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NEW SYSTEM OF STANDARD EXERGIES OF CHEMICAL ELEMENTS.
II. STANDARD EXERGIES OF COMPLEXOGEN ELEMENTS

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The present work is a continuation of [1, 2] and is devoted to substantiation of the method of calculation of standard exergies of chemical elements that presumably exist in sea water in the form of complex substances, namely, complex neutral compounds or ions of the type $[R(OH)_dO_f\Gamma_g]^k$. *Standard exergies of 41 elements are calculated.*

1. Derivation of the Basic Equation. The "sea" system of standard exergies of chemical elements is based on the postulates given in [1, 2] and partially used by J. Szargut [3] and H. Kameyama [4]. According to the main postulate of the system, in considering reactions of devaluation the components of sea water in environmental concentrations that by agreement can be adopted as mean characteristics of the liquid-phase composition of the World Ocean are used as reference substances or environmental substances [5]. Here, it is assumed, in particular, that the relative thermodynamic activity of H_2O [4] is equal to 0.9849 both in the liquid phase and in the atmosphere. Owing to this the atmosphere and the ocean are in equilibrium relative to H_2O. The activities of ionic components of sea water are assumed to be equal to their concentration, with the exception of H^+ and OH^-, being in equilibrium with molecular H_2O, the activity coefficient of which, as a result, must be equal to $\sqrt{0.9849}$, i.e., 0.9924213.

According to the definition, the exergy of a substance [5] taken in some initial or standard concentration is equal to its maximum useful work of "the devaluation process" in "a reference substance" in the environmental concentration. Here, it is not ruled out that reference substances participate in the reaction of devaluation not only as a product but also as a reagent, the difference between which lies in the sign of the stoichiometric coefficient, being positive for products and negative for reagents.

As is shown in [1, 2], the reaction of devaluation can be accomplished in some ideal fuel cell, namely, a galvanic cell with two electrodes. The first electrode, the unchanged, basic one, is referred to as the air electrode. It is reversible in relation to O_2, H_2O, and the OH^- ion in two environmental media, namely, the atmosphere and sea water and is used in the normal case for conversion of O_2 from the atmosphere to the hydrosphere in the form of the OH^- ion. On this electrode the half-reaction of devaluation of a free electron proceeds according to the following stoichiometric equation:

$$(e^-)_e \rightarrow \left\{OH^-\right\}_e - \frac{1}{2}\left\{H_2O\right\}_e - \frac{1}{4}(O_2)_e . \tag{1}$$

In the formulas of the reagents of the half-reaction of devaluation (1) and henceforth the subscript e is used in order to show their environmental concentration or, in the case of an electron, its relation to half-reaction (1).

The other electrode – variable – serves for devaluation of an arbitrary chemical element R into the R-element environmental substance R_e, in which the element R has an oxidation number – valence – equal to z. As a result of this half-reaction (with both half-reactions being forward), the free electrons obtained are consumed in half-reaction (1) and the reaction of devaluation as a whole turns out to be balanced over electrons. In the simplest

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case, considered in [1], the environmental substance R_e of the element R is a free ion R^z in the environmental concentration with a formal charge z, and the half-reaction of devaluation of the m-atom substance R_m of the element R is described by the following stoichiometric equation:

$$R_m \rightarrow m\left\{(R^z)_e + z\,(e^-)_e\right\}. \qquad (2)$$

To make this half-reaction balanced over electrons, owing to half-reaction (1) it is necessary to add Eq. (1), multiplied by the factor (mz), algebraically to Eq. (2).

According to the fundamental postulate of chemical thermodynamics, a measure of the maximum useful work of chemical processes under isobaric-isothermal conditions is the change in the Gibbs function (energy). Here, the changes in the Gibbs energies in consecutive reactions (half-reactions) are summed up algebraically. Consequently, the total change in the Gibbs energy in the complete reaction of devaluation of the substance R_m is

$$\Delta_{\text{dev}}\, G\,(R_m) = mz\,\Delta_1 G\,(e^-) + \Delta_2 G\,(R_m). \qquad (3)$$

In conformity with the above definition of exergy, the latter can be calculated according to (3) by changing the sign to the opposite, i.e.,

$$W(R_m) = -\,\Delta_{\text{dev}}\, G\,(R_m). \qquad (4)$$

Equation (4) can be used in calculation of the standard exergy $W^0(R_m)$ of the element R if R_m in (2) is taken in the standard state. Henceforth, the environmental complex of the element R will be denoted for brevity by R_e independently of its chemical form – a free ion or a complex.

Now we show that the formula obtained in [1] for calculation of $W^0(R_m)$ can easily be generalized and extended to the case where the environmental form R_e of the element R (or R_m) is a complex ion if the function G_e, which will be called the "environmental" Gibbs energy, is used as a thermodynamic characteristic of environmental substances. For the environmental substance R_e the indicated function is determined by the following expression:

$$G_e\,(R_e) = \Delta_f G^0\,(R_e,\ \text{env. ph.},\ 298.15\ \text{K}) + \text{R}\,T_e \ln a_e. \qquad (5)$$

The activity of environmental complexes of the element R in sea water is replaced by concentrations in the expression for the Gibbs energy. As s result, we arrive at

$$G_e\,(R_e) = \Delta_f G^0\,(R_e,\ \text{aq.},\ 298.15\ \text{K}) + 2478.9204 \ln C_e. \qquad (6)$$

The second terms in (1) and (2) are equal to the work, taken with the opposite sign, of isothermal dilution of the environmental substance R_e from the standard concentration (equal to 1 mole/liter or 1 mole/kg H_2O depending on the agreement about standard concentrations) to the environmental concentration C_e

$$W^0_{\text{dil}}\,(R_e) = -\,2478.9204 \ln C_e. \qquad (7)$$

Note that $W^0_{\text{dil}}(R_e)$ is equal to the standard exergy of the reference substance. Using (6), we can write (4) and (5) in a more coincise form:

$$G_e\,(R_e) = \Delta_f G^0\,(R_e,\ \text{env. ph.},\ 298.15\ \text{K}) - W^0_{\text{dil}}\,(R_e). \qquad (8)$$

The values of G_e for some substances and ions are given in Table 1.

We will illustrate the use of G_e in the formula for calculation of $W^0(R_m)$ in the case where the ion R^z is the environmental substance R. For this, we use (4) by preliminarily determining $\Delta_1 G$ and $\Delta_2 G$ in (3). The general

TABLE 1. "Environmental" Gibbs Energies of Some Reference Substances in Sea Water

Reference substance R_e	$\Delta_f G^0(R_e)$ [7], J/mole	C_e or a_e [8] (with the exception of O_2 and H_2O), mole/liter	$W^0_{\text{dil}}(R_e)$, J/mole	$G_e(R_e)$, J/mole
O_2	0	0.20284774	3954.62	−3954.62
H_2O	−237178.4	0.9849	37.72	−237216.12
OH^-	−157293.29	$9.97608 \cdot 10^{-8}$	39961.44	−197254.73
F^-	−279993.3	$6.8427 \cdot 10^{-5}$	23772.2	−303765.5
Cl^-	−131260.44	0.535921	1546.27	−132806.71
Br^-	−104223.44	$8.135 \cdot 10^{-4}$	17635.43	−121858.87
I^-	−51588.72	$4.728 \cdot 10^{-7}$	36104.48	−87693.2
CO_3^{2-}	−527895.28	$2.3311672 \cdot 10^{-3}$	15025.7	−542921.98
HCO_3^-	−586847.84	$2.3322672 \cdot 10^{-3}$	15025.7	−601873.54
PO_4^{3-}	−1018804	$2.26 \cdot 10^{-6}$	32226.35	−1051030.4
HPO_4^{2-}	−1089262.56	$2.26 \cdot 10^{-6}$	32226.35	−1121488.9
$H_2PO_4^-$	−1130391.28	$2.26 \cdot 10^{-6}$	32226.35	−1162617.655
H_3PO_4	−1142650.4	$2.26 \cdot 10^{-6}$	32226.35	−1174876.855
SO_4^{2-}	−743831.52	$2.604491 \cdot 10^{-2}$	8898.771	−752730.29

expression for the Gibbs-energy change in a chemical reaction and, consequently, for exergy is the following formula (we write it immediately for the half-reaction or reaction of devaluation) [6]:

$$W = -\Delta_{\text{dev}} G = -\sum_j \nu_j G(A_j) . \tag{9}$$

The initial substances – e^- in (1) and R_m in (2) – can be characterized (as adopted, by agreement, in chemical thermodynamics for all chemical elements in the standard state) by the standard Gibbs energies $\Delta_f G^0 = 0$. On the other hand, since $\Delta_1 G$, characterizing process (1), will be used below as the "environmental" Gibbs energy of a "free" electron, it can be denoted by $G_e(e^-)$. In [1] it already occurred in the term of $\Phi = -77658.0$ and now we must check the foregoing by an arithmetic calculation using the data in Table 1 and formula (9):

$$G_e(e^-) = \Phi = \Delta_1 G = G_e(\text{OH}^-) - \frac{1}{2} G_e(\text{H}_2\text{O}) - \frac{1}{4} G_e(\text{O}_2) =$$

$$= -197254.73 - \frac{1}{2} 37.72 - \frac{1}{4} 3954.62 = -77658.0 . \tag{10}$$

As is seen, within the limits of the error of rounding the thermodynamic quantities the value obtained coincides with the value of Φ found earlier in [1].

For the case where the z-charged ion R^z is the environmental form of the element R_m, the exergy of the elemental substance R_m in the standard state can be expressed by (4) with account for (6) and (9) and half-reaction (2):

$$W^0(R_m) = -m \left\{ G_e(R^z) + z\, G_e(e^-) \right\} =$$

$$= -m \left\{ \Delta_f G^0(R^z, \text{aq., } 298.15 \text{ K}) + \text{R}\, T_e \ln C_e + z\Phi \right\} . \tag{11}$$

Formula (11) coincides with the relation in [1], thus confirming the correctness of the methodology based on the environmental Gibbs energies.

Now we extend the above method to the more complicated case where the environmental substance R_e of the element R is the complex ion $[R(OH)_d O_f \Gamma_g]^k$, the formation of which is described by the following half-reaction of devaluation of the element R:

$$R_m \to m \left\{ [R\,(OH)_d\,O_f\,\Gamma_g]_e^k + f\,(H_2O)_e - (d + 2f)\,(OH^-)_e - g\,(\Gamma^r)_e + z\,(e^-)_e \right\}. \tag{12}$$

We apply (9) to (12)

$$W^0\,(R_m) = -\,\Delta_{dev}G_e\,(R_m) = -\,m\,\Big\{G_e\,[R\,(OH)_d\,O_f\,\Gamma_g]^k + fG_e\,(H_2O) -$$

$$-\,(d + 2f)\,G_e(OH^-) - gG_e\,(\Gamma^z) + z\Phi\Big\}. \tag{13}$$

When Eq. (13) is used for calculation of $G_e[R(OH)_d O_f \Gamma_g]^k$, the reference quantities $\Delta_f G^0\{[R(OH)_d O_f \Gamma_g]^k$, aq. 298.15 K$\}$ and $C_e(R)$, for instance, from [7, 8] and formula (6) can be employed.

Expression (13) can be simplified owing to the thermodynamic relation

$$G_e\,(H_2O) - 2G_e\,(OH^-) = \Delta_f G^0\,(OH^-,\ \text{aq.},\ 298.15\ \text{K}), \tag{14}$$

with the help of which we obtain

$$W^0\,(R_m) = -\,m\,\Big\{\Delta_f G^0\,\Big\{[R\,(OH)_d\,O_f\,\Gamma_g]^k,\ \text{aq.},\ 298.15\ \text{K}\Big\} + 2478.9204\,\ln C_e\,(R) -$$

$$-\,f\Delta_f G^0\,(OH^-,\ \text{aq.},\ 298.15\ \text{K}) - dG_e\,(OH^-) - gG_e\,(\Gamma^r) + z\Phi\Big\}. \tag{15}$$

2. Calculation of Exergies of Chemical Elements. *2.1. Sulfur.* The sulfate ion is a well-known component of sea water, and therefore recognition of the ion SO_4^{2-} as the environmental form of sulfur is quite justified. To calculate the exergy of elemental sulfur, it is sufficient to substitute in (15) the values $d = 0$, $f = 4$, $k = -2$, $g = 0$, $z = 6$ and the thermodynamic characteristics of the OH^- group in Table 1, whence we find (J/mole)

$$W^0\,(S) = -\,[-\,752730.29 - 4\,(-\,157293.29) + 6\,(-\,77658)] = 589505.6\,.$$

The value obtained can be compared to ones published earlier: 598830 [3], 608221 [4], 608340 [5], 523000 [9], 300120 [10]. The value found in [3] is closest to ours, which is not surprising, since the SO_4^{2-} ion in sea water is also adopted there as a reference substance. It is noteworthy that the value obtained in [5] does not differ from ours by very much, although it has been calculated under the assumption of the following reaction of devaluation:

$$[S] + \frac{3}{2}O_2 + [CaCO_3] + \{H_2O\} = [CaSO_4\,.\,2H_2O] + CO_2\,.$$

which is apparently attributable to some proximity (or differences of one-three orders of magnitude) of the composition of sea water to the state of saturation with respect to $CaCO_3$ and $CaSO_4 \cdot 2H_2O$. The data of [9] and [10] are obviously incorrect. As regards [9], this has been noted earlier in [2].

2.2. Phosphorus and the other elements. The fact that it is easy to obtain a result for sulfur gives rise to the illusion that it is easy to calculate the exergies of the other chemical elements. However, even in the case of such an element as phosphorus we come across a situation where an enormous number of complex phosphorus-containing anions exist but which of them is environmental is not known a priori. Therefore, it is necessary to repeat calculations by formula (15) for different phosphorus-oxygen, phosphorus-hydrogen-oxygen, and other complexes that can be present in sea water and for which the necessary thermodynamic data are available in the literature. However, additional information and additional principles are also needed.

TABLE 2. Standard Exergies of Complexogens of Chemical Elements

Elemental substance R_m	Content in sea water [8], g/liter	Environmental form R_e	$-\Delta_f G^0(R_e)$ [7], J/mole	Standard exergy $W^0(R)$, J/mole
Ag	$3 \cdot 10^{-7}$	$AgCl_2^-$	215392.3	76772.6
Al	$1 \cdot 10^{-5}$	$Al(OH)_3^0$	1111061.2	788979.06
As	$3 \cdot 10^{-6}$	$HAsO_4^{2-}$	717681.5	479061.2
Au	$4 \cdot 10^{-9}$	$AuCl_2^-$	151167.92	24243.53
B	$4.6 \cdot 10^{-3}$	H_3BO_3	968637.84	629089.45
Ba	$3 \cdot 10^{-5}$	$BaSO_4^0$	1304822.2	745426.54
Be	$6 \cdot 10^{-10}$	$(BeOH^+)_3$	1804140	617529.26
Bi	$2 \cdot 10^{-7}$	BiO^+	146021.6	273182.52
Ca	0.410	$CaSO_4^0$	1098885.7	724374.1
Cd	$1.1 \cdot 10^{-7}$	$CdCl_2^0$	355640	296767.41
Ce	$4 \cdot 10^{-7}$	$CePO_4^0$	1800375.2	1031089.8
Co	$5 \cdot 10^{-7}$	$CoSO_4^0$	813411.44	262068.05
Cr	$5 \cdot 10^{-8}$	CrO_4^{2-}	720861.36	609104.56
Cu	$3 \cdot 10^{-6}$	$Cu(CO_3)_2^{2-}$	1048510.4	159798.51
Fe	$1 \cdot 10^{-5}$	$Fe(OH)_3^0$	677389.6	357110.78
Ga	$3 \cdot 10^{-8}$	$Ga(OH)_4^-$	1012946.4	510363.25
Ge	$7 \cdot 10^{-8}$	$Ge(OH)_4^0$	967131.6	540206.07
Hg	$3 \cdot 10^{-8}$	$Hg(OH)_3^-$	507100.8	126734.05
J_2	$6 \cdot 10^{-5}$	JO_3^-	125938.4	156908.03
La	$3 \cdot 10^{-7}$	$LaCO_3^+$	1258128.8	997643.4
Mg	1.35	$MgCO_3^0$	1002486.4	622045.93
Mn	$2 \cdot 10^{-6}$	$MnSO_4^0$	986670.88	431716.95
Mo	$1 \cdot 10^{-5}$	MoO_4^{2-}	838055.32	714682.77
Nb	$1 \cdot 10^{-8}$	$HNbO_3$	98532	918677.55
Ni	$2 \cdot 10^{-6}$	$NiSO_4^0$	802198.32	247409.1
P	$7 \cdot 10^{-5}$	$H_2PO_4^-$	1130391.28	841811.59
Pb	$1 \cdot 10^{-7}$	$Pb(CO_3)_2^{2-}$	1127169.6	249818.9
S	0.885	SO_4^{2-}	743831.52	589505.13
Sb	$5 \cdot 10^{-7}$	$Sb(OH)_3^0$	647264.88	336344.12
Sc	$4 \cdot 10^{-8}$	ScF_3^0	12540130.4	913468.8
Se	$4 \cdot 10^{-6}$	SeO_4^{2-}	441412	319828.14
Si	$3 \cdot 10^{-3}$	H_4SiO_4	1309884.88	854166.23
Sn	$3 \cdot 10^{-6}$	$Sn(OH)_4^0$	794165 [11]	359142.86
Sr	$1.363 \cdot 10^{-2}$	$SrCO_3^0$	1117098.7	751229.11
Th	$7 \cdot 10^{-7}$	$Th(OH)_3^+$	1353524	1121026
Ti	$1 \cdot 10^{-6}$	$Ti(OH)_4^0$	1318378.4 [12]	883830.26
U	$3 \cdot 10^{-6}$	$UO_2(CO_3)_3^{4-}$	2672739.2	1240424.5
V	$2 \cdot 10^{-6}$	$HV_{10}O_{28}^{5-}$	7706928	756838.57
W	$1 \cdot 10^{-7}$	$H_7W_6O_{24}^{5-}$	5947974.4	834363.1
Y	$3 \cdot 10^{-7}$	YSO_4^+	1448919.2	977519.36
Zn	$1 \cdot 10^{-5}$	$ZnSO_4^0$	904580.8	346068.55

Thus, phosphorus forms compounds with oxygen where its valence is equal to five or less. Intuitively, it is clear to any chemist that in equilibrium with the air atmosphere phosphorus compounds with a valence less than five are hardly likely to exist. On the other hand, the number of different phosphates is rather large and monophosphates (orthophosphates), diphosphates (pyrophosphates), triphosphates (tripolyphosphates), etc. can be present. However, in a state of equilibrium, polymer phosphates must pass into a monomer of orthophosphate to a considerable degree, if not fully. Here, as the phosphorus concentration falls, the degree of hydrolysis and, consequently, the probability of orthophosphate formation increase. As is seen from Table 1, the phosphorus concentration in sea water is rather low and amounts to $2.26 \cdot 10^{-6}$ M. At this concentration, the existence of di- , tri- , and polyphosphate anions can apparently be neglected. As a result this preliminary screening, in the subsequent analysis we will retain only the forms H_3PO_4, $H_2PO_4^-$, HPO_4^{2-}, and PO_4^{3-}, or for the purpose of using formula (15), $[P(OH)_3O]$, $[P(OH)_2O_2]^-$, $[P(OH)_3]^{2-}$, and $[PO_4]^{3-}$.

The final choice of an environmental complex for each element is based on the principle of the highest or "supremum" exergy. Its essence lies in the fact that environmental complexes of the chemical element under consideration to which the highest exergy value of the element does not correspond cannot be stable, since their conversion to a more stable form is then thermodynamically probable and will be accompanied by an additional loss of exergy. Consequently, from the very beginning it is necessary to choose the environmental complex of an element in relation to which the exergy is highest as compared to all other environmental complexes (ions, substances) of the element under consideration. Here we have a case where the concept of exergy is used fruitfully and is effective in solving the problem of substantiation of environmental forms of chemical elements.

Using the data of Table 1, we calculate exergies of phosphorus in relation to environmental forms (indicated at the end of the calculations), J/mole:

$$W_1^0\,(P) = [-\,1142650.4 - 32226.35 - 3\,(-\,197254.73) - (-\,157293.29) +$$

$$+\,5\,(-\,77658)\,] = 814109.27\ (H_3PO_4)\,;$$

$$W_2^0\,(P) = -\,[-\,1130391.28 - 32226.35 - 2\,(-\,197254.73) - 2\,(-\,157293.29) +$$

$$+\,5\,(-\,77658)\,] = 841811.59\ (H_2PO_4{}^-)\,;$$

$$W_3^0\,(P) = -\,[-\,1089262.5 - 32226.35 - (-\,197254.73) - 3\,(-\,157293.29) +$$

$$+\,5\,(-\,77658)\,] = 840644.31\ (HPO_4{}^{2-})\,;$$

$$W_4^0\,(P) = -\,[-\,1018804 - 32226.35 - 4\,(-\,157293.29) + 5\,(-\,77658)\,] = 810147.19\ (PO_4{}^{3-})\,.$$

The highest exergy is the value corresponding to the environmental anion ($H_2PO_4^-$), and it amounts to $W^0(P) = W_2^0(P) = 841811.59$ J/mole. It is noteworthy that the value $W^0(P) = W_3^0(P) = 840644.31$ J/mole corresponding to the environmental form (HPO_4^{2-}) is close to it. In fact, an equilibrium mixture of the anions $H_2PO_4^-$ and HPO_4^{2-} seems to exist in sea water. However, consideration of this case is beyond the scope of the present work. We carried out similar calculations for the other elements. Their results are given in Table 2, where only the most stable environmental forms of the elements and their corresponding exergies are indicated.

NOTATION

a_e, thermodynamic activity of the environmental form of the element R; A_j, j-th reactant participating in the devaluation reaction; C_e, environmental concentration of the element R; e^-, electron; Φ, Gibbs-energy change in the first half-reaction (on the air electrode); G_e, environmental Gibbs energy; $\Delta_f G^0(R_e$, env. ph., 298.15 K), standard Gibbs energy of the environmental form of the element R at 298.15 K; $G(A_j)$, Gibbs energy of the reactant

A_j participating in the devaluation reaction; $\Delta_1 G(e^-)$ or $G_e(e^-)$, Gibbs-energy change in the first half-reaction (on the air electrode); $\Delta_2 G(R_m)$, Gibbs-energy change in the second half-reaction on the R-element electrode; $\Delta_{dev} G(R_m)$, Gibbs-energy change in the complete devaluation reaction of the elemental substance R_m; $\Delta_{dev} G$, Gibbs-energy change in an arbitrary devaluation reaction; v_j, algebraic stoichiometric coefficient of the reactant A_j participating in the devaluation reaction; $[R(OH)_d O_f \Gamma_g]_e^k$, formula of the environmental substance R_e of the element R or R_m (a complex ion with the charge k and the valence z of the element R that is composed of an OH ligand (hydroxyl) in an amount of d units, an O ligand (oxo-ligand) in an amount of f units, and an arbitrary ligand Γ differing from the hydroxyl and the oxo-ligand, in an amount of g units); Γ^r, r-charged ion in the form of which the ligand Γ exists in sea water; R, arbitrary chemical element; R_m, m-atom elemental substance; R_e, environmental substance of the element R or R_m; R, universal gas constant; T_e, environmental temperature ($T_e = 298.15$ K); $W(R_m)$, molar exergy of the elemental substance R_m, J/mole; $W^0(R_m)$, standard molar exergy of the elemental substance R_m; $W^0_{dil}(R_e)$, work isothermal of dilution of the environmental substance R from the standard to the environmental concentration; z, formal ion charge, oxidation number, or valence of the element R.

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