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Potential barrier in a two-layer porous structure based on ZrO_2 and $Ca_6H_2O_{19}Si_6$, depending on filling of the pores with electrolyte (H_2O)

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Abstract. In this paper, current-voltage characteristics of a two-phase disperse system $Ca_6H_2O_{19}Si_6 + ZrO_2$ as functions of filling of the pores with electrolyte (H_2O) have been investigated. It has been shown that presence of free water in the pores of the $Ca_6H_2O_{19}Si_6 + ZrO_2$ structure maximizes the current through this structure as well as induces blurring of the potential barrier in it due to the appearance of charge carriers (mobile ions), capable of moving through the entire two-layer disperse system, in the free water layer. Outflow of the free water leads not only to the disappearance of an additional channel for free movement of mobile ions through the entire structure, but also to an increase in the ion concentration in the diffuse layer. As a result, the efficiency of the potential barrier at the boundary between the layers of the structure as a rectifying element increases.

Keyword: disperse system, potential barrier, double electric layer, diffuse layer, potential determining layer, interface, $Ca_6H_2O_{19}Si_6 + ZrO_2$, porous structure.

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1. Introduction

Just as the study of current flow processes in electrolytes and superionic conductors led to the emergence of the concept of solid state ionics [1], which has been developed in the last few decades, the research of current flow in disperse systems led to the need to formulate a new concept – semiconductor ionics [2, 3].

Semiconductor ionics as a new area at the intersection of solid state physics and chemistry, electronics and electrochemistry, inorganic chemistry, materials science and energetics, was developed over the past 2-3 years. The impetus for creating and developing this area was the emergence of new approaches to generate environmentally friendly electricity by using disperse systems with ionic conductivity, which to some extent reproduce the functions of semiconductor structures and nanostructures [2–5].

Considering the amount of moisture contained in the air, it may be assumed that the mentioned structures and constructions based on them can serve as some alternative to solar panels in autonomous power supply systems [6–9]. However, widespread use of such constructions remains unclaimed due to their very low specific power [7, 10]. Currently, the development of constructions capable of generating electricity due to interaction with moisture is at a preliminary stage. If you recall, this is reminiscent of the development of solar cells after the discovery of the gate photoelectric effect in Schottky contacts based on metallic copper and copper oxide (Cu-Cu_2O), which for some time remained unclaimed due to very low efficiency.

Of particular interest are heterogeneous (disperse) systems with a highly developed interface as well as various processes occurring in these systems, namely in the surface layer of the interface and in the dispersion environment [11–13]. In this case, materials with so-called electroselective or ion-selective surfaces play an important role. From the point of view of practical application, the demand for such materials is very high. These surfaces allow only one type of ions, anions or cations, to pass through. Examples of electroselective surfaces are semipermeable electrical membranes, electrodes, and artificial micro- or nanochannel systems [14–17]. The behavior of an electrolyte near these surfaces exhibits many unusual features that are useful in practical applications [18–20]. Moreover, when several layers with different ion-selective surfaces are combined, current transfer in them reveals a number of peculiarities. For example, in [21–23], asymmetry of the diffusion permeability of a “two-layer fine-porous membrane” caused by difference in the effective exchange capabilities of its layers was considered. This asymmetry leads to an asymmetric pattern of the distribution of ion concentrations in the system when the direction of the external electric field changes. Materials with ion-selective surfaces are mainly porous structures obtained by pressing powders [24–27]. To raise the efficiency of these materials as catalysts, membranes, *etc*., their surface area is usually increased relative to the volume or mass [28–30]. At the same time, rather little attention is paid to more complex structures that are a combination of the layers with the same chemical composition, but with different geometric dimensions of the constituent elements. Such structures may allow for even more efficient use of the capabilities of highly dispersed materials.

In this work, we investigate *I–V* characteristics of a two-phase disperse system $Ca_6H_2O_{19}Si_6 + ZrO_2$, obtained by bringing two layers of compacted particles in the form of pressed tablets of ZrO_2 and xonotlite ($Ca_6H_2O_{19}Si_6$) in contact by a pressing method, depending on the filling of the pores with electrolyte (H_2O).

2. Experimental

In this work, we investigated two-layer disperse systems $Ca_6H_2O_{19}Si_6$+ZrO_2 obtained by bringing in contact by a pressing method two layers of compacted particles in the form of pressed tablets of ZrO_2 + 3mol.% Y_2O_3 (hereinafter ZrO_2) and xonotlite ($Ca_6H_2O_{19}Si_6$). The layers were initially completely saturated with distilled water. To compact the material into tablets with the diameter $d = 20$ mm and the hight $h \sim 2$ mm, powders of xonotlite $Ca_6H_2O_{19}Si_6$ and ZrO_2 were compressed under uniaxial pressure of ~10 MPa.

ZrO_2 nanopowders were synthesized by the method of inverse co-deposition from aqueous solutions of zirconium and yttrium chlorides taken in a stoichiometric ratio, with an aqueous solution of ammonia at pH > 9 [31]. The hydroxide was dried in a microwave oven until the weight ceased to decrease [31]. Calcination was carried out at a temperature of 700 °C in a resistance furnace. The holding time was 2 hours [31]. The ZrO_2 powders were a tetragonal modification of $ZrO_2$with a specific surface area of 50 m^2/g, as obtained by the measurements using the BET method [32].

Quantitative analysis of the elemental composition of the $Ca_6H_2O_{19}Si_6$ obtained on a Rigaku NEXQC+ fluorescence spectrometer is presented in [33].

The potential difference measurements were performed using a MASTECHMS 8040 voltmeter. The circuit included a load resistance R_{load} = 1.5 MOhm. The applied potential range was ±2 V. The measurements were carried out at room temperature (T = 295 K). The investigated samples were in contact with the ambient atmosphere (Rh ~ 45%).

The *I–V* measurements were performed immediately after complete saturation of the structure under study with moisture, after 3 hours, and after 24 hours. Gold pressure contacts were applied to both sides of the structure.

3. Results and discussion

As shown in [34], saturation of the single-layer ZrO_2 structure with moisture from Rh ~ 40% to ~100% leads to an increase in the current through this structure by approximately 1.8-2 times. Saturation of the xonotlite ($Ca_6H_2O_{19}Si_6$) structure with moisture from Rh ~ 40% to ~100%, as in the case of the ZrO_2, leads to an increase in the current through the structure by approximately 22-24 times. The comparison was made for the current values at a voltage of ±2 V. In this case, the current value in the xonotlite ($Ca_6H_2O_{19}Si_6$) based structure is 3 orders of magnitude smaller than that in the structure based on ZrO_2: 10^{-9} A for $Ca_6H_2O_{19}Si_6$ and 10^{-6} A for ZrO_2 [34]. It should be noted that for the single-layer $Ca_6H_2O_{19}Si_6$ and ZrO_2 structures, the *I–V* characteristics are almost symmetrical relative to zero. Moreover, there is no current in the structures at the beginning of the measurements, *i.e.* in the absence of a potential difference on the contacts, which indicates the symmetry of the applied contacts. After dehydration (by annealing), the ZrO_2 and $Ca_6H_2O_{19}Si_6$ structures practically do not conduct electric current [3].

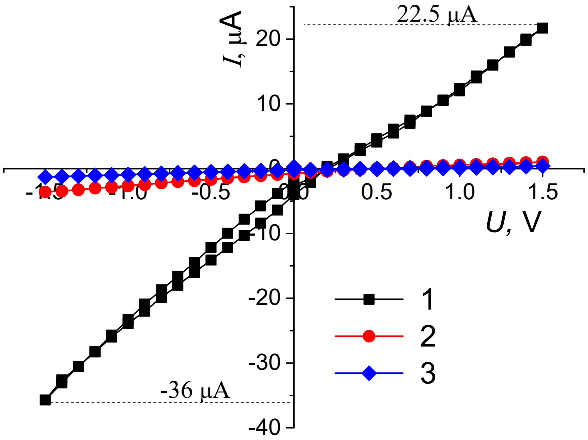

Fig. 1. *I–V* characteristics of the $Ca_6H_2O_{19}Si_6$+ZrO_2 structure immediately after saturation with moisture (*1*), after 3 hours (*2*) and after 24 hours (*3*).

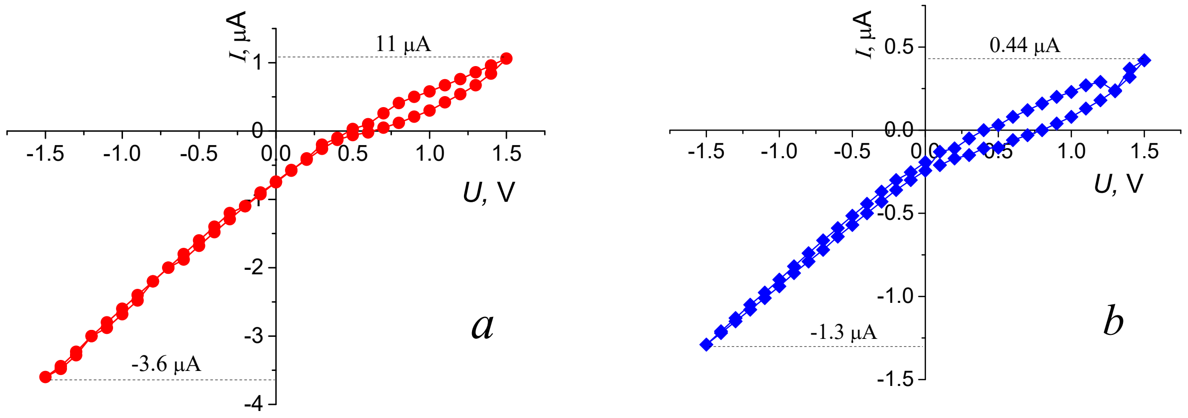

Fig. 2. *I–V* characteristics of the $Ca_6H_2O_{19}Si_6 + ZrO_2$ structure at different times after saturation with moisture: (a) 3 hours, (b) 24 hours.

Fig. 1 shows the *I–V* characteristics of the $Ca_6H_2O_{19}Si_6 + ZrO_2$ structure obtained at different times after complete saturation of the structure with moisture.

As can be seen from Fig. 1, the *I–V* characteristics of the structure immediately after the saturation with moisture have a weakly expressed asymmetric shape relative to the zero potential. The current value on the positive branch is approximately 21.5 μA, and on the negative branch – 35.5 μA, *i.e.* the rectification coefficient of the structure is 1.65. At the same time, the shape of the *I–V* characteristics of the structure measured after 3 hours or more shows that the structure conductivity depends more strongly on the current direction (Figs 2a, 2b). The rectification coefficients obtained from the *I–V* characteristics of the structure measured after 3 and 24 hours are approximately 3.27 and 2.95, respectively.

The observed shift of the *I–V* characteristics relative to zero is caused by the asymmetry of the contact-structure interface, *i.e.* by that one contact is applied to the ZrO_2 layer, and the other one to the $Ca_6H_2O_{19}Si_6$ layer.

As noted in [34], each individual layer of the structure is a disperse system consisting of a solid porous dispersed phase, which are pressed $Ca_6H_2O_{19}Si_6$ or ZrO_2 particles, as well as a dispersion environment – H_2O. Taking into account that OH^- groups and H_2O molecules, oriented depending on the sign of the surface states, are mainly adsorbed on the ZrO_2 surface, and H^+ and also H_2O molecules on the $Ca_6H_2O_{19}Si_6$ surface, the main mobile charge carriers in the diffuse layer of the porous ZrO_2 are positively charged counterions, *i.e.* hydrated protons $H^+(H_2O)_n$, whereas in the diffuse layer of the porous $Ca_6H_2O_{19}Si_6$, the negatively charged counterions are OH^-. When water molecules are adsorbed on the surface of the dispersed phase, the thickness of the diffuse layer may reach 90 nm [35].

Manifestation of the rectifying effect when current passes in different directions through a two-phase disperse system saturated with moisture means that a potential barrier appears in this structure. The authors of [2, 34] showed that when layers of materials with different types of electrical conductivity come in contact, due to the concentration gradient of mobile charge carriers, diffusion of counterions through the interface occurs in the region with the opposite types of electrical conductivity. This leads to formation of a space charge region consisting of two oppositely charged layers, *i.e.* to a potential barrier at the interface between the layers.

As can be seen from the data presented in Fig. 1, the current in the $Ca_6H_2O_{19}Si_6$+ZrO_2 structure drops sharply over time. After 3 hours, it drops almost 10-fold, and after 24 hours, it drops 30-fold as compared to the current value immediately after the saturation with moisture (Fig. 1). This behavior is probably caused by the specificity of the electrolyte structure (dispersion environment) filling the pores of the dispersed phase $Ca_6H_2O_{19}Si_6 + ZrO_2$. In this case, the electrolyte was distilled water, which was chosen as the material that serves as the basis for most electrolytes.

Initially, complete saturation of the dispersed phase $Ca_6H_2O_{19}Si_6 + ZrO_2$ with distilled water leads to filling the pores with moisture with its subsequent structuring inside the pores. As a result, a double electric layer is formed in the pores, which consists of a potential-determining layer, *i.e.* a layer of adsorbed water molecules and ions formed as a result of water dissociation, as well as a diffusion layer and a layer of free water. Note that mobile ions (counterions) that have diffused into the free water layer are not subject to the electrostatic influence of the field of the potential-determining layer, since this field is screened by the diffusion layer. Screening of the potential-determining layer field makes possible movement of the ions with the same charge sign as the ions forming the potential-forming layer, in the free water layer. This leads to the appearance of charge carriers (mobile ions) capable of moving in the free water layer through the entire two-layer disperse system. These mobile ions, unlike the counterions located in the diffuse layer, are not affected by the potential barrier at the boundary of the layers of the $Ca_6H_2O_{19}Si_6 + ZrO_2$ disperse system. Therefore, presence of free water in the pores of the structure can lead to an increase in the value of the current through the structure and to blurring of the potential barrier in it.

It should be noted that configuration of the interface between the layers of the structure (in the case under consideration, the layers are brought into contact by pressing) significantly influences the parameters of the potential barrier in the dispersion environment (H_2O) of the two-phase disperse system. In the presence of a gap between the layers of the structure, the passing through it counterions of the opposite sign lose part of their kinetic energy. Hence, their energy becomes insufficient to overcome mutual attraction during their migration within the gap. Interaction of the counterions of the opposite sign leads to appearance of neutral water molecules and a decrease in the concentration of mobile ions in the entire structure [34].

As can be seen from Figs 2a, 2b, outflow of free water from the disperse system $Ca_6H_2O_{19}Si_6+ZrO_2$ leads to a decrease in the current and an increase in the rectification coefficient (from ~1.6 to ~3). This is a consequence of several factors. The first factor is the absence of a free water layer, and, hence, absence of ions capable of freely moving through the entire structure. Moreover, as is well known, the current I in a conductor is determined as $I = q \times n \times v \times S$ or $I = q \times n \times \mu \times E \times S$, where n is the concentration of charge carriers, q is the electric charge, v is the drift velocity of the charge carriers, μ is the mobility of the charge carriers, E is the electric field strength, and S is the cross-sectional area of the conductor, respectively. It is evident from these expressions that reduction in the cross-section area of the current flow channel by removing free water from the $Ca_6H_2O_{19}Si_6 + ZrO_2$ structure and, consequently, narrowing of the current flow channel to the diffuse layer dimensions leads to a decrease in the current strength (I) in the entire disperse system. Further reduction of the diffuse layer thickness due to moisture desorption will lead to an even greater drop in the current value.

Moreover, outflow of free water leads not only to the disappearance of an additional channel for movement of mobile ions, but also to an increase in their concentration in the diffuse layer. This, in turn, leads to an increase in the efficiency of the potential barrier at the boundary between the layers of the structure, and, consequently, to an increase in the rectification coefficient of the $Ca_6H_2O_{19}Si_6 + ZrO_2$ structure (Figs 2a, 2b).

4. Conclusions

In conclusion, use of several composite materials as disperse systems, each of which consists of a porous matrix ($Ca_6H_2O_{19}Si_6$ and ZrO_2) filled with electrolyte (H_2O), due to the differences in the properties of the porous matrix, makes it possible to obtain structures with a potential barrier formed in the electrolyte environment. This barrier defines the characteristics of movement of ions of a certain sign (anions or cations) through these structures.

The degree of filling of the pores with water as well as the presence or absence of a free water layer in them define both the current flow in a disperse system and the efficiency of the potential barrier at the boundary of two layers in this system.

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Ahmadov F.I.: verification, study design and manuscript preparation.
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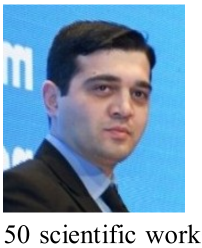

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Потенціальний бар’єр у двошаровій пористій структурі на основі ZrO_2 та $Ca_6H_2O_{19}Si_6 + ZrO_2$ в залежності від заповнення пор електролітом (H_2O)

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Анотація. У цій роботі досліджено вольт-амперні характеристики (ВАХ) у двофазній дисперсній системі $Ca_6H_2O_{19}Si_6 + ZrO_2$ в залежності від заповнення пор електролітом (H_2O). Показано, що наявність у порах структури $Ca_6H_2O_{19}Si_6 + ZrO_2$ вільної води приводить до максимальних значень струму, що протікає крізь структуру, а також розмиття потенціального бар’єра в ній, що зумовлено появою у шарі вільної води носіїв заряду (рухливих іонів), здатних рухатися крізь усю двошарову дисперсну систему. У той же час витік вільної води приводить не тільки до зникнення додаткового каналу для вільного руху рухомих іонів крізь усю структуру, але і до підвищення їх концентрації в дифузному шарі. Внаслідок чого ефективність потенціального бар’єра на межі між шарами структури як елемента, що випрямляє, підвищується.

Ключові слова: дисперсна система, потенціальний бар’єр, подвійний електричний шар, дифузний шар, потенціал-визначальний шар, межа поділу, $Ca_6H_2O_{19}Si_6 + ZrO_2$, пориста структура.