairs Yb–Pt, Yb–W, and W–Pt made of polycrystalline materials (the electrodes were flat plates). Note that tungsten is a cathode in the Yb–W pair and an anode in the W–Pt one. The electron work function determined for W naturally oxidized in air was $\varphi_W = 4.75$ eV (see the Table). The value of φ_W for “clean” tungsten obtained by thermionic methods amounts to 4.55 eV.

In Fig. 1, the time dependences of the effective conductivity $AJ(t)$ for electrochemical systems including those three pairs of electrodes are depicted; the dependences were measured either when no additional external voltage ($V_0 = 0$) was inserted into the electric circuit (curves 1, 2, and 3) or when the voltage $V_0 = +9.7$ V (curves 1⁽⁺⁾, 2⁽⁺⁾, and 3⁽⁺⁾) or $V_0 = -9.7$ V (curves 1^(–), 2^(–), and 3^(–)) was applied to the anode electrode. Curves 1, 2, and 3 are similar to one another by the character of their time dependence and do not differ strongly by $AJ(t)$ -value at the same t . A certain reduction of the $AJ(t)$ -value with time in the interval $t < 2$ min is caused by moving apart H^+ and OH^- ions emerging at electrodes owing to the catalytic processes running at the latter before the electric circuit was closed (at $t \leq 0$). After attaining a minimal value, the effective conductivity grows within the interval 2–15 min to

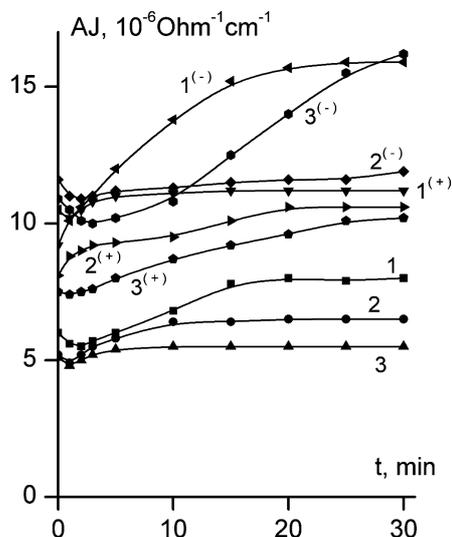


Fig. 1. Time dependences of the conductivity $AJ(t)$ measured in the electrochemical systems of Yb–Pt (curves 1, $1^{(+)}$, and $1^{(-)}$), Yb–W (curves 2, $2^{(+)}$, and $2^{(-)}$), and W–Pt (curves 3, $3^{(+)}$, and $3^{(-)}$) electrode pairs in distilled water without application of an external voltage V_0 (i.e. $V_0 = 0$) (curves 1, 2, and 3) and at $V_0 = \pm 9.7$ V on the first-in-the-pair electrode (Yb, Yb, and W, respectively) (curves $1^{(+)}$, $1^{(-)}$, $2^{(+)}$, $2^{(-)}$, $3^{(+)}$, and $3^{(-)}$)

saturation. Such a behavior is associated with the variation of the catalytic activity of electrodes in the course of water decomposition at them due to physical and chemical changes occurring on the electrodes' surfaces. We note that the simultaneous monitoring of the variation of ΔV_c -values in time (it was taken into account while calculating $AJ(t)$) showed that this parameter changed within the time interval $t = 0 \div 30$ min from 1.3 to 1.0 V, from 1.15 to 0.45 V, and from 0.5 to 0.35 V for the Yb–Pt, Yb–W, and W–Pt electrode pair, respectively. This means that the variation of the voltage $\Delta V_c(t)$ between electrodes cannot explain the growth of the $AJ(t)$ -value in time.

The character of the time dependences $AJ(t)$ after the inclusion of an additional voltage $V_0 = \pm 9.7$ V into the electric circuit (curves $1^{(+)}$, $2^{(+)}$, $3^{(+)}$ and $1^{(-)}$, $2^{(-)}$, $3^{(-)}$) became different not only by the $AJ(t)$ -amplitude, but also by the features of the AJ -variation in time t . This circumstance is associated with the manifestation – by electrodes – of their individual properties, and how – as an anode or a cathode – the specific electrode is made use of. The common feature of the curves obtained in the case where the voltage $V_0 = \pm 9.7$ V was applied is the appreciable growth of the $AJ(t)$ -values in comparison with the corresponding values observed at $V_0 = 0$. This fact, as has been

directly demonstrated in works [2–4], is associated with an enhancement of the catalytic water decomposition at structural and relief inhomogeneities of electrodes, when the voltage between the latter increases.

By analyzing the dependences $AJ(t)$ exhibited in Fig. 1, one can draw a number of important conclusions, namely:

- 1) the catalytic activity of tungsten as for the water decomposition is approximately equal to that of platinum;
- 2) the catalytic activity of tungsten is a little bit lower than that of ytterbium;
- 3) penetration of hydrogen into electrodes made of tungsten or ytterbium is somewhat larger than that in the case of platinum electrodes, which manifests itself in a difference between the relevant $AJ(t)$ -dependences.

In works [3, 4, 6, 7], we showed that doping the surfaces of silicon, diamond-like carbon films, titan, and chromium silicide Cr_3Si with transition metals (Pd, Pt, Cr, and Ni) by various methods (chemical and electrochemical deposition from the solutions of salts of those metals or thermal vacuum sputtering) gives rise to an enhancement of the catalytic activity of the surfaces of the indicated materials as for the water decomposition. Therefore, in this work, we studied the surfaces of n -Si and Cr_3Si electrodes after doping them with a W impurity, which was electrochemically deposited onto the surface from the aqueous solution of those metals or thermal vacuum sputtering) gives rise to an enhancement of the catalytic activity of the surfaces of the indicated materials as for the water decomposition. Therefore, in this work, we studied the surfaces of n -Si and Cr_3Si electrodes after doping them with a W impurity, which was electrochemically deposited onto the surface from the aqueous solution of those metals or thermal vacuum sputtering) gives rise to an enhancement of the catalytic activity of the surfaces of the indicated materials as for the water decomposition. Therefore, in this work, we studied the surfaces of n -Si and Cr_3Si electrodes after doping them with a W impurity, which was electrochemically deposited onto the surface from the aqueous solution of those metals or thermal vacuum sputtering) gives rise to an enhancement of the catalytic activity of the surfaces of the indicated materials as for the water decomposition.

In Fig. 2, the dependences $AJ(t)$ for the Yb– n -Si electrode pair with a clean n -Si surface and after the

Electron work function φ of electrodes in their different physical and chemical states

Electrode	Electrode prehistory	φ , eV
Yb	oxidized in the air	3.64
W	polycrystalline, oxidized in the air	4.75
n -Si	oxidized in the air	4.45
n -Si(W) ₁	W-doped ($2 \times 10^{18} \text{ cm}^{-2}$)	4.12
n -Si(W) ₂	W-doped ($2 \times 10^{19} \text{ cm}^{-2}$)	4.31
n -Si(W) ₂	aged for 7 days	4.69
n -Si(W) ₂	aged for 15 days	4.77
Cr_3Si	oxidized in the air	4.67
$\text{Cr}_3\text{Si(W)}$	W-doped ($6 \times 10^{18} \text{ cm}^{-2}$)	4.47
$\text{Cr}_3\text{Si(W)}$	aged for 12 days	4.72
$\text{Cr}_3\text{Si}^\text{g}$	ground	5.12
$\text{Cr}_3\text{Si}^\text{g}(\text{Cr})$	Cr-doped ($1.6 \times 10^{19} \text{ cm}^{-2}$)	5.01
$\text{Cr}_3\text{Si}^\text{g}(\text{Cr, W})$	Cr- i W-doped ($6 \times 10^{18} \text{ cm}^{-2}$)	4.89
$\text{Cr}_3\text{Si}^\text{g}(\text{Cr, W})$	aged for 14 days	5.12
$\text{Cr}_3\text{Si}^\text{g}(\text{Cr, W})$	aged for 40 days	5.03

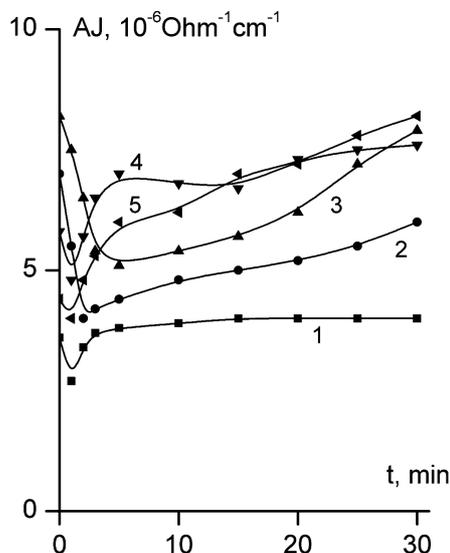


Fig. 2. Time dependences of the conductivity $AJ(t)$ of the following electrochemical systems in distilled water and at $V_0 = 0$: 1 – Yb– n -Si, 2 – Yb– n -Si(W)₁, 3 – Yb– n -Si(W)₂, 4 – Yb– n -Si(W)₂ after 7 days of ageing, 5 – Yb– n -Si(W)₂ after 15 days of ageing in the air. The concentration of doping tungsten: $[W]_1 = 2 \times 10^{18} \text{ cm}^{-2}$, $[W]_2 = 1 \times 10^{19} \text{ cm}^{-2}$

electrolytic deposition of W onto n -Si up to the concentrations $[W]_1 = 2 \times 10^{18} \text{ cm}^{-2}$ and $[W]_2 = 1 \times 10^{19} \text{ cm}^{-2}$ are depicted. In addition, for the system Yb– n -Si(W)₂, where the notation n -Si(W)₂ stands for an n -Si electrode with the surface concentration $[W]_2$ of deposited tungsten, we also plotted the dependences $AJ(t)$ measured after the ageing of the n -Si(W)₂ electrode for 7 and 15 days. The values of φ for corresponding states of the n -Si surface are given in the Table. One can see that the deposition of W onto the n -Si surface enhances the catalytic activity of the electrode, especially in the vicinity of the $t = 0$ and $t = 30$ min points (curves 1, 2, and 3); and that the corresponding dependences $AJ(t)$, although having identical characters, are slightly different from one another owing to the variation of the depth and the width of their minima.

The ageing of the doped n -Si(W)₂ electrode for 7 and 15 days diminishes its catalytic activity only in the time interval $t = 0 \div 3$ min after the immersion, while, at $t > 3$ min, the values of $AJ(t)$ become larger than those obtained right after the doping (curves 4 and 5). This is a positive effect of the n -Si(W)₂ electrode ageing. Ageing is accompanied by some oxidation of the n -Si electrode covered with a W film, which is confirmed by the growth of the work function φ of the electrode (see the Table). We note that the ΔV_c -values for both undoped and W-

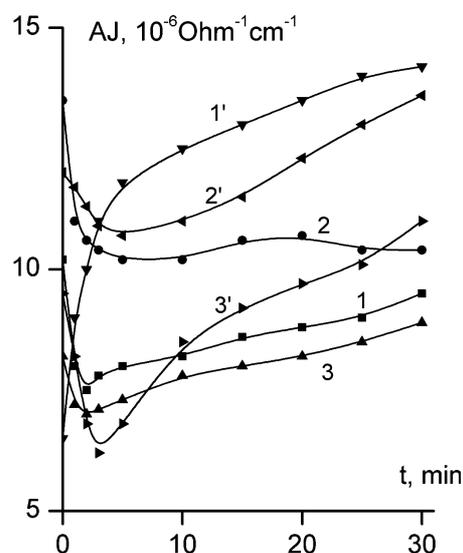


Fig. 3. Time dependences of the conductivity of the systems Cr₃Si–Pt (curved 1, 2, and 3) and Cr₃Si–Yb (curves 1', 2', and 3') at $V_0 = 0$ before (curves 1 and 1') and after (curves 2 and 2') the doping of the Cr₃Si electrode with tungsten ($[W] = 6 \times 10^{18} \text{ cm}^{-2}$), and after the 12-day electrode ageing (curves 3 and 3')

doped n -Si electrodes practically did not change in time t . However, they were different after the doping with W and the ageing, namely, $\Delta V_c = 0.4$ V in the n -Si case, 0.55 V in the n -Si(W)₁ case, and 0.75 V in the n -Si(W)₂ case, but $\Delta V_c = 0.45$ and 0.30 V in the n -Si(W)₂ case after the electrode ageing for 7 and 15 days, respectively.

Although the catalytic activity of Cr₃Si electrode with respect to the water decomposition is higher than that of the n -Si one, doping the Cr₃Si electrode with a W impurity to $6 \times 10^{18} \text{ cm}^{-2}$ turned out less effective than that in the case of n -Si electrode. In Fig. 3, the dependences $AJ(t)$ for the electrode pairs Cr₃Si–Pt (curves 1, 2, and 3) and Cr₃Si–Yb (curves 1', 2', and 3') before (curved 1 and 1') and after the W-doping (curves 2 and 2'), as well as after the 12-day ageing of the Cr₃Si(W) electrode in air (curves 3 and 3'), are presented. Note that the Cr₃Si electrode was an anode in pair with the Pt counter electrode, and a cathode in pair with the Yb counter electrode, both before and after the doping with tungsten. Figure 3 makes it evident that an appreciable increase of the $AJ(t)$ -value at the W-doping was observed in the whole time interval of measurements in the case where the Cr₃Si electrode played the role of anode (curves 1 and 2). The ageing of the Cr₃Si(W) electrode for 12 days compensated the growth of $AJ(t)$ in this case. However, when the Cr₃Si electrode, being in pair with the Yb one, played the role of cathode, the

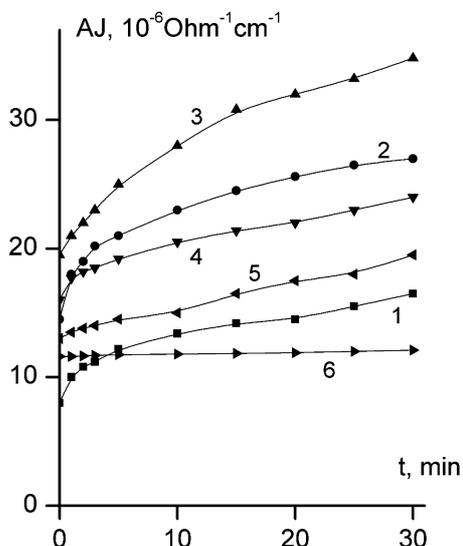


Fig. 4. Time dependences of the conductivity of the Yb-Cr₃Si system in distilled water at $V_0 = 0$: 1 - Yb-Cr₃Si (oxidized in the air), 2 - Yb-Cr₃Si^{asg} (as-ground), 3 - Yb-Cr₃Si^{asg}(Cr) (Cr-doped, [Cr] = 1.6×10^{19} cm⁻²), 4 - Yb-Cr₃Si^{asg}(Cr,W) (doped with Cr and W, [W] = 6×10^{18} cm⁻²), 5 - Yb-Cr₃Si^{asg}(Cr,W) aged for 14 days, 6 - Yb-Cr₃Si^{asg}(Cr,W) aged for 40 days

increase of $AJ(t)$ -values at the W-doping was observed only within the time interval $t = 0 \div 3$ min. (curves 1' and 2'). Ageing brought about a reduction of $AJ(t)$ -values in this case as well, but now they are larger – in particular, at $t > 10$ min – than the corresponding values of curves 1 and 3 measured for the pair with the Pt electrode. Hence, doping Cr₃Si with a W impurity is less effective than the similar doping of the *n*-Si electrode, especially if the ageing effect is made allowance for.

Taking into account that the maximal – by magnitude – current generation was earlier observed in the case where a Pd-doped Cr₃Si electrode was used as a cathode [4], we doped a Cr₃Si electrode two times in this work – first with a Cr impurity and then with a W one. In work [7], the doped films made of Pd and Ni were demonstrated to age most quickly in the air environment, as a result of their oxidation, which diminished the values of $AJ(t)$. That is why Cr rather than Pd was selected as a first doping impurity.

In Fig. 4, the dependences $AJ(t)$ for the electrode pair Yb-Cr₃Si with the untreated Cr₃Si electrode (curve 1), after its mechanical grinding (curve 2), and after its following doping with Cr to 1.6×10^{19} cm⁻² by the method of electrolytic deposition from the aqueous solution of chromium salt (curve 3) are shown. One can see a substantial growth of the $AJ(t)$ -value within the whole time interval of observation. In work [7], it was

also shown that a Cr film on the Cr₃Si electrode ages as well, which results in a reduction of $AJ(t)$, although it does not occur so quickly as if a Pd film was used. Taking into account the positive effect of the tungsten coverage on the *n*-Si electrode – in particular, an insignificant role of ageing after the W-deposition – we hoped for that the additional covering with W could “stabilize” the Cr₃Si(Cr) surface. Curve 4 was obtained after a W film (6×10^{18} cm⁻²) had been electrolytically deposited onto a Cr₃Si(Cr) electrode. An appreciable reduction of the $AJ(t)$ -values was observed (from curve 3 to curve 4), which was evidently caused by a lower catalytic activity of the W film in comparison with the Cr one.

The ageing of the Cr₃Si(Cr,W) electrode in the air environment was also observed, though it was not so fast as in the absence of the W film. Curve 5 illustrates the $AJ(t)$ -values after 14 days of the Cr₃Si(Cr,W) electrode ageing. We note that the $AJ(t)$ -values for curve 5 grew in the course of measurements within 5 h and achieved a value of 32×10^{-6} Ω×cm, which can be regarded as a positive effect. In so doing, the growth of $AJ(t)$ in time is associated with structural and morphological modifications in the Cr₃Si(Cr,W) electrode, which stimulates the growth of its catalytic activity with respect to the water decomposition. At the same time, the amplitude of the voltage ΔV_c between the Yb and Cr₃Si(Cr,W) electrodes separated by a 1.6-cm gap remained almost constant and equal to 1.44 V within the time interval $t = 0 \div 5$ h, whereas the current increased from 9 to 30 μA/cm² within the same time interval. The parameters became worse after the 40-day ageing of the Cr₃Si(Cr,W) electrode (see curve 6). Nevertheless, the application of a Cr₃Si electrode as a cathode should be considered as promising, especially provided that tungsten silicides are formed on its surface; those compounds are practically not oxidized in air, which would enable a stable cathode electrode to be developed.

4. Conclusions

1. By researching the time dependences of the current generation between the electrode pairs Yb-Pt, Yb-W, and W-Pt in distilled water with and without application of an external voltage $V_0 = \pm 9.7$ V, it was found that the catalytic activity of a tungsten electrode with respect to the water decomposition is close to that of a Pt electrode, but lower than the corresponding catalytic activity of an Yb electrode. Penetration of hydrogen into the depth of W and Yb electrodes is a

little bit larger than into a Pt electrode, which manifests itself in the values of conductivity $AJ(t)$ at $V_0 = \pm 9.7$ V.

2. The differences between the $AJ(t)$ -dependences measured at $V_0 = 0$ and ± 9.7 V stem from the enhancement of the electrode catalytic activity with respect to the water decomposition reaction $H_2O \rightarrow H^+ + OH^-$, when the voltage between electrodes increases, due to the emergence of higher electric fields at structural and relief inhomogeneities of the electrode surfaces.

3. Electrolytic deposition of W films from the aqueous solution of tungsten salt onto the surfaces of n -Si and Cr_3Si electrodes increases, as a rule, the catalytic activity of electrodes with respect to the water decomposition. In addition, n -Si(W) electrodes practically do not deteriorate their properties under ageing in the air environment.

4. A consecutive deposition of Cr and W films onto the surface of a Cr_3Si electrode enables one to obtain relatively large $AJ(t)$ -values for $Cr_3Si(Cr,W)$ cathodes and inhibit their ageing.

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РОЛЬ ВОЛЬФРАМУ У СТРУМОУТВОРЕННІ ПРИБИ КАТАЛІТИЧНОМУ РОЗКЛАДАННІ ВОДИ

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Резюме

Досліджено струмоутворення при каталітичному розкладанні води $H_2O \rightarrow H^+ + OH^-$ на електродах з полікристалічного вольфраму W і на електродах з n -Si і Cr_3Si з домішками електролітично осадженого на них W з водного розчину його солі. Встановлено, що каталітична активність W в реакції розкладання води дещо нижча в порівнянні з іншими перехідними металами, проте може бути використане уповільнене його окислення в електрохімічних елементах на основі води.