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ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY CHARACTERIZATION OF KITCHEN UTENSILS USED AS MATERIALS FOR LOCAL COOKING IN TWO CULINARY MEDIA

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Abstract: This study is inscribed in the framework of the valorization of traditional kitchen utensils recycled from aluminum waste in Burkina Faso. In fact, these traditional kitchen utensils made of recycled aluminum alloys occupy a very important place in Burkina Faso's kitchen. The effect of foods for consumption on its local utensils was studied using the non-stationary technique and electrochemical impedance spectroscopy. For this purpose, a sample of utensil has been deducted on traditional production site. The corrosion behavior of the recycled aluminum alloy ok know chemical composition was evaluated by analyzing the impedance spectra obtained at the open circuit potential, in the salt media titrated at 3 g·L^{-1} and rice. Modeling electrical properties by the using of a simple equivalent circuit made it possible to interpret the results obtained by impedance spectroscopy. The results showed a susceptibility to pitting corrosion and were confirmed by the electrochemical impedance spectroscopy method.

Keywords: *corrosion, electrochemical impedance spectroscopy, equivalent circuit, foods, recycled aluminum alloy*

INTRODUCTION

The pots made by craftsmen from recycled aluminum alloys play an important role in the cooking process in Burkina Faso. These alloys stand for very reactive materials and react instantly to media containing oxygen. This is why their outside surface is covered with an isolating oxide film. The thickness of this film reached around 10 nm and plays a protective role on those materials to corrosion which was generally observed in some aggressive media. The state in which these materials are located is called passivity state. The passivity condition can be stopped at any time when defects are found in the oxide film (discontinuity, heterogeneity) or the presence of aggressive ions in the electrolytic media (halogen, cyanide, etc.). This can lead to a release of a localized aggression [1]. Aluminum alloys have a low density (2.7 $g \cdot cm^{-3}$), a good thermal and electrical conductivity, a low melting point, easy to shape, a relatively low price which is advantageous for local people [2]. Moreover, they are of high mechanical characteristics which make them to be utilized as structural materials. In Africa and particularly in Burkina Faso, craft industry turns to profit these properties in the recycling of aluminum alloys for kitchen utensils manufacturing; the raw material used in this field is made of combined or non-aluminum waste, from old car spare parts, beverage cans and tins [3]. Manufacture techniques remain empirical and recycled aluminum alloys are not homogeneous. Corrosion phenomena is favored when utensils are used for food cooking at high temperature or for long cooking time and when acidic or alkaline food are stocked in these same containers for long time [4 – 5]. The humidity, the high temperature and the cooking times are factors which favor the metallic materials corrosion, from which some of the component elements of the corroded material get through the surrounding aqueous media. Despite the numerous studies related to aluminum and their alloys corrosion, few scientific, strict and comprehensive studies on the behavior to corrosion, recycled allows in the craft industry have been conducted. The objective of this work was to study the corrosion behavior of a recycled aluminum alloy collected in the city of Ouagadougou (Burkina Faso) in various culinary media and to evaluate the anti-corrosion effect in these media.

MATERIALS AND METHODS

Sample preparation

The chemical composition of the recycled aluminum alloy is showed in Table 1 [6].

***Table 1**. Chemical composition of the recycled aluminum alloy determined by X-ray fluorescent technique*

Chemical elements	Al	Si	Fe	Cu	Zn	Mg	Cl	Mn	P	Pb	remainder
Wt. %	82.8	12.76	0.76	1.21	1.27	0.48	0.19	0.27	0.22	0.01	0.03

Before each measurement, the aluminum allow surface preparation of these discs for electrochemical tests was the followings: the discs were first ground with 400 through 4000 grit SiC papers and then polished with diamond down to 6 µm and followed by 1 µm alumina- 30 % chrome oxide suspension, and finishing with 5 % oxalic acid solution. Later, each polished sample was rinsed with acetone and put in an ultrasonic cleaner for 10 min. Subsequently, it was rinsed with milliQ water and ethyl alcohol, and finally dried under a hot air flow. The contact area with the culinary media is 3.46 cm^2.

Culinary media preparation

The testing media was local culinary media whose composition is given below. The media used in this study are: salt water titrated at 3 $g \cdot L^{-1}$ and broken rice in tap water (5 g of broken rice in 250 mL water) reserve for local consumption. The selection is made based on the fact that rice is the most consumed cereal in Burkina Faso. In this country, the people consume on the average once daily prepared with vegetables, fish and meats. These media were chosen to simulate a cooking process similar to that of Burkina Faso. All electrochemical measurements were performed in five replicates for each cooking media and show a reproducibility up to around 3 to 9 %. Before each test, we assure that all the electrodes are submerged in the media, at the same depth in the electrochemical cell. As the cooking is most of the time performed at a hot temperature, the media were tested at boiling temperature (100 °C) in order to simulate the real cooking conditions [7 – 8].

Electrochemical measurements

The electrochemical measurements were conducted in the Analytical Chemistry and Interfacial Chemistry (CHANI) of the University of Brussels (ULB). The EIS measurements were determined in a three electrodes electrochemical cell containing the culinary media. There are three electrodes - the reference electrode, the auxiliary electrode and the working electrode. A saturated calomel electrode (SCE) was used as the reference electrode, a platinum metal gate as the auxiliary electrode and a recycled aluminum alloy as working electrode (WE) made in the laboratory. The EIS measurements were performed with employed Princeton Applied Research potentiostat (model PGSTAT 50). A microcomputer was used for data acquisition. The measurements were carried out after 60 minutes of cooking.

RESULTS AND DISCUSSION

Open – circuit potential measurements

Open-circuit potential, Eoc changes were measured against a standard saturated calomel electrode placed in the same compartment. The recycled aluminum alloy was immersed in the culinary media exposing a circular area of about 3.46 cm^2. A copper wire was soldered at the rear of the electrode which was housed in a glass tube to protect it from the test culinary media. Results of the open circuit potential (E_{oc}) are shown in Figure 1. In the curve (Figure 1), a rapid increase of the open circuit potential was observed followed by a decrease of the value in the two culinary media. Open-circuit potential was studied for 15 min of cooking in the various culinary media. From the curve, a rapid increase of the open-circuit potentials followed by a decreasing of the value in the two culinary media (salt water at 3 $g \cdot L^{-1}$ and broken rice) were observed. It can be noticed that these curves varies towards higher values during the first 150 seconds of cooking but after that, an almost decrease of the potential is observed. In this case, we can observe the aluminum passivation tendency which could have many forms: passivation caused by hydroxides which are absorbed at the metal surface, that caused by absorption of the existing components of the two cooking media or their combination.

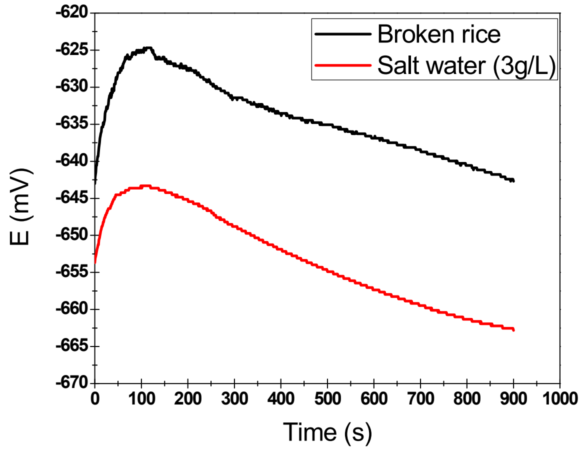

Figure 1. *Open-circuit potential for recycled aluminum alloy in salt water and broken rice*

A comparison of the behavior of recycled alloy in the media (broken rice and salt water) indicated that significantly higher corrosion potential was recorded in the salt water compared to broken rice media. This could be explained by their negative effect susceptible of influencing the passivation during the first minutes of cooking. According to literatures, presence of chloride ions in study media could compete with media hydroxides ions when absorbed at the surface, allowing a localized corrosion and then a deterioration of the passive film [9]. In order to understand more about the existing behavior for metal/media in the cooking media, a series of curves was set out by electrochemical impedance spectroscopy in the context of comparative study in the different media.

Electrochemical impedance spectroscopy (EIS) measurements

Behavior to corrosion from recycled alloy in the two cooking media simulating a similar process to Burkina Faso cooking habit was studied by electrochemical impedance spectroscopy at 100 °C and different cooking times. The frequency ranged from 100 KHz to 100 mHz, and the amplitude was set at 10 mV. Nyquist and Bode plots were used in broken rice media and that of salt water titrated at 3 $g \cdot L^{-1}$ and up to boiling temperature after various cooking times in an open-circuit. Data acquisition and analysis were performed with microcomputer. The spectra were interpreted using the ZSimpWin program. These measures were performed in five replicates to ensure the results reproducibility.

Effect of cooking times

Measuring electrochemical impedance consists in studying the response of the electrochemical system, due to disturbance which is most often a low amplitude double signal. The strength of this technique is to differentiate the reaction phenomena from their relaxation times. Only quick processes are characterized in high frequencies; when the applied frequency decreases, appears the contribution of slower steps as transport phenomena or solution diffusion. To evaluate the behavior of the passive layer in various culinary media, the sample of aluminum alloy was immersed continuously for 60 minutes (00 min, 15 min, 30 min and 60 min) for broken rice and salt water. During these cooking times, only measurements of impedances have been regularly performed since they do not disturb the system. Nyquist graph (Figures 2 and 3) illustrates the experimental impedance diagrams to corrosion potential obtained from the aluminum alloy in the studied culinary media. Indeed, (Figure 3) shows a progressive decrease in the size of the impedance spectrum in a more or less flattened half circle shape, characterizing the formation of the protective layer (alumina Al_2O_3). This leads to a decrease of the total recycled aluminum resistance with regards to the cooking time. In contrast to the salt water media, the broken rice media (Figure 2) shows an increase in the spectra size, confirming the sample resistance of the media [10] .We find a phase difference with respect to axis of real (Figures 2 and 3), which may be explained by the surface none-homogeneity. However, for a better correlation between the experimental data and simulation, we introduced into the procedure for calculation a constant phase element and the surface none-homogeneity is realized through this constant phase element as fallow Equation 1 [11 – 14].

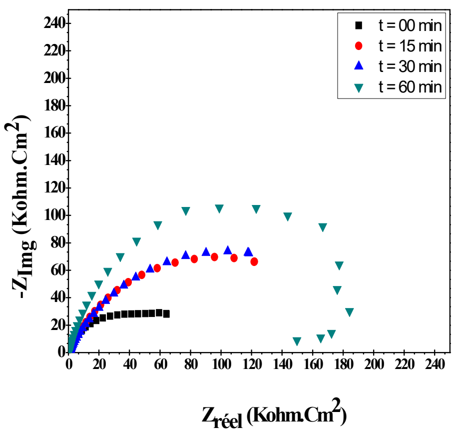

Figure 2. *Nyquist plots for recycled aluminum alloy tested in broken rice media*

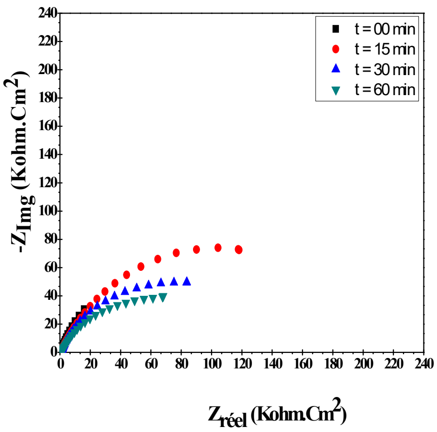

Figure 3. *Nyquist plots for recycled aluminum alloy tested in salt water media*

Despite of a constant phase element being utilized for data fitting instead of an ideal capacitor, since n values obtained from data fitting were in the range from 0.85 to 0.95, the value obtained from data fitting was taken as the capacitance (Equation 1).

$$Z_{CPE} = C\left[(j\omega)\right]^{-n} \qquad (1)$$

Z_{CPE} = the impedance of the CPE;
C = the capacitance associated to an ideal capacitor;
j = the complex imaginary number;
ω = the angular frequency and
$-1 \prec n \prec 1$ [15 – 16]. When n = 1, CPE is an ideal capacitor (Equation 2).

$$Z_{dc} = C[(j\omega)]^{-1} \tag{2}$$

Z_{dc} = double layer capacitance.

A true capacitive behavior is rarely obtained. The n values close to 1 represent the deviation from the ideal capacitive behavior [17].
The best simulation is obtained from the use of equivalent circuit proposed for metal/electrolyte interface and illustrated in Figure 4. This equivalent circuit was proposed by Zhang et al. [9] to describe the bi-layer oxide film formed on aluminum and aluminum corrosion aqueous media.

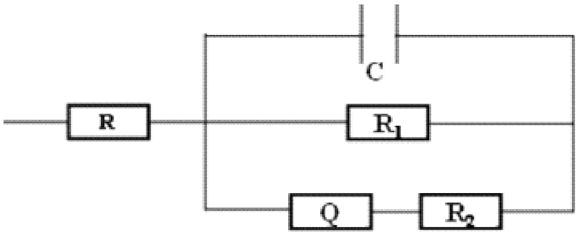

Figure 4. *Equivalent circuit to aluminum alloy in the cooking food of Burkina Faso*

This circuit is valid for all determinations. In the equivalent circuit, R is the salt water and the broken rice resistance, R_1 is the resistance to polarization, C is the corresponding capacity to the dense oxide layer, R_2 is the resistance in porous oxide position and Q is the corresponding component of the constant phase to porous oxide positions. The results of the parameters in the equivalent circuit are shown in Table 2.

Table 2*. Electrical parameters of equivalent circuit obtained by fitting the experimental results of EIS tests*

Media	**Times [min]**	**R KΩ·cm²**	**C F·cm⁻²**	**R₁ KΩ·cm²**	**Q S sⁿ·Cm⁻²**	**n**	**R₂ KΩ·cm²**
Broken rice	**0**	0.697	0.0017	0.3480	0.0048	0.73	75.4
	15	$1.81.10^{-8}$	$1.72.10^{-5}$	0.4830	0.0053	0.78	196.8
	30	$3.21.10^{-8}$	$1.53.10^{-5}$	0.5364	0.0060	0.76	223.8
	60	$1.96.10^{-6}$	$9.14.10^{-6}$	0.3061	0.0015	0.87	259.2
Salt water	**0**	$6.70.10^{-11}$	$1.41.10^{-5}$	0.4474	0.0426	0.80	330.9
	15	$2.30.10^{-7}$	$2.04.10^{-5}$	0.5586	0.0080	0.74	155.9
	30	$2.40.10^{-8}$	$1.53.10^{-5}$	0.5366	0.0060	0.76	222.3
	60	$2.89.10^{-7}$	$1.58.10^{-5}$	0.6573	0.0105	0.66	152.8

For the recycled aluminum alloy, different resistivity profile in both media, regardless of the cooking time is observed as the impedance diagrams vary with the immersion time (Figures 5 and 6). It shows that parameters in the salt water media decrease in contrast to those in broken rice media for different cooking times up to 60 minutes.

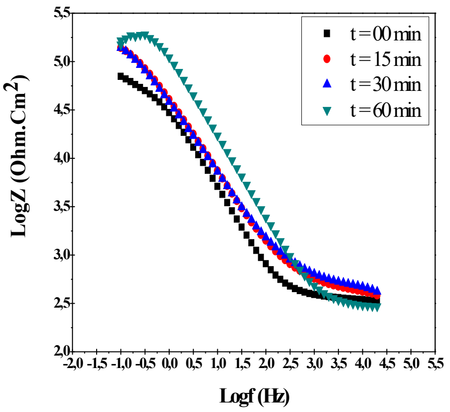

Figure 5. *Bode plots spectra for recycled aluminum alloy tested in broken rice media*

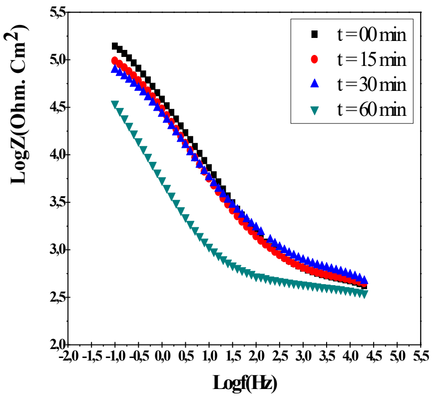

Figure 6. *Bode plots spectra for recycled aluminum alloy tested in salt water media*

This behavior may be associated with physicochemical variations which occurred in the oxide film (alumina) during cooking in the salt water media (penetration of the electrolyte into the oxide layer and hydration of alumina) containing chloride ions. Comparison of the curves (Figures 5 and 6) clearly shows that resistivities in the layer alumina developed on the alloy recycled aluminum are higher in the broken rice media than those in the salt water. This could be explained by the presence of a more homogeneous and dense layer for the recycled aluminum in the media and also that of chloride ions in the salt water. Because, the behavior of interface/medias completely different with the latter. The overall behavior is reflected in the impedance diagram by a decrease in size of the capacitive phenomenon. This can be explained by the weakening and destruction of a film which is likely to be developed on the surface of the studied alloy allowing disappearance of the distribution phenomenon and the decrease of the resistance. These differences may be explained by the oxide layer composition developed on the alloy which is influenced by the chemical composition of material solid media and by the chemical composition of the intermetallic particles [18 – 20].

In conclusion, resistivity profiles obtained for recycled aluminum alloy showed that the oxide layer developed is less protective in the salt water media than the broken rice. This result would be bound to the zinc presence which would return this less resistant system [21 – 23]. The negative effect of chlorides in the salt water media are presented in Table 2. This result was translated by the decrease in the polarization resistance. It appeared also an increase in the capacity associated with the polarization resistance. This increase may reflect the dissolution of the recycled alloy in the salt water media.

The polarization resistance stands for the sum of the dense oxide layer resistance and that of the two cooking media (salt water and broken rice) [24]. In this case, R_2 is much larger than R_1, therefore, it can be considered as the polarization resistance. Table 2 illustrated the simulation parameters. It shows that the polarization resistance increases gradually with the increased cooking time up to 60 minutes for media broken rice while for the salt water media a decrease is observed followed by a slight increase. Highest values of the polarization resistance in broken rice media as compared to the salt water can be explained partly by the chemical composition of the recycled alloy capable of modifying the physical and chemical properties of the oxide layer into more or less noble depending on the studied media, and secondly, by the resistance of the charge transfer (R) which is not identical for both media. Figure 7 indicated a clear difference between the polarization resistance values from the two cooking media.

Observation Figure 7, curves show that the sample from the broken rice media is less corroded than that from the salt water media. This confirmed the destructive effect of the salt water media on our sample [22, 25, 26].

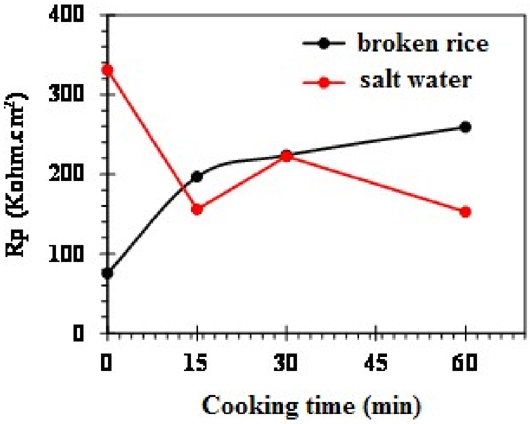

Figure 7. *Polarization resistance according to cooking times*

CONCLUSIONS

This study contributed to the characterization by electrochemical impedance spectroscopy of the local kitchen utensils used for cooking. From this study, we conclude that variations of the impedance spectra in Niquist Z diagram based on the cooking time confirms the development of a protective oxide layer (alumina) of this alloys in electrochemical tests, resulting in an increase of the polarization resistance jointly with a decrease in the capacity of the double layer. Electrochemical tests showed a good efficacy of the sample in the broken rice media and having a good resistance to corrosion comparatively to salt water media. The low resistance to corrosion of sample in the salt water media is certainly caused by chloride ions. Susceptibility to corrosion by pitting has been confirmed by the method of electrochemical impedance spectroscopy.

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