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INFLUENCE OF NON-IONIZING ELECTROMAGNETIC FIELDS ON REDOX SYSTEMS IN SOLUTIONS

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Abstract

This paper presents the experimental results obtained during testing in an electromagnetic field with non-ionizing radiations from low-intensity microwaves acting on a group of substances consisting of: double distilled water, neutralized to pH = 7; drinking water with pH = 7.3; slightly acidic aqueous solutions obtained by diluting the acetic acid at concentrations of 2.25%, 4.5% and 9% in double distilled water, for pH = 6.8, 6.7 and 6.5, respectively. The exposure of the samples was performed in a resonance cavity at 700 MHz in successive periods from 10 seconds to 5 minutes. After each period the evolution of redox potential was determined. Three samples were taken for each experiment, with a volume of 2 mL/sample and initial temperatures of: 19 °C, 21 °C and 22 °C, which represent temperature variations of the ambient environment. Experimental diagrams were established for each sample, based on which related analytical functions describing the evolution rate of the redox potential were identified, which showed that: (i) distilled water with neutral pH showed no significant influences during exposure; (ii) the redox potential in drinking water varies mainly due to temperature variation in the resonance cavity; (iii) The evolution of the redox potential variation is linear for samples containing water.

For weak acid solutions, the variation of the redox potential is determined both by the thermal effects in the resonance cavity, dominant in the first part of the exposure, as well as the effects of radiation, dominant in the last part of the exposure, the diagrams for the process being nonlinear.

Key words: electromagnetic field, polynomial fit, redox potential

Received: March, 2011; *Revised final:* July, 2012; *Accepted:* July, 2012

1. Introduction

Studies on the effect of the electromagnetic field on living organisms gain greater significance as the number of equipment for communications continues to develop and grow (Miclaus and Calota, 2010). Most studies are usually targeted towards the low frequency range (50Hz, 60Hz) and the frequencies encountered in mobile communications (EUREKA, 2003; Valentine, 1995). Thus, the influence of the low frequency electromagnetic field is largely approached (Di Loreto et al., 2009), for example in terms of redox reactions in mice brain (Jajte and Zmyslony, 2006) and also regarding some

biological and chemical reactions in water (Vallée et al., 2005). The effect of the electromagnetic field with a frequency of 900MHz (a frequency close to that discussed in this paper) on the chemical processes in the brain of guinea pigs was discussed by Meral et al. (2007).

This paper focuses on the physical and chemical changes produced by an oscillating electromagnetic field of low intensity on aqueous solutions with a frequency of 700 MHz (the exact frequency used was 709.5 MHz) in terms of redox potential (ICNIRP, 1998). The choice for this frequency band was motivated by the fact that it is used for VHF TV systems and it is in the proximity

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of the 900 MHz band, used for mobile communications.

The present article is part of a larger research project which concerns high frequency (700 – 900 MHz) radiant field and influences on pH modification in aqueous solutions (Morariu et al., 2009). The aim of research in this field is to identify areas of interest regarding the influences of low intensity high frequency radiation on the natural state of various substances and in order to support competent authorities to formulate security norms and measures.

2. Experimental study

2.1. Materials and methods

Experimentally, the following samples were analyzed for the aqueous solutions (the sample volume is 20 mL for each experiment):

- double distilled water (pH=7);
- slightly alkaline drinking water (pH=7.3);
- slightly acidic aqueous solutions (acetic acid in water with concentrations of 2.25‰, 4.5‰, and 9‰), with the pH values of 6.8, 6.7, and 6.5, respectively, used in the food industry.

The redox potential was measured relative to SHE+197, electrode Ag/AgCl sat KCl. The samples were placed in a cylindrical resonance cavity (ICNIRP, 1998) at 709.5 MHz (TM₁₁₀ oscillation mode) in order to avoid external perturbations, for different time intervals from 10 seconds to 5 minutes, with the growth rate of 10 seconds, each measurement being repeated 3 to 10 times.

Different measurements were performed for the following parameters: time of exposure; sample temperature; radiation power; redox potential. The architecture of the measurement line for a given period of irradiation is presented in Fig. 1.

The elements of the working stand are as follows:

1. microwave frequency generator, with programmable working time and transient limiter with a bandwidth of 600 - 750 MHz;
2. microwave wattmeter;
3. examined sample from resonance cavity;
4. cylindrical plane resonance cavity;
5. IR camera for detection of the sample temperature;
6. redox probe;
7. redox meter.

The power gain of the cavity, depending on frequency, after the sample was fixed in optimal position, is presented in the diagram of Fig. 2. Thus, the experiment could be run at a frequency around 700 MHz, which is used by television systems and is situated in the proximity of the 900 MHz band of mobile networks. The effective power for the experiments was set to 0.55 W. The additional power consumption of the samples for the entire duration of experiment was less than 35 mW (ANSI/IEEE, 1999), (NRPB, 2004) and the energy consumption was 6.5 J.

2.2. Experiments and results with samples of distilled and drinking water

Three samples of distilled water were used for performing the tests in the resonance cavity, with the initial temperatures of 19°C, 21°C, and 22°C. During experiments running, the evolution of the temperature was recorded for each irradiated sample (Zmirou, 2001). For an overall evaluation of the process, the average temperature evolution was obtained in a similar manner from the three samples; the results are presented in Fig. 3 together with the analytical equation for approximating the evolution trajectory of the process (Fig. 3b) (Eq. 1):

$$T_{di} = 1.3E^{-7} \cdot t^2 - 0.003 \cdot t + 20.76 \quad (1)$$

where T_{di} represents the evolution in time of the temperature for the samples containing distilled water.

The temperature variation in the samples exposed to the microwave radiation phenomenon involves small variations in the radiation power (Sheppard and Swicord, 2002). The averaging of the variable power characteristics within the temperature range previously determined for the three samples is shown in Fig. 4 along with analytical equation for approximating the evolution trajectory of the process (Eq. 2):

$$P_{di} = 3.9E^{-9} \cdot t^3 - 2E^{-6} \cdot t^2 - 4.3E^{-4} \cdot t + 0.523 \quad (2)$$

where P_{di} represents the evolution of power over time for the samples containing distilled water and is expressed in W .

The redox processes have developed in the samples, in the context of the evolution of temperature parameters – irradiation power and the simultaneous variation of these two parameters. The evolution of these redox processes is shown in Fig. 5a. Interpolating the three samples of distilled water, the variation of the redox process was obtained; its characteristic is shown in Fig. 5b and the analytical equation for approximating the corresponding evolution trajectory is also represented, according to Eq. (3):

$$V_{di} = 0.001 \cdot t^2 - 0.36 \cdot t + 175.5 \quad (3)$$

where V_{di} (mV) represents the evolution of redox potential over time for the samples containing distilled water.

The evolution characteristic of the redox process tends towards a stable equilibrium at long exposure. This phenomenon is highlighted by a decrease in the redox potential for shorter periods of exposure (up to 200 seconds) after which it tends to revert to a value close to the initial value, remaining in a quasi-stable equilibrium (NEXUS, 1995). Fig. 5c presents the average of redox potential (of distilled water) variation, as a temperature function, for an exposure of 300 s and under the influence of radiant magnetic field.

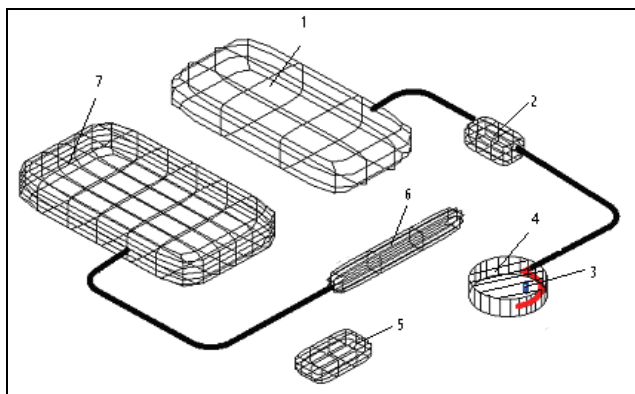


Fig. 1. The block diagram of the work stand

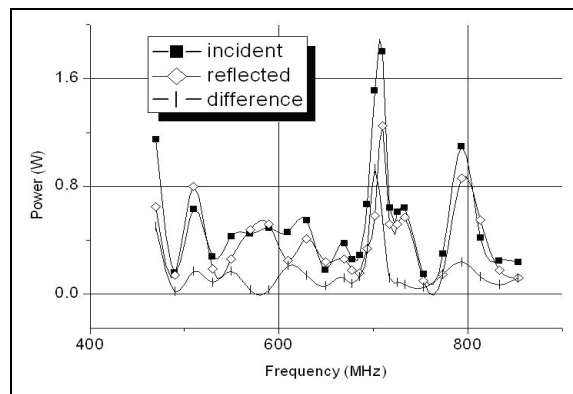


Fig. 2. Characteristic frequency of the resonance cavity in fundamental mode

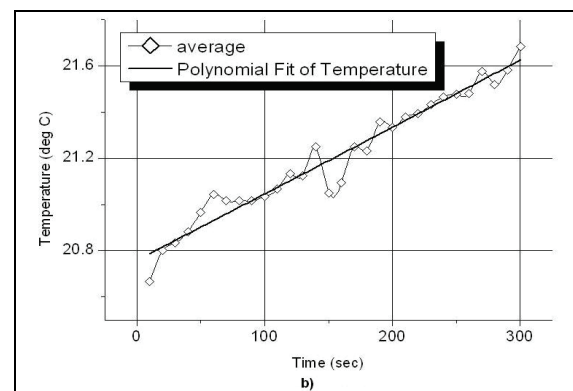
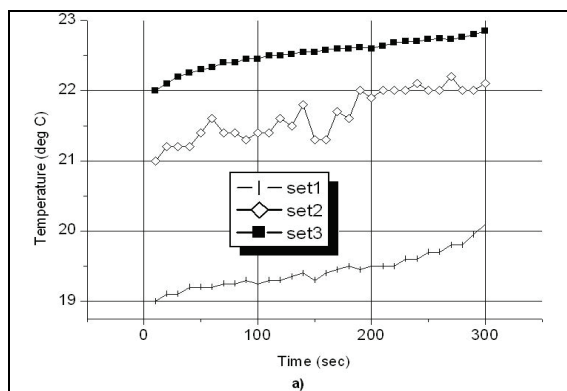


Fig. 3. Temperature variation with distilled water during the irradiation process: a) in the samples; b) average

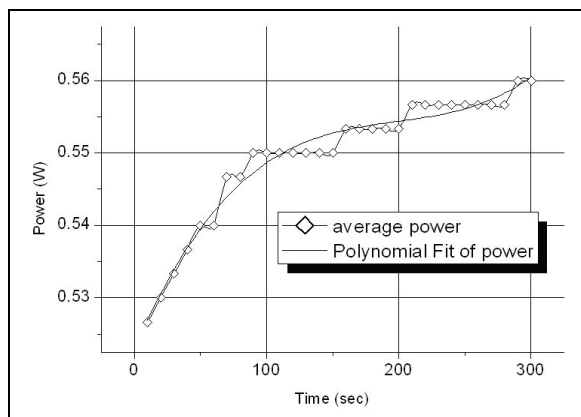
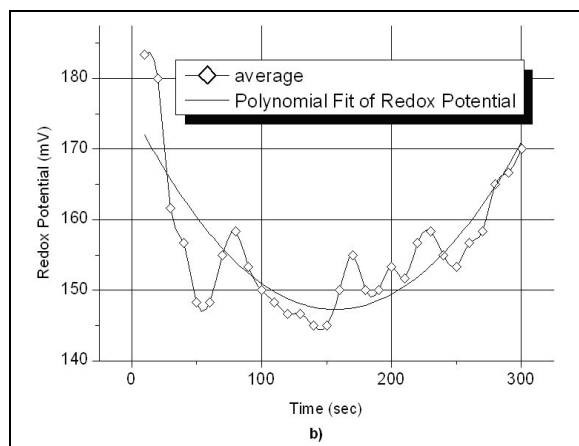
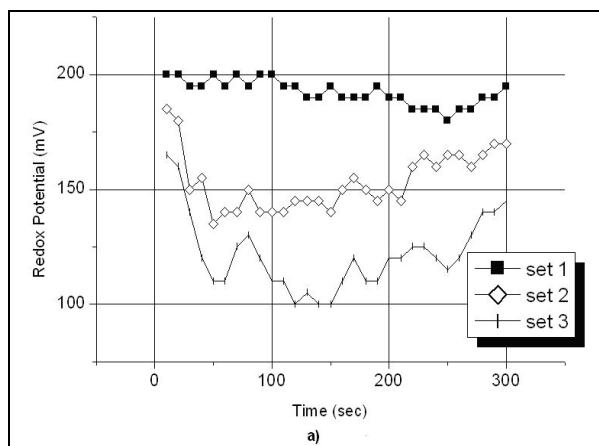


Fig. 4. Average power evolution for distilled water



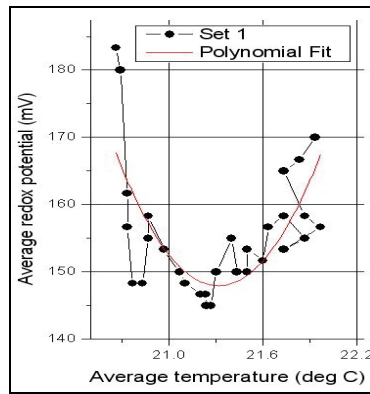


Fig. 5. Evolution of the redox processes in distilled water under the irradiation process: a) for the three samples; b) average; c) Average redox potential as a function of temperature for distilled water

From this experimental graph, it can be observed that there are differences between the experimental data for the radiant field and the graphical representation of the equation for non radiant field defined by Nerst ($E_h = E_{red}^\theta - \frac{R \cdot T}{z \cdot F} \cdot \ln Q$, a quasi-linear function for a narrow temperature domain). The redox potential average tends to gradually re-gain the initial level and stabilize. Under the same test conditions as for distilled water, three samples of drinking water with pH 7.3 were tested in the resonance cavity, with the initial temperatures of 19°C, 21°C, and 22°C, both for cavity and solution. During experiments running, the evolution of the temperature variation for each sample was recorded (Zmirou, 2001) and the results are presented in Fig. 6 together with the average temperature evolution for the samples (Eq. 4):

$$T_{di} = 1.3E^{-6} \cdot t^2 - 0.005 \cdot t + 20.78 \quad (4)$$

where T_{dr} represents the evolution of temperature over time for the samples containing drinking water.

The diagrams in Fig. 7 present the average power variation (similar to that found by Sheppard and Swicord, 2002) within the temperature range for the three samples (Eq. 5):

$$P_{dr} = 5.6E^{-10} \cdot t^3 - 3.66E^{-7} \cdot t^2 - 1.65E^{-4} \cdot t + 0.53 \quad (5)$$

where P_{dr} represents the evolution of power over time for samples containing drinking water.

Simultaneously with the evolution of the temperature parameters, others such as: irradiation power, redox processes have developed in the samples. The evolution of these redox processes is presented in Fig. 8a. Interpolating the results of three samples for drinking water, the variation of the redox process was obtained, as shown in Fig. 8 b (Eq. 6):

$$V_{di} = 0.002 \cdot t^2 - 0.85 \cdot t + 135.4 \quad (6)$$

where V_{dr} represents the evolution of redox potential over time for samples containing drinking water.

Note that drinking water with a pH of approximately 7.3 follows the evolution profile of the redox equilibrium similar to that of distilled water,

but with an important distinction, namely, that the phenomenon of returning to an initial stable equilibrium is not immediate, even after the removal of the irradiation field (Fig. 8b).

Fig. 8c shows the variation of redox potential in of drinking water as a function of temperature under electromagnetic radiation with 300s exposure. The following aspects can be noted: the experimentally obtained graph is deformed when compared to the normal graph for redox potential as a temperature function in non-radiant field, as defined by the Nerst equations. It seems to be a trend towards equilibrium, but in this case the trend is much more subtle than for distilled water (Fig. 5 c). According to Eqs. 3 and 6, the variation rates of the redox potentials for distilled and drinking water, represented in Fig. 9, can be expressed through derivation as given by Eqs. (7-8):

$$V_{di} = \frac{V_{di}}{dt} = 2 \cdot (0.001 \cdot t - 0.18) \quad (7)$$

$$V_{di} = \frac{V_{di}}{dt} = 4 \cdot (0.001 \cdot t - 0.213) \quad (8)$$

Comparing and combining the two rates, it can be found the expression for the evolution of redox potential over time for the samples containing distilled water (Eq. 9):

$$\frac{V_{di}}{dt} \cong \frac{1}{2} \quad (9)$$

The difference in the temperature evolution of the two samples, namely distilled water and slightly alkaline drinking water (Eqs. 1 and 4) is presented in Fig. 10, which shows that:

- the evolution profiles of temperature for distilled water and for drinking water are similar;
- during the entire duration of the experiment, the temperature in the sample of distilled water remained identical to the cavity temperature, whereas the temperature of drinking water differs from that of the cavity, due to the fact that it contains traces of impurities.

The drinking water and the distilled water samples followed a quasi-linear (with different trajectories) increase with the duration of exposure.

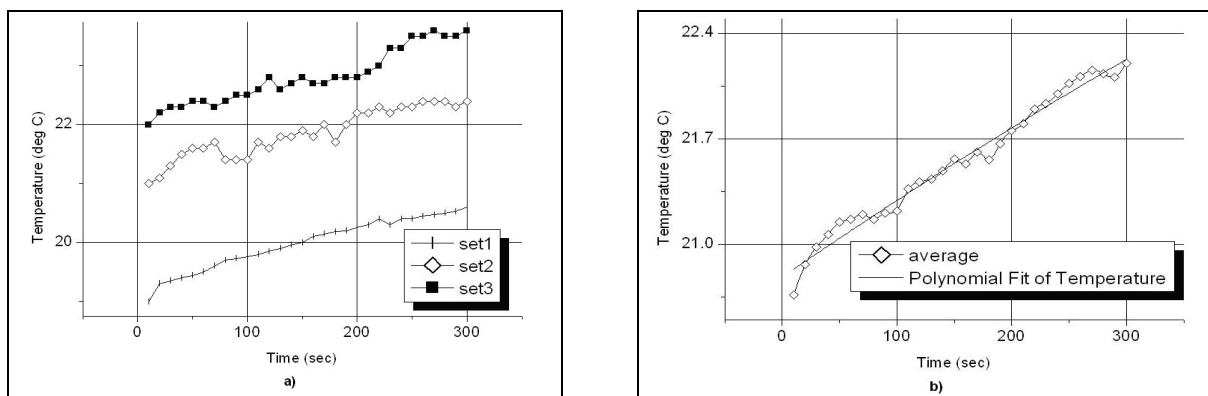


Fig. 6. Temperature variation with drinking water during the irradiation process: a) in the samples; b) average

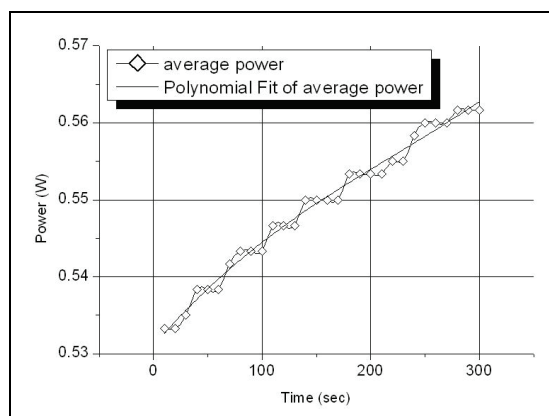


Fig. 7. Average power evolution for drinking water

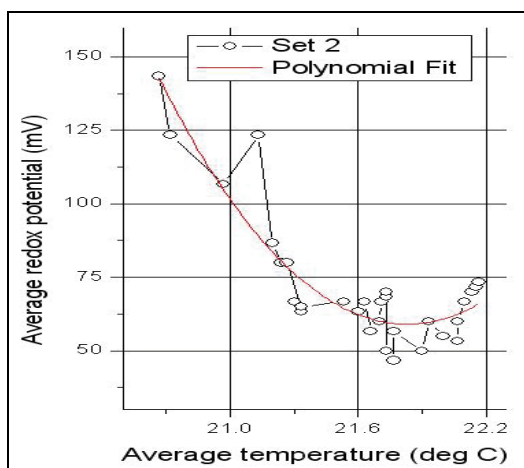
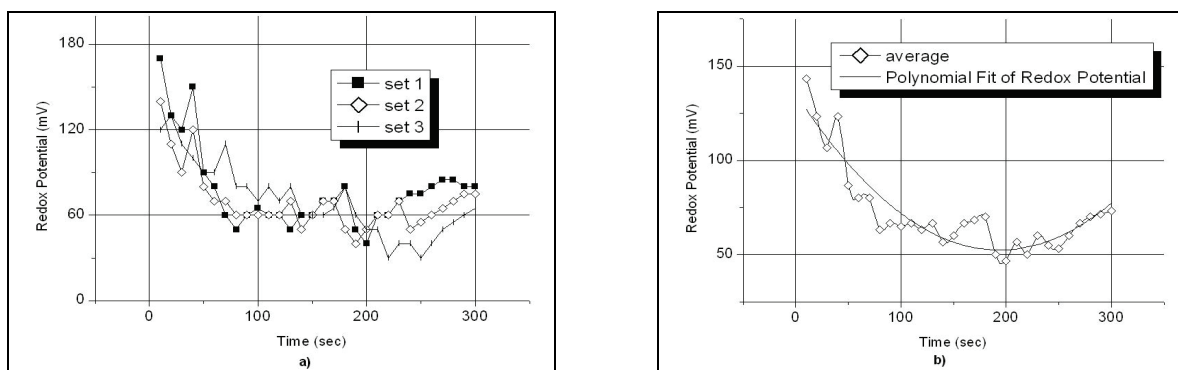


Fig. 8. Evolution of the redox processes in drinking water under the irradiation process: a) for the three samples; b) average; c) average redox potential as a function of temperature for drinking water

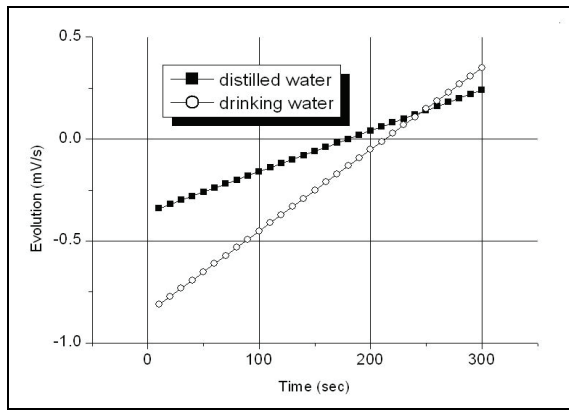


Fig. 9. Variation rates of the redox potentials for distilled and drinking water

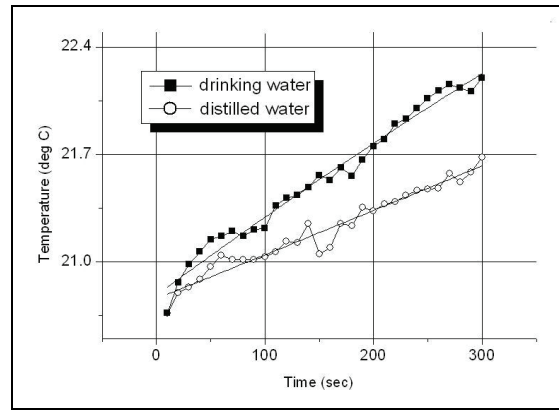


Fig. 10. Comparative evolution of the average temperature for distilled and drinking water

2.3. Experiments and results for samples of slightly acidic aqueous solutions

The slightly acidic aqueous solutions obtained from acetic acid (HCN, 2002) dissolved in drinking water, which are used in the food industry, were tested at non-ionizing electromagnetic field exposure in the same physical conditions as the two previous sets of samples with water. In order to make a comparative study with the samples of distilled water and drinking water, each sample set consisting of three samples with identical low concentration of acidic solution was analyzed with the initial temperatures of 19°C, 21°C, and 22°C. The evolution of the redox process for each sample was recorded in the diagrams below as follows:

1. acetic acid solution, 2.25‰ (Fig. 11);
2. acetic acid solution, 4.5‰ (Fig. 12);
3. acetic acid solution, 9‰ (Fig. 13).

We have noted a specific change in the redox process different from the behavior of the waters (Figs. 11-13). The analysis of the comparative evolution of the redox process for the tested solutions of acetic acid complies with the evolution diagrams from Fig. 14 and the corresponding equations.

The redox potentials for acetic acid with concentrations of 2.25‰, 4.5‰ and 9‰ have the following expressions (Eqs. 10-12):

$$V_{2.25} = -0.00001 \cdot t^3 + 0.004 \cdot t^2 - 0.346 \cdot t + 265.3 \quad (10)$$

$$V_{4.5} = -0.00001 \cdot t^3 + 0.003 \cdot t^2 - 0.202 \cdot t + 345.4 \quad (11)$$

$$V_9 = -0.000003 \cdot t^3 - 0.006 \cdot t^2 + 0.538 \cdot t + 336.8 \quad (12)$$

The variation rates of the redox potentials for aqueous solutions of acetic acid with the above-mentioned concentrations are illustrated in Fig. 15 and can be expressed through derivation as (Eqs. 13-15):

$$V_{2.25} = \frac{dV_{2.25}}{dt} = -0.00003 \cdot t^2 + 0.0086 \cdot t - 0.346 \quad (13)$$

$$V_{4.5} = \frac{dV_{4.5}}{dt} = -0.00003 \cdot t^2 + 0.006 \cdot t - 0.202 \quad (14)$$

$$V_9 = \frac{dV_9}{dt} = -0.000009 \cdot t^2 - 0.012 \cdot t + 0.17 \quad (15)$$

Figs. 16 (a-c) illustrate the variation of redox potential in slightly acidic solutions (concentrations 2.25‰, 4.5‰ and 9‰ respectively). These solutions were exposed to an electromagnetic radiant field for 300 s. There were several observations to note. When exposed to a radiant field for a short period of time, these solutions supported a very slight temperature increase, but were not affected with respect to the redox potential average (it fits to the Nerst equation). However, when the exposure is prolonged and the temperature change is more obvious, the graph is deformed and a trend towards the initial potential value cannot be seen, by contrast with drinking and distilled water, discussed previously. For a lower acid concentration, the temperature at which the graph starts to deform is higher and that implies that the moment when the graph starts to be affected is further in time (Fig. 14).

3. Discussion

The redox potential from the graphs presented in the paper represents the measured redox potential (E_m) resulting from the equation: $E_m = E0/SHE - E_p$, where $E0/SHE$ is the absolute redox potential compared to the standard hydrogen electrode and E_p is the potential of the measuring electrode, and in this case $E_p = 197$ mV.

The aqueous solutions used in measurements (experiment) have the following features: (i) the double distilled water was obtained from drinking water with pH = 7.3 and was neutralized to pH = 7; (ii) the slightly acidic solutions with pH = (6.9, 6.7, 6.5) were obtained by diluting acetic acid in the double distilled water, used for the analysis.

The experiment was carried out based on the redox potential of double distilled water, primarily, in order to maintain the reference stable to temperature variations. Because pH = 7 is thermo-invariant, the effects of the microwave radiation were related to the neutralized double distilled water. Double distilled

water was also used in order to preserve the composition of the dominant substance in the solution regarding the type of the impurities (which, in very small quantities, lead to a fairly large dispersion in the range of the redox potential, making it difficult to interpret the results).

The pH and the value of the redox potential are interdependent. For the substances analyzed in the radiant microwave field, the redox potential variation is substantial, highlighting the radiant effect, whereas the temperature variation is minimal. The sway of the redox potential for solutions with pH = 7 is strong, which has led to an increase in the measurement sensitivity. The use of three identical samples with the initial temperatures of 19°C, 21°C, and 22°C respectively, was necessary in order to trace the evolution trajectory of the irradiation process effect. The lower the acid concentration is, the longer the exposure time is, and this has a small influence on the variation of the potential.

For example, from the redox-time characteristic (Fig. 14), one can observe that the sample with concentration of 2.25 ‰ ($V_{2.25}$) has a duration of insensibility or stability to exposure around 200 seconds. The characteristic $V_{4.5}$ has a time interval of stability to exposure around 150 seconds. For V_9 the redox potential of the solution with a concentration of 9 ‰ is unstable once exposed, as can be seen in Fig. 15.

This trajectory of the redox process in the weak acid aqueous solutions, above-mentioned, is better observed in the diagrams of Fig. 15, in which it is shown that for concentrations of 2.25 ‰ and 4.5 ‰, the variation rate of the redox potential is modified during the sufficiently slow exposure, which in turn highlights the balancing between the effects produced by temperature variation and the effects produced by irradiation. In aqueous solutions which are exposed to high frequency radiation, the redox process has a non-linear evolution over time.

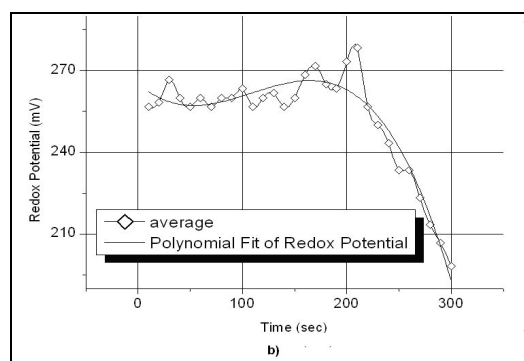
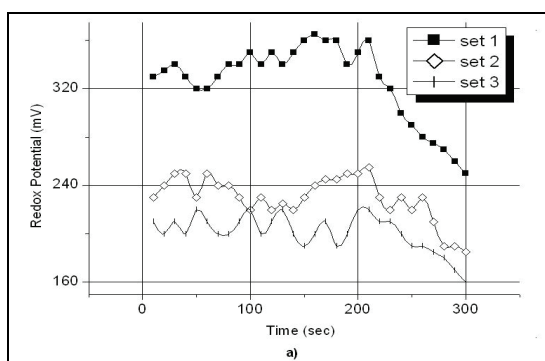


Fig. 11. Evolution of the redox process for aqueous solutions of acetic acid with a concentration of 2.25‰: a) in the samples; b) average

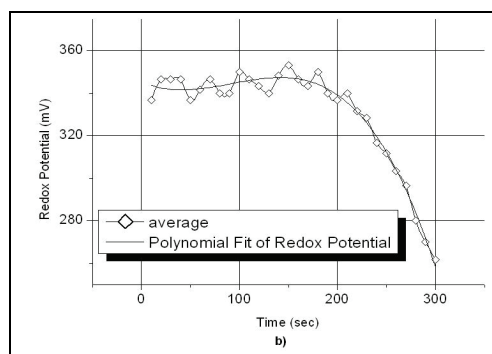
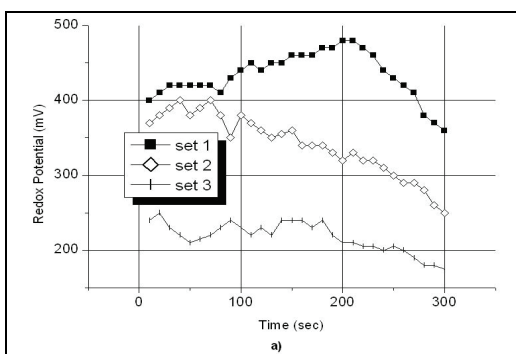


Fig. 12. Evolution of the redox process for aqueous solutions of acetic acid with a concentration of 4.5‰: a) in the samples; b) average

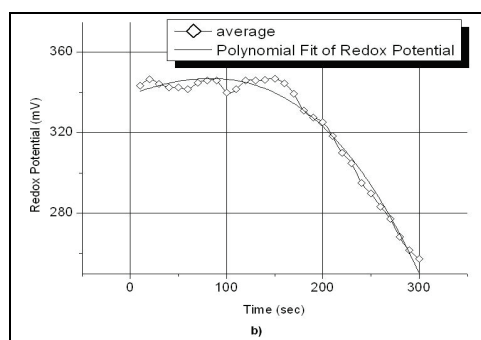
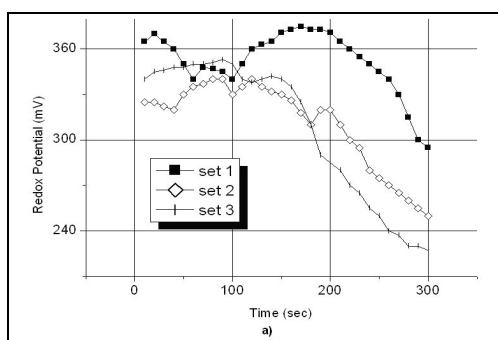


Fig. 13. Evolution of the redox process for aqueous solutions of acetic acid with a concentration of 9‰: a) in the samples; b) average

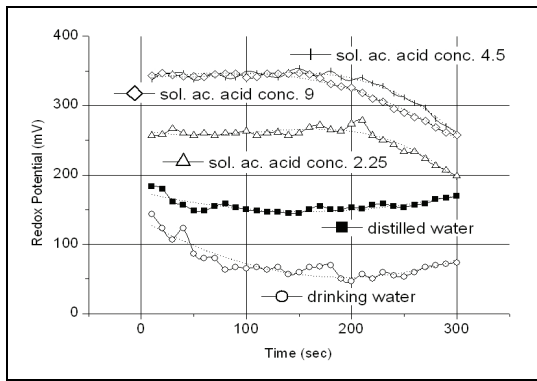


Fig. 14. Average evolutions of the redox processes for the five types of liquids used in the research study

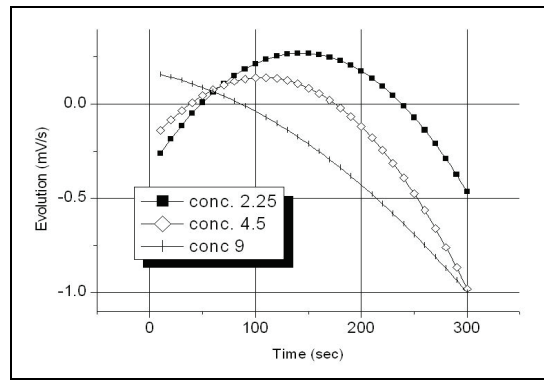


Fig. 15. Variation rates of the redox potentials for aqueous solutions of acetic acid, as presented in Eqs. (13-15)

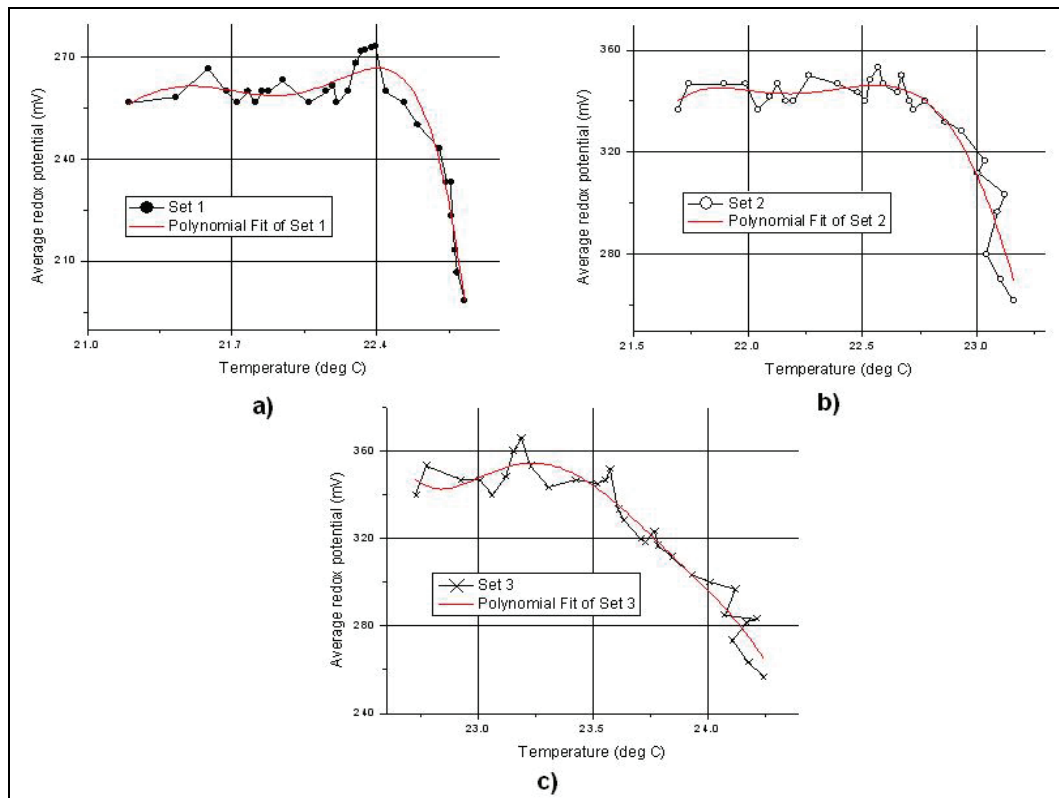


Fig. 16. Redox potential average variation as a function of temperature for slightly acidic solutions

4. Conclusions

Based on the experimental results, the following conclusions can be drawn:

The variation of the redox potential for distilled water (which has a high degree of purification) has not been influenced substantially when exposed to the microwave radiant field, the variations of the redox potential being driven by the temperature changes in the resonance cavity used for measurements.

The slightly alkaline drinking water (pH=7.3), when exposed to microwave radiation, changes its redox potential, due to both the thermal effect and the radiant effect. Also, the rate of variation of the redox potential during exposure was double than for distilled water. Maintaining the microwave radiant

field of low intensity, the redox process tends toward balance remaining constant.

Compared with the water samples at which the redox potential is less affected, the slightly acidic solutions present a fundamental change in the redox potential depending on the duration of exposure, with a strong drop for long exposures, over 200 seconds, and with no tendency to rebalance.

The redox process, for the entire slightly acidic domain, has an graphical evolution with concave profile. The evolution of the graphical profile for the water samples is slightly convex.

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