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MORE DETAILS CONCERNING THE USE OF THE THOMSON RADIAL FREQUENCIES $\omega_{Th,s}$, $\omega_{Th,p}$ (OF THE SERIES, RESPECTIVE PARALLEL, CIRCUITS, CONSIDERED INSTEAD OF THE WARBURG PSEUDO CAPACITANCE C_W), AS CRITERIA OF CLASSIFYING THE DRUGS

NICOLAE BONCIOCAT[a] AND ADINA COTARTA[a]

ABSTRACT. In a very recent communication we have shown that the EIS method may be used for classifying the drugs. More precisely it has been shown that to explain the phase difference between the current and the tension it is necessary to use instead of the Warburg pseudo-capacitance $C_W(\omega_1)$, two physically quantities, namely: a pseudo-inductance $L_W(\omega_1)$ and a pseudo-capacitance $C_W(\omega_1)$, which may be arranged in series or in parallel. Consequently, two criteria of classifying the drugs have resulted: for the series arrangement the Thomson radial frequency $\omega_{Th,s} = [L_s(\omega_1)C_s(\omega_1)]^{-1/2}$, respective for the parallel arrangement, the Thomson radial frequency $\omega_{Th,p} = [L_p(\omega_1)C_p(\omega_1)]^{-1/2}$. As for ω_1 it represents the lowest radial frequency used in getting the Nyquist plots(e.g., 1.256 s^{-1}) and has a very important physical meaning: it represents the resonance Thomson radial frequency of both series or parallel circuits, and expresses the highest efficiency that a drug may have, irrespective of the fact that it belongs to the class characterized by the criterion $\omega_{Th,s}(\omega_1)$ or by the criterion $\omega_{Th,p}(\omega_1)$. At the resonance Thomson radial frequency, ω_1 the electric energy of the drug, transforms in its magnetic energy and back in its electric energy and this oscillation between electric and magnetic energies occur permanently with a maximum amplitude. This explains why at the resonance radial Thomson frequency $\omega_{Th,s} = \omega_{Th,p} = \omega_1$ the efficiency of drug is maximum.

Keywords: multielectrode, Thomson radial frequencies, drugs classification

INTRODUCTION

In a series of papers, Bonciocat et al., have shown that the faradaic current density of an electrode redox reaction occuring with combined limitations of charge transfer and nonstationary, linear, semiinfinite diffusion is the solution

[a] *University Politehnica of Bucharest, Department of Applied Physical Chemistry and Electrochemistry, Computer Aided Electrochemistry Laboratory UPB-LEAC, Bucharest, Romania, PO BOX 12-112, e-mail: adina@cael.pub.ro*

of an integral equation of Volterra type[1-6]. By solving this integral equation, new methods of direct and cyclic voltammetry have been developed [7-15]. The above mentioned equation has led to a new approach to the Electrochemical Impedance Spectroscopy (EIS) when only the charge transfer and diffusion limitations are present. Very recently has been shown that the E I S method may have important applications in drug research[16-19].

In this paper, one analysis more deeply the physical meaning of this *resonance* Thomson radial frequency, and how it is related to the *normal* Thomson radial frequencies $\omega_{Th,s}$, respective $\omega_{Th,p}$. Two formulae have resulted, expressing the quantities $\omega_1^2/\omega_{Th,s}^2$, respective $\omega_{Th,p}^2/\left(\omega_{Th,p}^2-\omega_1^2\right)$, in terms of the contributions that a drug may have upon the processes occurring at the electrode/ solution interface. Thus, it may introduce new electrode reactions, or it may change the *Warburg diffusion resistance*, if the drug adsorbs at the electrode/ solution interface, and, in this way, the surface of the electrode changes from A to ${}_iA^*$, or ${}_iA^{**}$. Of course, the drug may introduce new ionic species in the RRD solution, but, because the concentrations of these species are very small in comparison with those of $\left[Fe(CN_6)\right]^{3-}$, $\left[Fe(CN_6)\right]^{4-}$ the effect of these new ionic species may be neglected. Consequently, taking into account only the two effects that a drug may have, i.e., the change of both, charge transfer and Warburg diffusion resistances, Bonciocat and Adina Cotarta have proposed the following expressions for the two quotients $\omega_1^2/[\omega_{Th,s}^2]_0$, respective :$[\omega_{Th,p}^2]_0/[\omega_{Th,p}^2]_0-\omega_1^2$:

$$J^*=\frac{\omega_1^2}{\left[\omega_{Th,s}\right]_0^2}=\frac{\left(A_{ct}\right)_{RRD}-\left({}_iA_{ct}^*\right)_{(ME)_{Di}^*}}{\left[R_W(\omega_1)\right]_{RRD}\cdot\frac{A}{\left({}_iA^*\right)}} \qquad (1*)$$

respective:

$$J^{**}=\frac{\left[\omega_{Th,p}\right]_0^2}{\left[\omega_{Th,p}\right]_0^2-\omega_1^2}=\frac{\left(A_{ct}\right)_{RRD}-\left({}_iA_{ct}^{**}\right)_{(ME)_{Di}^{**}}}{\left[R_W(\omega_1)\right]_{RRD}\cdot\frac{A}{\left({}_iA^{**}\right)}} \qquad (1**)$$

where the Thomson radial frequencies $[\omega_{Th,s}]_0$ and $[\omega_{Th,p}]_0$ have the meanings:

$$\left[\omega_{Th,s}\right]_0=\left\{\omega_{Th,s}\left[\alpha_D^*(\omega_1)\right]\right\}_{\eta(0)=0}=\frac{\omega_1}{\sqrt{1-\alpha_D^*(\omega_1)}}$$

$$\left[\omega_{Th,p}\right]_0 = \left\{\omega_{Th,p}\left[\alpha_D^{**}(\omega_1)\right]\right\}_{\eta(0)=0} = \omega_1\sqrt{1-\alpha_D^{**}(\omega_1)} \qquad (2*)$$

and:

$$0 \le \alpha_D^{*}(\omega_1) \le 1; \qquad 0 \le \alpha_D^{**}(\omega_1) \le 1 \qquad (3*)$$

In this paper one shows that J^* and J^{**} represent two criteria very useful for a better and more correct classification of the drugs which belong to the classes (I^*), respective (I^{**}).

THEORETICAL SECTION

Introduction

The quotients (1^*) and (1^{**}) permit to consider the following situations:

A) $D_i \in$ (I), i.e., has no effect. Then:

$$\frac{\omega_1^2}{\left[\omega_{Th,s}\right]_0^2} = \frac{\left[\omega_{Th,p}\right]_0^2}{\left[\omega_{Th,p}\right]_0^2 - \omega_1^2} = 0 \qquad (4*)=(4**)$$

which means:

either $\left[\omega_{Th,s}\right]_0 = \left\{\omega_{Th,s}[\alpha_{D_i}^{*}(\omega_1)=1]\right\}_{\eta(0)=0} = \infty$

or $\left[\omega_{Th,p}\right]_0 = \left\{\omega_{Th,p}[\alpha_D^{**}(\omega_1)=1]\right\}_0 = 0 \qquad (5*)=(5**)$

B) $D_i \in$ (I^*) and has effect if:

$$J^* = \frac{\omega_1}{\left[\omega_{Th,s}\right]_0} = \frac{\omega_1}{\left\{\omega_{Th,s}\left[1 \rangle \alpha_D^{*}(\omega_1) \rangle 0\right]\right\}_0} =$$ finite and positive value less than 1, and:

$D_i \in$ (I*) has a maximum effect if:

$$J^* = \frac{\omega_1}{[\omega_{Th,s}]} = \frac{\omega_1}{\left\{\omega_{Th,s}\left[\alpha_D^{*}(\omega_1)=0\right]\right\}_0} = 1 \qquad (6*)$$

C) $D_i \in$ (I^{**}) and has effect if:

$$J^{**} = \frac{\left[\omega_{Th,p}\right]_0^2}{\left[\omega_{Th,p}\right]_0^2 - \omega_1^2} = \frac{\left\{\omega_{Th,p}[1 \rangle \alpha_D^{**}(\omega_1) \rangle 0]\right\}_0^2}{\left\{\omega_{Th,p}[1 \rangle \alpha_D^{**}(\omega_1) \rangle 0\right\}_0^2 - \omega_1^2}$$ =finite and negative value (7*)

and

$D_i \in (I^{**})$ has a maximum effect, if :

$$J^{**} = \frac{\left[\omega_{Th,p}\right]_0^2}{\left[\omega_{Th,p}\right]_0^2 - \omega_1^2} = \frac{\left\{\omega_{Th,p}\left[\alpha_D^{**}(\omega_1) = 0\right]\right\}_0^2}{\left\{\omega_{Th,p}\left[\alpha_D^{**}(\omega_1) = 0\right]\right\}_0^2 - \omega_1^2} \rightarrow - \infty \qquad (8^{**})$$

Therefore, from (5^{*}) and (6^{*}), respective (7^{*}) and (8^{**}), it follows:

$$0 \le J^{*} \le 1 \qquad (9*)$$

respective:

$$-\infty \le J^{**} \le 0 \qquad (9**)$$

Anti -oxidizer and Pro -oxidizer drugs

The inequalities (9∗) and (9∗∗) show that the pair (J^{*} , J^{**}) satisfies the necessary conditions for being a pair of criteria of classifying the drugs, because the values of J^{*} are greater than zero, while those of J^{**} are less than *zero.* The class of drugs characterized by the criterion J^{**} will be a sub-class of the class (I^{**}), while the class of drugs characterized by the criterion J^{*}, will be a sub-class of the class (I^{*}).

We shall write these two subclasses by $\left(J_{A.O}^{**}\right)$, respective $\left(J_{P.O}^{*}\right)$, because, as we shall see further, in these sub-classes enter the *anti-oxidant* drugs (i.e., A.O-drugs), respective the *pro-oxidant* drugs (i.e., P.O-drugs). Thus, $(J_{A.O}^{**}) \subset (I^{**})$, while $(J_{P.O}^{**}) \subset (I^{*})$.

The most important conclusion concerning the Theoretical Section:

- A drug $D_i \in (I^{*})$ is characterized by the radial Thomson frequency $\omega_{Th,s}[\alpha_{D_i}^{*}(\omega_1) = 0]$ and has a maximum effect if his radial Thomson frequency is equal to the *resonance* Thomson frequency

$$res\ \omega_{Th,s}\ [\alpha_{D_i}^{*}(\omega_1) = 0] = \omega_1\ ;$$

- A drug $D_i \in (I^{**})$ is characterized by the radial Thomson frequency $\omega_{Th,p}\,[\alpha_{D_i}^{*}(\omega_1) = 0]$ and has a maximum effect if his radial Thomson frequency ω is equal to *the resonance* Thomson frequency

$$\text{res}\ \omega_{Th,p}\ [\alpha_{D_i}^{**}(\omega_1) = 0] = \omega_1.$$

- An A.O- drug is characterized by the criterion $J_{A.O}^{**}$, and has a maximum effect, if his $J_{A.O}^{**}$ criterion has the value - ∞.

- An P.O –drug is characterized by the criterion $J_{P.O}^{*}$, and has a maximum effect, if his $J_{P.O}^{*}$ criterion has the value 1.

EXPERIMENTAL

In the paper hold at Journees d'Electrochimie 2009, Sinaia (6-10 Jouillet), Roumanie, we have investigated 6 drugs and 2 mixtures of drugs, namely:

B (Sweedish Bitter), Am (Achillea Millefolium), Cf_1 and Cf_2 (Calendula floss), Uh_1 and Uh_2 (Urticae herba), M_1 = Uh + Am, M_2 = B + Am + Cf + Uh[1].

By using eqs.(37') and (37'') [1], where $Re^{**}(\omega_1)$, $Re^{**}(\omega_2)$ represent the abscissa of the first two points of the Nyquist plots(i.e., corresponding to ω_1=1.256s^{-1} and ω_2=1.582s^{-1}) recorded for the drugs and mixtures of drugs investigated, while $Re(\omega_1)$, $Re(\omega_2)$ represent the abscissa of the first two points of the Nyquist plots, recorded for the RRD-dielectrode, it was possible to get the values of the quantities a^{**} and b^{**}[1]. Because the values thus obtained have proved to obey all the inequality $a^{**} \langle b^{**}$, it results that all drugs and mixtures of drugs investigated belong to the class (I^{**}), i.e., in their electrical schemes, the Warburg pseudo-capacitance $C_W(\omega)$ must be replaced by a parallel circuit of a pseudo-inductance $L_W(\omega)$, in parallel with a pseudo-capacitance $C_W(\omega)$. Once the values a^{**} and b^{**} known, the radial Thomson frequency $\left\{\omega_{Th,p}[\alpha_D^{**}(\omega_1)=0]\right\}_0 = [\omega_{Th,p}]_0$, have resulted by means of eq.(37) [1].

In Table 1 is shown, in *details,* the procedure by which have been obtained the values of a^{**} , b^{**} and $[\omega_{Th,p}]_0$ in the case of the drug B.

$$a^{**} = 1.122\,[\mathrm{Re}(\omega_2) - \mathrm{Re}^{**}(\omega_2)]; \qquad b^{**} = \mathrm{Re}(\omega_1) - \mathrm{Re}^{**}(\omega_1)$$

(37') and (37'')[1]

$$\left[\omega_{Th,p}\right]_0 = \left\{\frac{-a^{**}+b^{**}}{1.121\left[\mathrm{Re}^{**}(\omega_1) - \mathrm{Re}^{**}(\omega_2)\right] - a^{**}+b^{**}}\right\}^{1/2} \cdot \omega_1$$

(37)[1]

Further, the procedure exemplified in Table 1, has been used for obtaining the values $[\omega_{Th,p}]_0$, corresponding to all drugs and mixtures of drugs investigated. The results are given in Table 2, where for sake of simplicity, are given the multielectrods containing the respective drugs or mixtures of drugs, and the mean values of $[\omega_{Th,p}]_0$ obtained from 4 experimental values.

Table 1. Example of obtaining the values a^{**} and b^{**} (by means of eqs.(37') and (37'')) and of the values of $[\omega_{Th,p}]_0$ (by means of eq.(37))[1]

We mention that $[\omega_{Th,p}]_0$ is the mean value of the 4 values $\left\{\omega_{Th,p}[\alpha_D^{**}(\omega_1)=0]\right\}_0$ obtained by applying eq.(37)[1].

Dielectrode RRD V= 300mL	Re(ω_1) (Ω)	Re(ω_2) (Ω)	
	319 329 318 320	291 291 289 290	

Multi electrode	$\mathrm{Re}^{**}(\omega_1)$ (Ω)	$\mathrm{Re}^{**}(\omega_2)$ (Ω)	a^{**} (Ω)	b^{**} (Ω)	$\left\{\omega_{Th,p}[\alpha_D^{**}(\omega_1)=0]\right\}_0$ (s^{-1})
$(ME)_B$ V=300mL V=20mL	10447 10936 11688 12721	9944 10409 11125 12266	-10830 -10761 -12158 -13370	-10128 -10614 -11370 -12401	0.935 0.561 0.936 0.994

Table 2. The Thomson radial frequencies $[\omega_{Th,p}]_0 = \left\{\omega_{Th,p}[\alpha_D^{**}(\omega_1)=0]\right\}_0$ estimated by using eq.(37)[1]

Multielectrode	$[\omega_{Th,p}]_0$ Mean values	Multielectrode	$[\omega_{Th,p}]_0$ Mean values
$(ME)_B$ V= 300mL v= 20mL	0.857	$(ME)_{Uh}$ V= 300mL v= 20mL	0.949
$(ME)_{Am}$ V= 300mL v= 30mL	0.823	$(ME)_{Uh2}$ V= 300mL v= 30mL	1.002

Multielectrode	$[\omega_{Th,p}]_0$ Mean values	Multielectrode	$[\omega_{Th,p}]_0$ Mean values
$(ME)_{Cf_1}$ V= 300mL v= 20mL	1.067	$(ME)_{Uh+Am}$ V= 300mL v=20mL+20mL	0.898
$(ME)_{Cf_2}$ V= 300mL v= 30mL	0.874	$(ME)_{B+Am+Cf+Uh}$ V= 300mL v=(10+10+10+10)mL	0.954

Because all drugs and mixtures of drugs investigated have proved to belong to the class (I**), it is important to decide if they are AO-drugs, i.e., if they belong to the sub-class $J^{**}_{A.O} \subset \left(I^{**}\right)$.

Therefore it is necessary to estimate the values of the criterion:

$$J^{**} = \frac{[\omega_{Th,p}]_0^2}{[\omega_{Th,p}]_0^2 - \omega_1^2}$$

for these 6 drugs and 2 mixtures of drugs.

The corresponding values are given in Table 3.

As one sees in Table 3, all the values of the criterion J** are negative, and belong to the interval (9**). Consequently, *the most important conclusions of the Experimental Section are:*

- All the drugs and mixtures of drugs investigated, belong to the sub-class ($J^{**}_{A.O} \subset \left(I^{**}\right)$), i.e., are A.O-drugs.
- The drug Cf_1 has the maximum effect, because $J^{**}_{Cf_1}$ = -2.586, i.e., has the most negative value. This means that this drug acts by changing both, the charge transfer resistance and the Warburg diffusion resistance, i.e., this drug *adsorbs significantly* at the electrode/solution interface. This adsorption process explains also why, by increasing the concentration of the drug, i.e., by passing from Cf_1 to Cf_2 the value of $\left|J^{**}_{Cf}\right|$ decreases from +2.586 to +0.939 because the quotient A/ $_i A^{**}$ increases (see eq.(1**)).
- In the case of the drug Uh, by increasing its concentration, the value of $\left|J^{**}_{Uh}\right|$ increases too, from 1.331 to 1.749. This may be explained by the fact that the drug Uh acts *firstly by changing the charge transfer resistance*, which, of course, *increases* if the concentration of drug *increases.*

Table 3. The values of the criterion $J^{**} = [\omega_{Th,p}]_0^2 / \left\{[\omega_{Th,p}]_0^2 - \omega_1^2\right\}$ for the investigated drugs and mixtures of drugs

Drugs and mixtures of drugs	$[\omega_{Th,p}]_0^2$ (s^{-2})	ω_1^2 (s^{-2})	J^{**}
B	0.901	1.578	-1.331
Am	0.677	1.578	-0.751
Cf_1	1.138	1.578	-2.586
Cf_2	0.764	1.578	-0.939
Uh_1	0.901	1.578	-1.331
Uh_2	1.004	1.578	-1.749
Uh+Am	0.806	1.578	-1.044
B+Am+Cf+Uh	0.910	1.578	-0.247

This two effects (i.e., changing of the charge transfer resistance and of the Warburg diffusion resistance may compensate each other, and this may explain why the values $\left|J^{**}\right|$ corresponding to the two mixtures of drugs investigated are not far away from the values $\left|J^{**}\right|$ corresponding to their individual components.

It remains to explain how an A.O-drug acts. Let's start by remembering the reactions of the RRD-dielectrode. They are:

$\frac{1}{4}O_2 + H^+ + e \rightarrow \frac{1}{2}H_2O$ cathodic reaction

$[Fe(CN)_6]^{4-} \rightarrow [Fe(CN)_6]^{3-} + e$ anodic reaction

The electrons resulted by the oxydation of $[Fe(CN)_6]^{4-}$ are consumed by the reduction of the physically dissolved oxygen and H^+-ions (i.e., $\frac{1}{4}O_2 + H^+$) to $\frac{1}{2}H_2O$.

Therefore, $\frac{1}{4}O_2 + H^+$ acts as an *oxidizer* of $[Fe(CN)_6]^{4-}$. It thus results that an A.O- drug **reduces** the oxidating effect of $\frac{1}{4}O_2 + H^+$, *by consuming a part, or all electrons resulted by the oxidation* of $[Fe(CN)_6]^{4-}$ to $[Fe(CN)_6]^{3-}$.

This consuming of electrons may take place, either by a *chemical* reaction in solution, e.g.,

$$(A.O-D)_{\mathrm{Re}d}+\frac{1}{4}O_2+H^+\rightarrow(A.O-D)_{Ox}+\frac{1}{2}H_2o$$

when the A.O-drug (see A.O-D) passes from its reduce form, to its oxidize form, when at the electrode / solution interface, occur three electrode reactions, e.g.,

$$\frac{1}{4}O_2+H^+ + e\rightarrow\frac{1}{2}H_2O$$
$$[Fe(CN)_6]^{4-}\rightarrow[Fe(CN)_6]^{3-}+e$$
$$[A.O-D]_{\mathrm{Re}d}\rightarrow[A.O-D]_{Ox}+e$$

In both ways of action, the important effect of the A.O - D is the reduction of the intensity of the *important oxydation reaction.*

If this important *oxidation* is a biological reaction, responsible for a very dangerous illness, the A.O-drugs may have *very important therapeutic applications*, in fighting against this dangerous illness.

As for the P.O-drugs, there are two ways in which they may act.

A first possibility is to act chemically, i.e., by a reaction in solution, e.g.,

$$(A.O-D)_{Ox}+\frac{1}{2}H_2O\rightarrow(A.O-D)_{\mathrm{Re}d}+\frac{1}{4}O_2+H^+$$

increasing the concentration of the $\frac{1}{4}O_2+H^+$ and in this way *increasing* the intensity of the oxidation reaction:

$[Fe(CN)_6]^{4-}\rightarrow[Fe(CN)_6]^{3-}+e$

or electro-chemically when at the electrode / solution interface, occur the reactions:

$$\frac{1}{4}O_2+H^+ + e\rightarrow\frac{1}{2}H_2O$$
$[Fe(CN)_6]^{4-}\rightarrow[Fe(CN)_6]^{3-}+e$
$$[P.O-D]_{\mathrm{Re}d}\rightarrow[P.O-D]_{Ox}+e$$

when, a part of the electrons necessary to the first cathodic reaction come from the de-electronation (i.e., oxydation) of the P.O-D- drug.

In both ways of action, the important effect of a P.O.-drug is the *increase* of the intensity of the important oxydation reaction.

CONCLUSIONS

Because the efficiency of an A.O-drug increases if the value of $\left| J^{**} \right|$ increases, it results the following sequence concerning the efficiencies of the 6 drugs and 2 mixtures of drugs (all A.O-drugs) investigated:

$$\left| J^{**}_{Am} \right| < \left| J^{**}_{B} \right| < \left| J^{*\cdot *}_{C f_2} \right| < \left| J^{**}_{Uh+Am} \right| < \left| J^{**}_{Uh_1} \right| < \left| J^{**}_{B+Am+Cf+Uh} \right| < \left| J^{**}_{Uh_2} \right| < \left| J^{**}_{Cf_1} \right|$$

Finally, it is very important to underline that, presently, are known many oxidation reactions, occurring in biological systems, about which one supposes that are at the origin of many illnesses, and for this reason, the development of experimental methods for estimating the therapeutic efficiencies of the A.O.-drugs represents the most important aim of the future researchers in the domain of biological and pharmaceutical sciences.

From this point of view, one may conclude that the two EIS methods presented at Journees d'Electrochimie 2009 and RICCCE 2009, have the necessary conditions for playing a very important role for a *scientific classification of drugs*, produced by the *chemical industry*, or taken from the **God pharmacy** [i.e., by an *adequate transforming of the medicinal plants*].

Unfortunately, such an action necessitates about 4-5 years, and a team of researchers in which must enter: electrochemists, biochemists, physicist and specialists in medicinal plants. Although such an action implies so many difficulties, it must be done, because the results that will be obtained, will justify the efforts, and what it is much more important, will decide the directions in which the future developments of the medical sciences in Romania must be made. This also explains the importance of the criteria $[\omega_{Th,s}]$, $[\omega_{Th,p}]_0$, respective $J^{*}_{P.O}$, $J^{**}_{A.O}$, for the future development of drug-sciences not only in Romania, but also in the world.

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