

# On the Galvanic Modification of Seawater

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**How to cite this paper:** Shimkevich, A. (2019) On the Galvanic Modification of Seawater. *World Journal of Condensed Matter Physics*, 9, 112-121.

<https://doi.org/10.4236/wjcmp.2019.94009>

**Received:** October 3, 2019

**Accepted:** October 27, 2019

**Published:** October 30, 2019

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## Abstract

Chemical properties of seawater are studied at forced shifting of Fermi level  $\varepsilon_F$  in the band gap of liquid water due to deviation of its composition  $H_2O_{1-z}$  ( $|z| < 10^{-13}$ ) from the stoichiometric one ( $z = 0$ ). It is shown that the hypo-stoichiometric state ( $z > 0$ ) of seawater emerges when Fermi level is shifted to the local electron level  $\varepsilon_{H_3O}$  of hydroxonium  $H_3O^+$  in galvanic cell with the strongly polarized anode and the quasi-equilibrium cathode. Then, each  $\varepsilon_{H_3O}$  is occupied by electron and hydroxonium radicals  $[H_3O]$  together with hydroxide anions  $[OH^-]$  form in seawater hydrated electrons  $[(H_2O)_2]^-$ . The opposite hyper-stoichiometric state ( $z < 0$ ) of seawater is gotten in galvanic cell with the strongly polarized cathode and the quasi-equilibrium anode. Then, Fermi level is shifted to the local energy level  $\varepsilon_{OH}$  for removing electron from each hydroxide ion  $OH^-$  and forming hydroxyl radicals  $[OH]$  as strong oxidizers. It turned out that the ions of sodium and chlorine are connected into hydrates of sodium hypochlorite  $NaClO$  in this case.

## Keywords

Non-Stoichiometric Seawater, Band Gap, Fermi Level, Galvanic Cell, Electron Donor, Sodium Hypochlorite

## 1. Introduction

Liquid water can be described as a dielectric [1] with the wide band gap  $\varepsilon_g = 6.9$  eV [2] where Fermi level  $\varepsilon_F$  as an identifier of water oxidation-reduction potential (*ORP*) is easily varied by action of the galvanic cell with the strongly polarized one electrode and the other in the quasi-equilibrium with water solution. Such cell works as *ORP* changer of any aqueous solution by a forced shift of Fermi level  $\varepsilon_F$  in the band gap at an expense of insignificant

deviation ( $|z| < 10^{-12}$ ) of water composition  $\text{H}_2\text{O}_{1-z}$  from the stoichiometric one ( $z = 0$ ) [1]. This variation is occupied between two allowed local electron states in the band gap of liquid water such as an occupied-by-electron energy level  $\varepsilon_{\text{OH}^-}$  of hydroxide anion  $\text{OH}^-$  and the vacant one  $\varepsilon_{\text{H}_3\text{O}^+}$  of hydroxonium ion  $\text{H}_3\text{O}^+$ .

These electronic levels are located symmetrically nearby the band-gap middle with the energy difference between them of 1.75 eV [3]. This theoretical concept allows eliminating the inconsistencies [4] in reconciling the electrochemical properties of these well-known aqueous ions in the frame of electronic band structure. Then, Fermi level  $\varepsilon_{\text{F}}$  as an electrochemical potential indicates the tendency of liquid water to donate or accept proton. If  $\varepsilon_{\text{F}}$  is high, there is a strong tendency for liquid water to donate protons, *i.e.* it is reducing. Opposite, if  $\varepsilon_{\text{F}}$  is low in the aqueous medium, there is the strong tendency for that to accept protons when this matter is oxidizing [1].

It is interesting to understand how galvanic modifying of seawater can be useful for a practical application of such approach. The solution to this problem is the subject of the given work.

## 2. The Band Structure of Seawater

We have shown in [3] that a weak aqueous solution of any atomic impurity  $i$  (at its little concentration) has a local electronic level  $\varepsilon_i$  in the band gap.

At the same time, such inherent constituents of liquid water as ions of hydroxonium  $\text{H}_3\text{O}^+$  and hydroxide  $\text{OH}^-$  emerge there due to the self-dissociation of water molecules in the reversible chemical reaction [5]



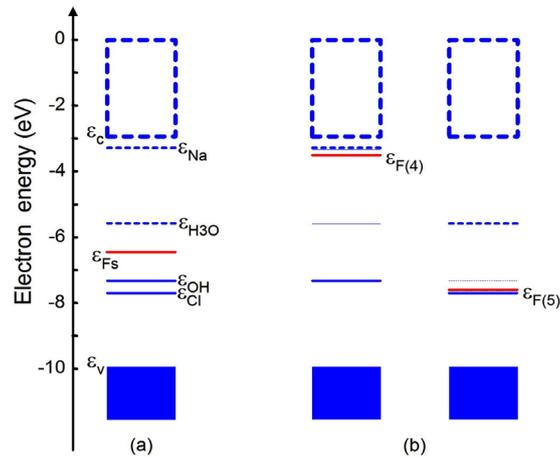
These water constituents have the electronic levels [3]  $\varepsilon_{\text{H}_3\text{O}^+} = -5.575$  eV and  $\varepsilon_{\text{OH}^-} = -7.325$  eV located symmetrically nearby the middle of the band gap as shown in **Figure 1(a)**. The electronic levels of sodium  $\varepsilon_{\text{Na}}$  and chlorine  $\varepsilon_{\text{Cl}}$  of their little impurities in liquid water are shown here too. They are obtained by Equations (1):

$$\varepsilon_{\text{Na}} = \varepsilon_{\text{SHE}} - eE_{\text{Na}} - k_{\text{B}}T \ln \left( \frac{[\text{Na}^0]_{\text{s}}}{[\text{Na}^+]_{\text{s}}} \right) = -3.29 \text{ eV} \quad (2)$$

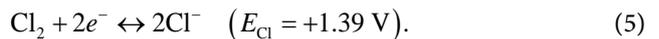
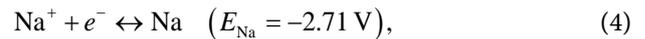
$$\varepsilon_{\text{Cl}} = \varepsilon_{\text{SHE}} - eE_{\text{Cl}} + k_{\text{B}}T \ln \left( \frac{[\text{Cl}^0]}{[\text{Cl}^-]} \right) = -7.7 \text{ eV} \quad (3)$$

where  $k_{\text{B}}$  is Boltzmann constant equal to  $8.617 \times 10^{-5}$  eV/K;  $T$  is Kelvin temperature;  $e$  is the charge of electron;  $[\text{Na}^0]_{\text{s}} = 3.3 \times 10^{-4}$  M is the solubility of neutral sodium atoms in liquid water [1];  $[\text{Na}^+]_{\text{s}}$  is its ionic mole fraction and  $[\text{Cl}^-]$  is the one of chlorine equal to 1.0 M;  $E_{\text{Na}} = -2.71$  V is the standard *ORP* of sodium in the aqueous solution and  $E_{\text{Cl}} = +1.39$  V is the one of chlorine [5];  $[\text{Cl}^0] = [\text{HClO}] = 0.09$  M is the mole fraction of chlorine atoms in the aqueous solution at  $P_{\text{Cl}_2} = 1$  atm,  $\text{pH} = 1$ ,  $T = 300$  K [6].

The electronic levels in Equations (1) and (2) are obtained according to standard electrode reactions [5]:



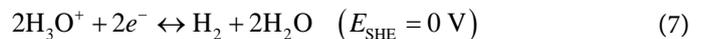
**Figure 1.** The band structure of seawater for the stoichiometric state (a) with Fermi level  $\varepsilon_{Fs}$  (red full line) in the middle of band gap and for the non-stoichiometric ones (b) with Fermi levels  $\varepsilon_{F(4)}$  and  $\varepsilon_{F(5)}$  defined by Equations (4) and (5); the blue boxes are the valence bands with the top  $\varepsilon_v$  and the dotted ones are the conduction bands with the bottom  $\varepsilon_c$ ; the blue full lines denote energy levels:  $\varepsilon_{Na}$ ,  $\varepsilon_{H_3O}$ ,  $\varepsilon_{OH}$ , and  $\varepsilon_{Cl}$  occupied by electron; the dotted ones are these local levels in the vacant state.



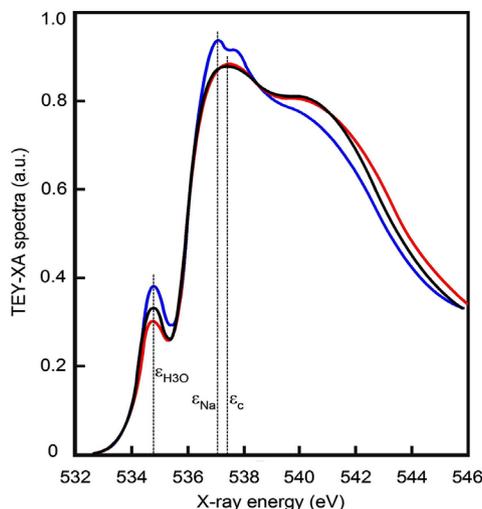
The correspondent Fermi levels  $\varepsilon_{F(4)}$  and  $\varepsilon_{F(5)}$  are shown in **Figure 1(b)** and obtained from Equation

$$\varepsilon_{F(n)} = \varepsilon_{SHE} - eE_{(n)} \tag{6}$$

The Standard Hydrogen Electrode (SHE) is described by the electrode reaction [5]



with the mole fractions:  $[H_3O] = \sqrt{K_{H_2} [H_2]} (P_{H_2} = 1 \text{ atm}) \sim 2 \times 10^{-11} \text{ M}$  and  $[H_3O^+] = 1.0 \text{ M}$  [3]. Here  $K_{H_2} \sim 2 \times 10^{-19} \text{ M}$  is the constant of hydrated dissociation of hydrogen molecule in water up to H-H<sub>2</sub>O. Then in [3], we have obtained  $\varepsilon_{SHE} = -6.21 \text{ eV}$ . These estimations agree with the experimental data [7] shown in **Figure 2** for Total Electron Yield (TEY) of the X-ray Absorption (XA) by water with  $\varepsilon_g = 7.0 \text{ eV}$  and the vacant electron levels:  $\varepsilon_{H_3O}$ ,  $\varepsilon_{Na}$ , and  $\varepsilon_c$  denoted by dotted lines where  $\varepsilon_c$  is the bottom of conduction band. The electron yield of salt NaCl into the identification of  $\varepsilon_{H_3O}$  level is positive due to the synergetic effect of ions  $[Na^+]$  and  $[Cl^-]$  on separating radioactive pairs  $[H_3O^+, OH^-]$  into basic and acidic micro-domains of the salt solution. The electron yield of acid HCl into the same identification is negative due to the inhibitory action of hydroxonium ions  $[H_3O^+]$  onto radioactive formation of pairs  $[H_3O^+, OH^-]$  in the acidic solution.



**Figure 2.** The oxygen K-edge TEY-XA spectra presented by authors of [7] together for pure liquid water (black line), 4 M HCl (red line) and 4 M NaCl (blue line) in aqueous solution; the vertical dotted lines denote the vacant electron levels:  $\varepsilon_{\text{H}_3\text{O}}$ ,  $\varepsilon_{\text{Na}}$ , and  $\varepsilon_c$ .

### 3. The Chemical Stability of Seawater

The population  $[\text{H}_3\text{O}]$  and  $[\text{OH}]$  of the energy levels  $\varepsilon_{\text{H}_3\text{O}}$  and  $\varepsilon_{\text{OH}}$  by electron and hole respectively can be defined by the proportions of the species concentrations  $[\text{H}_3\text{O}^+]/[\text{H}_3\text{O}]$  and  $[\text{OH}]/[\text{OH}^-]$  as shown in [3] [8]. They are given by Maxwell-Boltzmann distribution of electrons and holes in the corresponding energy levels [3]:

$$[\text{H}_3\text{O}^+]/[\text{H}_3\text{O}] = \exp[(\varepsilon_{\text{H}_3\text{O}} - \varepsilon_F)/k_B T] \quad (8)$$

$$[\text{OH}]/[\text{OH}^-] = \exp[(\varepsilon_{\text{OH}} - \varepsilon_F)/k_B T] \quad (9)$$

Using Equations (8) and (9), we can transform the index

$$z = 0.009 \text{ M}^{-1} \{ [\text{H}_3\text{O}] + [\text{H}_3\text{O}^+] - [\text{OH}^-] - [\text{OH}] \} \quad (10)$$

of non-stoichiometric water  $\text{H}_2\text{O}_{1-z}$  saline by NaCl and electrically charged ( $[\text{H}_3\text{O}^+] \neq [\text{OH}^-]$ ) to the form

$$z = 0.009 \text{ M}^{-1} \sqrt{K_w} \left\{ \Delta_{\text{ch}} + \exp\left[\alpha_{\text{ch}} + (\varepsilon_F - \varepsilon_{\text{H}_3\text{O}})/k_B T\right] - \exp\left[-\alpha_{\text{ch}} + (\varepsilon_{\text{OH}} - \varepsilon_F)/k_B T\right] \right\} \quad (11)$$

where  $\Delta_{\text{ch}} = ([\text{H}_3\text{O}^+] - [\text{OH}^-])/\sqrt{K_w}$ ,  $\alpha_{\text{ch}} = \ln(\sqrt{1 + \Delta_{\text{ch}}^2/4} + \Delta_{\text{ch}}/2)$ , and the dissociation constant  $K_w = 10^{-14} \text{ M}^2$  at  $T = 300 \text{ K}$  is defined by the known dissociation ratio [9]

$$[\text{H}_3\text{O}^+] \cdot [\text{OH}^-] = K_w \quad (12)$$

for the mole fractions of hydroxonium  $[\text{H}_3\text{O}^+]$  and hydroxide  $[\text{OH}^-]$  ions.

As seen in Equation (11), the non-stoichiometric index  $z$  of seawater at  $\Delta_{\text{ch}} = 0$  is controlled by  $\varepsilon_F$ . Then, we can present Equation (11) in the form

$$z = 1.68 \times 10^{-24} \{ \exp[38.76\Delta_F/eV] - \exp[-38.76\Delta_F/eV] \} \quad (13)$$

at  $k_B T = 0.0258 \text{ eV}$ ,  $\varepsilon_{\text{H}_3\text{O}} = -5.575 \text{ eV}$ ,  $\varepsilon_{\text{OH}} = -7.325 \text{ eV}$ , and  $\varepsilon_{\text{Fs}} = -6.45 \text{ eV}$  [3] for  $\Delta_F = \varepsilon_F - \varepsilon_{\text{Fs}}$ .

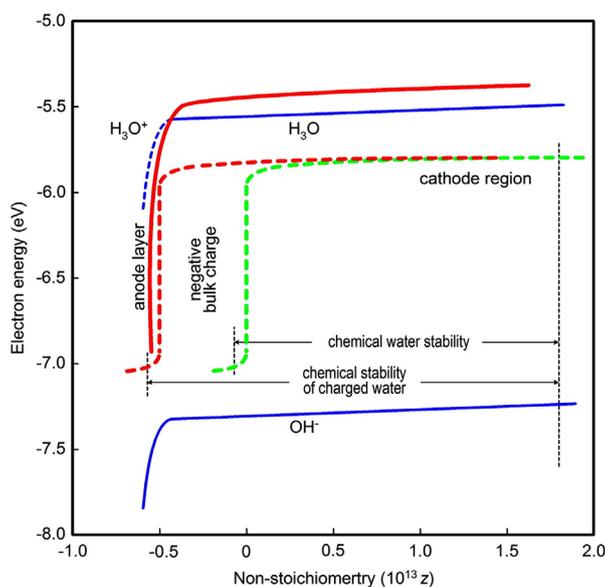
Then, Equation (13) allows plotting Fermi level in seawater as a function of its non-stoichiometry  $z$  shown in **Figure 3** by the green dotted line for  $[\text{H}_3\text{O}^+] = [\text{OH}^-] = 10^{-7} \text{ M}$ ,  $[\text{H}_2] = 1.3 \times 10^{-3} \text{ M}$ ,  $[\text{O}_2] = 2.7 \times 10^{-4} \text{ M}$ ,  $P_{\text{H}_2} = P_{\text{O}_2} = 1 \text{ atm}$ ,  $T = 300 \text{ K}$  [10],  $[\text{H}_3\text{O}] \sim 2 \times 10^{-11} \text{ M}$ , and  $[\text{OH}] \sim 8 \times 10^{-13} \text{ M}$  [3].

## 4. Discussion of the Obtained Results

### 4.1. The Electro-Reduced Seawater

For electrochemical reduction of seawater, one can use the galvanic cell under the voltage of  $\sim 2 \text{ V}$  between the strongly polarized anode and the quasi-equilibrium cathode. In this case, we will always have a negative bulk charge near the polarized anode as shown in **Figure 3**.

One can see that the region of chemical water stability is enlarged to the hyper-stoichiometric state  $\text{H}_2\text{O}_{1-z}$  ( $z < 0$ ). It is easily achieved when  $\varepsilon_{\text{H}_3\text{O}}$  is occupied by electron and hydroxonium radicals  $[\text{H}_3\text{O}]$  are joined to hydroxide anions  $[\text{OH}^-]$  forming in seawater the hydrated electrons  $[(\text{H}_2\text{O})_2^-]$  [11].



**Figure 3.** The changed  $z$ -dependence of Fermi level (red full line) in the band gap (see **Figure 1**) of negatively charged ( $\Delta_{\text{ch}} = -5.0 \times 10^{-5}$ ) seawater (red dotted line) obtained by the electrochemical cell under the voltage of  $\sim 2 \text{ V}$  between the quasi-equilibrium cathode and the strongly polarized anode at  $P = 1 \text{ atm}$ ,  $T = 300 \text{ K}$ ; the green dotted line is the  $z$ -dependence of Fermi level in neutral seawater ( $\Delta_{\text{ch}} = 0$ ); the vertical dotted lines denote the confines of chemical water stability; the blue line denote the local electronic levels  $\varepsilon_{\text{H}_3\text{O}}$  and  $\varepsilon_{\text{OH}}$  for hydroxonium ion/radical ( $\text{H}_3\text{O}^+/\text{H}_3\text{O}$ ) and hydroxide anion  $\text{OH}^-$  respectively.

It is well known [12] [13] that increasing mole fraction of different charges (ions of sodium and chlorine as well hydrated electrons) in seawater blocks the formation of gas hydrates there because these charges shift the equilibrium curve of gas-hydration towards low temperature. At the same time, the hydrated electrons are increasing as kinetic inhibitors of this process that do not change the chemical composition of seawater and can be gotten by its electro-reduction in the mentioned asymmetric electrochemical cell with the polarized anode and the cathode in equilibrium with the aqueous medium.

As seen in **Figure 3**, Fermi level  $\varepsilon_F$  is shifting higher the energy level  $\varepsilon_{H_3O}$  and the hydroxonium ions  $[H_3O^+]$  formed in the anode layer by reaction [1]



forcibly migrate in the bulk of seawater due to the action field between anode and negative bulk charge and are discharged in the cathode region to radicals  $[H_3O]$  by the quasi-equilibrium cathode reaction



These radicals join to hydroxide anions  $[OH^-]$  and form negative equilibrium bulk charges  $[(H_2O)_2^-]$  by reaction



that keep seawater in the stable hypo-stoichiometric state (see **Figure 3**) with the high mole fraction of the hydroxide anions ( $[OH^-] \sim [H_3O] \gg 10^{-7} M$ ) as proton acceptors and the hydroxonium radicals as electron donors in the bulk of seawater.

Thus, electrochemical processing of these very active antioxidants can be more effective than the gaseous hydrogen can do them in the aqueous solution ( $[H_3O] \sim 10^{-10} M$ ) for holding the negative *ORP* of water chemistry by the kinetically-limited reaction of hydrogen dissociation [14]



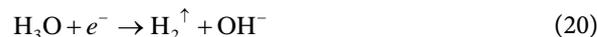
This effect of water processing appears also and in the alternative electrochemical cell with the strongly polarized cathode and the quasi-equilibrium anode as shown that in the next paragraph for comparing with the formation of strong oxidizers  $[OH]$  by the kinetically-limited reaction of oxygen dissociation



## 4.2. The Electro-Oxidized Seawater

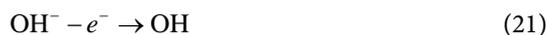
As seen in **Figure 3**, the region of non-stoichiometric composition  $z$  of water ( $H_2O_{1-z}$ ) is limited by the narrow interval  $|z| < 10^{-14}$  where Fermi level is changed in the band gap up to 1.5 eV. Such forced variation of  $\varepsilon_F$  can be carried out by the electrochemical cell with the voltage of  $\sim 2$  V between the strongly polarized cathode and the anode in quasi-equilibrium with medium. The external potential applied to the strongly polarized cathode intensively generated hy-

droxide anions  $[\text{OH}^-]$  in the narrow cathode layer (see **Figure 4**) by standard reactions:



At the same time, Fermi level (red full line in **Figure 4**) is shifting to the energy level  $\varepsilon_{\text{OH}}$  and the hydroxide anions migrate forcedly in the bulk of seawater due to the action of electric field between cathode and the positive bulk charge.

The anions  $[\text{OH}^-]$  are discharged with forming hydroxyls  $[\text{OH}]$  by quasi-equilibrium anodic reaction



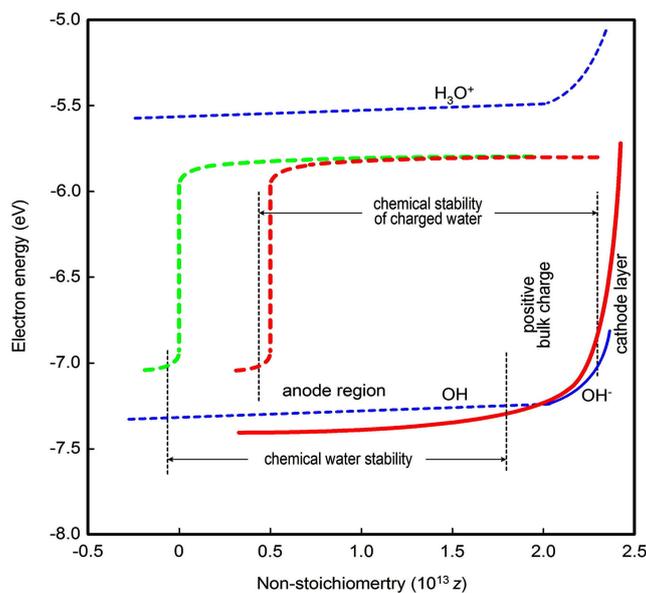
and hydroxonium radicals  $[\text{H}_3\text{O}]$  diffused out of the cathode layer in the bulk of electrochemical cell put electrons to hydroxyl radicals by reaction



All this forms the positive bulk charge  $[(\text{H}_2\text{O})_2^+]$  in seawater near the cathode by reaction



at the condition



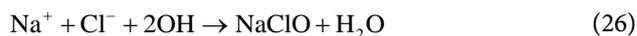
**Figure 4.** The changed  $z$ -dependence of Fermi level (red full line) in the positively charged ( $\Delta_{\text{ch}} = 5.0 \times 10^{-5}$ ) seawater (red dotted line) obtained by the electrochemical cell under the voltage of  $\sim 2$  V between the strongly polarized cathode and the quasi-equilibrium anode at  $P = 1$  atm,  $T = 300$  K; the green dotted line is the  $z$ -dependence of Fermi level in neutral seawater ( $\Delta_{\text{ch}} = 0$ ); the vertical dotted lines denote the confines of chemical water stability; the blue line denote the local electronic levels  $\varepsilon_{\text{H}_3\text{O}}$  and  $\varepsilon_{\text{OH}}$  for hydroxonium ion  $\text{H}_3\text{O}^+$  and hydroxyl/hydroxide species ( $\text{OH}/\text{OH}^-$ ) respectively.



which indicates on the strong acidic reaction of the chemically stable hypo-stoichiometric seawater with the high mole fraction ( $[\text{OH}] \gg 10^{-7} \text{ M}$ ) of hydroxyls as strongest oxidizers. They are able to generate molecules of hypochlorite acid HClO from hydroxyls  $[\text{OH}]$  and chlorine anions  $[\text{Cl}^-]$  by the quasi-equilibrium reaction in the anodic region (see **Figure 4**)



and to joint the salt ions  $[\text{Na}^+]$  and  $[\text{Cl}^-]$  together in the electro-oxidized seawater by reaction [15]



that essentially decreases their mole fraction in aqueous solution and shifts the equilibrium curve of hydrating the neutral species as  $[\text{NaClO}]$  and gaseous molecules towards high temperatures.

The advantages of this approach are the high efficiency and simplicity of oxidation process, low cost, and there is no need for special sorbents because seawater itself becomes the reagent for removing of pollutants.

## 5. Conclusions

The electro-reduced seawater can be easily obtained by galvanic cell under the voltage of  $\sim 2 \text{ V}$  between the strongly polarized anode and the quasi-equilibrium cathode. In this cell, the negative bulk charge is appeared near polarized electrode due to occupation of  $\varepsilon_{\text{H}_3\text{O}}$  by electron and joining of hydroxonium radicals  $[\text{H}_3\text{O}]$  to the hydroxide anions  $[\text{OH}^-]$  in forming hydrated electrons  $[(\text{H}_2\text{O})_2]^-$ .

Then, increasing content of different charges in seawater can block the formation of gas-hydrates there because these charges (as kinetic inhibitors of this process) shift the equilibrium curve of gas-hydration towards low temperatures.

At the same time, the negative bulk charge  $[(\text{H}_2\text{O})_2]^-$  keeps seawater in the stable hypo-stoichiometric state with high mole fraction of hydroxide anions ( $[\text{OH}^-] \sim [\text{H}_3\text{O}] \gg 10^{-7} \text{ M}$ ) as proton acceptors.

Opposite, the hydroxonium radicals  $[\text{H}_3\text{O}]$  as electron donors are very active antioxidants more effective than gaseous hydrogen.

It is shown that the region of non-stoichiometric composition of water ( $\text{H}_2\text{O}_{1-z}$ ) is limited by the very narrow interval  $|z| < 10^{-14}$  where Fermi level is changed in the band gap up to 1.5 eV. At the strong polarization of cathode in the electrochemical cell, the high mole fraction of formed hydroxyls ( $[\text{OH}] \gg 10^{-7} \text{ M}$ ) will essentially decrease content of ions in seawater by forming neutral species  $[\text{NaClO}]$ .

It is shown that the electron yield of NaCl into the identification of energy level  $\varepsilon_{\text{H}_3\text{O}}$  is positive due to the synergetic effect of ions  $[\text{Na}^+]$  and  $[\text{Cl}^-]$  on

separating radioactive pairs  $[\text{H}_3\text{O}^+, \text{OH}^-]$  into basic and acidic micro-domains of seawater. The electron yield of acid HCl into the same identification is negative due to inhibitive action of hydroxonium ions  $[\text{H}_3\text{O}^+]$  onto radioactive formation of pairs  $[\text{H}_3\text{O}^+, \text{OH}^-]$  in the aqueous solution of acid.

### Acknowledgements

The author thanks the Russian Foundation of Basic Research (RFBR) for supporting this work (grant #19-08-00149a) and appreciates his colleagues for active discussing all the aspects of galvanic modifying the chemical properties of seawater.

### Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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