

Effect of Cooking Condiments on the Mass Loss of Artisanal Aluminum Cooking Utensils: The Case of Congo—Brazzaville

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Abstract

Corrosion of cookware is a growing concern for the durability of materials. A rapidly emerging theme that is one of the major current societal challenges at the interface of environmental and health issues. In this present work, the corrosion of aluminum in food environments (salt water and fresh tomatoes) was studied. The three aluminum utensils were purchased in the various workshops in Brazzaville (Republic of the Congo). The weight loss method followed the effect of cooking media on cookware, X-ray diffraction (XRD) and fluorescence (XRF) are two methods used to characterize alloys. XRF analysis indicates that the samples consist of aluminum as a basic element. XRD reveals that the essential building blocks of cookware samples are aluminum, silicon, iron, copper, magnesium and zinc. Finally, gravimetric measurements are carried out to assess the mass losses of samples of artisanal kitchen utensils when cooking food. Aluminum is found to be sensitive in TF and OS media.

Keywords

Kitchen Utensils, Corrosion, Loss of Mass, Intermetallic Compounds, Aluminum, Salt Water, XRD

1. Introduction

The corrosion behavior of aluminum alloys has been widely studied in the industrial world (energy, transport, etc.). However, there is limited work on the

corrosion of aluminum and its alloys in the food industry. The authors are concerned with the degradation of materials itself seen through the concepts of materials science, so that innovation is related by both sides of the problem. The corrosion of aluminum in aqueous media has been widely studied, it is considered responsible for the degradation of materials. For aluminum, corrosion begins with the attack of the protective film (Al_2O_3) which covers these products spontaneously exposed to the open air [1]. Aluminum and its alloys are widely used in industry, transportation, construction and automobiles, as well as in the manufacture of kitchen utensils and food packaging. The great utility of aluminum is mainly due to its remarkable properties (relatively low cost, ease of shaping, good thermal conductivity, low specific and electrical weight, etc.). In developing countries, artisans use the properties of aluminum to make kitchen utensils for cooking and packaging food. Many artisanal kitchen utensils are made from recycled waste, which can originate from car wrecks, carpentry parts, tin cans, electrical cables and more. Aluminum is the main constituent of these types of waste. The use of aluminum utensils is a source of human ingestion of aluminum [2]. Handcrafted aluminum kitchen utensils are ubiquitous in collective kitchens for cooking and food storage, these utensils corrode when in contact with acidic and alkaline media such as tomato sauces and salted water [3]. The degradation caused by aluminum materials results in the passage of part of the constituent elements of the material into the surrounding aqueous medium [4] [5]. Studies have shown that the release of aluminum in saucepans varies depending on the temperature, the cooking time, the aggressiveness of the cooking environment, the quality of the old or new utensil and the topography of the dish. The strong solubilization of aluminum in cooking utensils is a source of heavy metals in the case of acidic food cooked for a long time.

It is in this context that we conducted this study, the objective of which is to understand the effects of two staple food condiments of Congolese cuisine on the degradation of kitchen utensils made in Congo Brazzaville. In addition, our study focuses on foods brought into contact with the alloys from which we seek to follow the evolution of mass loss. Specifically, we intend to characterize the alloys by carrying out mineralogy and chemical analysis, to follow the effect of degradation by loss of mass as a function of the firing time and finally to study the behavior of the alloys in the different media in order to understand the phenomenon of corrosion.

2. Methodology

2.1. Experiments

Three (03) samples of wafer-shaped aluminum alloys were taken from three Congolese cooking pots (casseroles) made in three different workshops: E1 alloy, E2 alloy and E3 alloy.

2.2. Study Media

To show an operation similar to Congolese cuisine, we placed the samples in a

Pyrex beaker containing the different media intended to understand the corrosion phenomena: the salted tap water medium (OS titrated at 3 g/l) at a temperature of boiling and the fresh tomato medium (TF titrated at 33.33 g/100ml) mashed in 100 ml of tap water at boiling temperature. Since most of the cooking is done hot, the media (OS and TF) are tested at boiling temperature (100°C) to simulate real cooking conditions, temperature recommended by directive 97/48/EC [6].

2.3. Product Characterization

Characterization of materials: X-ray fluorescence allowed a qualitative and semi-quantitative analysis to be carried out. The device used is an epsilon 3 (PANATICAL) type. X-ray diffraction is mainly used to identify and demonstrate the nature of the precipitated phases of the intermetallic compounds present. The diffractometer used is PANATICAL brand XPERT-3. The angular interval that it measures ranges from 10° to 90° on the 2θ scale. The sweep of this interval is not done step by step, the step length being 0.02° always on the 2θ scale. The X-ray source used has a Cu target, the radiation emitted is the $K\alpha_1$ - $K\alpha_2$ doublet of copper. The phase indexing is done by comparison of the experimental X-ray diffraction spectra and the theoretical spectra (pow_COD files) of the Qualx software [7].

Gravimetric technique: Mass loss measurements give a direct estimate of the corrosion rate of the three aluminum alloys in various culinary environments. The corrosion rate is determined after 60 minutes of cooking at boiling temperatures (100°C). It is calculated according to the following Equation (1):

$$V_{\text{corr}} = \Delta m / S \cdot t \left(\mu\text{g} \cdot \text{cm}^{-2} \cdot \text{j}^{-1} \right) \quad (1)$$

Δm represents the difference between the initial mass m_1 and the final mass m_2 after the different cooking times. S is the area of the metal exposed to the study solution and t the cooking time.

For the mass loss tests, the equipment necessary to carry out these mass loss measurements comprises an analytical balance precise to five decimal places, a multi-parameter indicating the pH and the temperature of the solution tested over a time range going from 15 min, 30 min and 60 min. The mass loss tests are carried out by placing the parallelepipedal plate of approximately 9 cm² (two faces + the sides) and 3 g of chips are immersed in a Pyrex beaker containing one of the media identified above, when this one reaches the expected temperature. The chips are taken by turning from our raw samples. We used them during tests aimed at quantifying the transfer of material, to increase the metal/medium contact surface [8]. At the end of each cooking cycle (15 min + 15 min + 30 min), during which additions of deionized water have compensated for the losses due to evaporation, the parallelepiped sample is removed and rinsed with water deionized then with acetone. The cleaned sample is then placed in an oven at 110°C, for at least half an hour, to remove all traces of humidity before being weighed with a SARTORIUS CPA225D precision balance, the result is

brought to the initial surface. After analysis, the tested sample is again placed in a culinary environment to undergo a new thermal cycle before being further analyzed. During the tests, the pH of each study medium was measured before and after the corrosion tests. We will also note that to compensate for the evaporation during the cooking phase, deionized water was added periodically to keep the volume of the mixture constant and thus avoid an overconcentration of elements dissolved in the water [8]. To ensure consistency, all experiments are performed in duplicate.

3. Results and Discussion

3.1. X-Ray Fluorescence Analysis

Table 1 groups together the results of the X-ray fluorescence spectrometry (XRF) analysis of the 3 selected samples.

These results show a diversity of chemical composition of the alloys studied, which can be explained by the choices made by the foundry craftsmen when constituting the melting charges. Silicon is the major addition element in our alloy samples ranging from 4.13% to 10.9%. The presence of silicon in a material lowers the melting temperature of an alloy and improves the castability and the wear resistance [9]. The origin of this element is certainly due to the use in large proportions of alloys from the automobile. The copper and zinc contents of the three samples exceed the values of the French standard NF EN 601 of July 2004 [10]. The high level of copper in an alloy reduces corrosion resistance. The iron contents of these alloys are high, although the contents of samples E1 and E2 are lower than the French standard. Only sample E3 has an iron content above the norm. Copper, iron and zinc sensitize alloys to intergranular corrosion. The components (Fe, Mn) are only found in so-called “hard” alloys which make up a maximum of 1/3 of the melt charge [9]. Manganese would form phases which would not influence the corrosion resistance of aluminum alloys. But iron, with aluminum, would form a precipitate which constitutes preferential sites for localized corrosion [11]. The elements magnesium, chromium, titanium, nickel and vanadium are present in small quantities in our alloys and have levels below the standard. Magnesium increases corrosion resistance, but decreases foundry and machinability properties.

3.2. X-Ray Diffraction (XRD) Analysis

Figure 1 shows the diffractograms (XRD) of the three samples.

Table 1. Chemical composition (XRF) in % by mass of the samples taken in the three workshops in the city of Brazzaville.

Ech	Al %	Si %	Mg %	Mn %	Fe %	Cr %	Cu %	Ti %	Zn %	Ni %	K %	Ca %	S %	v %
E1	81.782	10.934	0.599	0.557	1.588	0.04	3.099	0.069	0.954	0.102	0.119	0.077	0.08	-
E2	91.746	4.13	0.837	0.266	0.991	-	1.285	0.044	0.544	0.055	0.025	0.023	0.054	-
E3	86.342	6.037	0.808	0.377	3.098	0.044	2.48	0.045	0.562	0.103	0.047	0.025	0.02	0.012
NF	Base	13.5	11	4	2	0.35	0.6	0.3	0.25	3	-	-	-	-

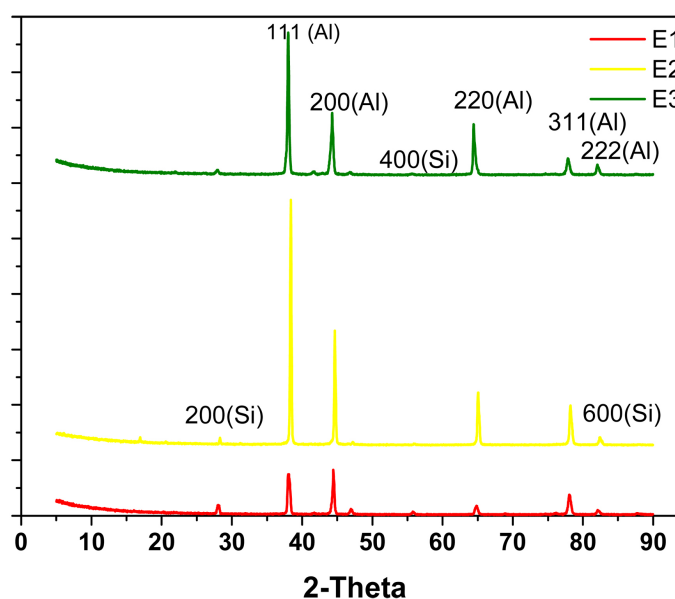


Figure 1. X-ray diffraction spectrum of the samples E1, E2 and E3.

The identification of the lines which appear in these alloys in the raw state gives the peaks corresponding to those characteristics of the α -aluminum matrix according to the ASTM (JCPDS-ICDD 1997) files [12]. Indeed, it should be noted that in the case of the diffractogram of sample E1 in **Figure 1**, the most intense reflection is that oriented along the plane (200), the diffraction angle of which is located at 44.40° and not that due to the plan (111) as prescribed by the ASTM files. This assumes the existence of a privileged orientation (texture) with the plane (100). The absence of other lines of the species is probably due to the low concentration of the transformed fraction of the precipitated phases and the variation in the intensity of the peaks is related to anisotropy. On the other hand, one can clearly notice on the diffractograms of samples E2 and E3 (**Figure 1**) a preferential orientation along the plane (111) corresponding to the diffraction angle of 38.3° . This observation was observed by Sawadogo *et al.* [13]. We notice the presence of small silicon peaks on the diffraction spectrum of samples E2 and E3 (**Figure 1**). On the other hand, a weak silicon peak (200) is observed at the diffraction angle 27.99° in the diffraction spectrum of sample E1. The presence of the small silicon peaks is due to the low silicon content as reported by chemical analysis which showed low levels in samples E2 and E3, in the spectrum (**Figure 1**). The presence of iron is also observed at the same angular values as the aluminum peaks. However, these spectra also manifest less intense unidentified peaks which can result in the presence of traces of foreign phases in the aluminum matrix of his three samples which would be made up of the intermetallic phases of iron (Al_2FeCu_2 , $\text{Al}_{77.69}\text{Fe}_{25.46}\text{Si}_{18.86}$, $\text{Al}_{78.2}\text{Si}_{19.7}\text{Cu}_{32.5}\text{Fe}_{16}$) as well as probable phases of Mg_2Si , CuZn_5 , Mn_3Si and MnO_2 . The formation of these phases, with regard to the low contents of the elements seen in chemical analysis can also be linked to the slow cooling after forming which favors the heterogeneous precipitation of the intermetallic particles at the level of the grain joints

which can constitute preferential sites of corrosion.

3.3. Corrosion Process of Aluminum Alloy Samples

In the case of aluminum, the layer of alumina is continuous and begins to gradually thicken when submerged in boiling water. In the presence of chloride ions contained in the drinking water with a value less than 200 $\mu\text{g/L}$ at the low of food environments [14]. This layer disappears locally. The chloride ions present disrupt the passivity of the Al and this could be due to the ability of the chloride ions to penetrate the oxide film through the pores of defects on the surface of the metal in completion with dissolved oxygen [15]. As described by Vagel in his work on the behavior of aluminum and its alloys, aqueous pitting corrosion occurs [16]. This mechanism is governed by the concentration of hydronium H^+ ions in the solution, it means the pH and the electrical potential of the part in relation to the solution [9]. The average mass loss results for the three cookware samples are shown in **Table 2**.

The aluminum alloys submitted to our study are formed from a mixture of phases present in the matrix of the alloy or at the grain joints according to the results of XRD (**Figure 1**), consisting mainly of aluminum as the primary phase, silicon and intermetallic compounds as the secondary phases. The different phases have different corrosion potentials, a galvanic coupling is established between the different constituents of the alloy, thus the compounds forming the anodic zones go into solution [17] [18]. In addition, material degradation has been reported to be manifested by the appearance and development of pitting. Intermetallic compounds and silicon at the grain boundaries of aluminum (intergranular corrosion) can be activated by the presence of chloride ions. Considering the extreme diversity of the chemical composition of the alloys, the origin of the materials, the method of preparation and cooking of food, etc., the study of the corrosion of metallic materials is therefore complicated. It is therefore very difficult to control all of these factors to have a stable and reproducible environment in the tests, and to compare the results of the tests with those obtained by other authors. The corrosion of metallic materials by food environments becomes difficult because the chemical species they contain vary depending

Table 2. Variation of the $\mu\text{g/cm}^2$ mass loss of the three cooking utensils as a function of cooking time.

Backgrounds	Mass loss $\mu\text{g/cm}^2$					
	OS		TF			
t (min)	0 - 15	0 - 30	0 - 60	0 - 15	0 - 30	0 - 60
Ech						
E1	1.7483719	2.8997388	4.9892565	3.0154719	6.332491	7.4956016
E2	2.0380712	2.8872675	5.8594546	3.3525927	5.2437988	5.5876545
E3	1.4567339	1.7566497	5.5698649	2.7344818	3.3655161	3.7020677

on the origin of the product, the method of preparation and cooking of the food, etc. Knowledge of corrosion mechanisms becomes essential to understand the role of alloying elements in the transfer process.

Aluminum corrodes preferentially at the level of silicon and intermetallic precipitates, for this, the mechanism of corrosion by galvanic coupling in aqueous media of the different phases present in industrial aluminum alloys [9]. However, the Mg-rich precipitates which are anodic to aluminum are dissolved. Since magnesium is associated with zinc in compounds of the $MgZn_2$ type, these elements will therefore go into solution in the food medium [19]. In our diffractograms (**Figure 1**) we have identified the compounds capable of forming a galvanic coupling with aluminum. The aluminum matrix attacked by the food environment will release Al^{3+} ions, which will pass into the food environment. The attack of the aluminum-silicon or aluminum-intermetallic compound interface will lead to a detachment of the latter, which will be transferred to foods in their solid form depending on the medium. Therefore, where the intermetallic compounds remain insoluble, the mode of degradation of the materials is influenced by the amount of copper in the alloys produced by the smelters. The results of the X-ray fluorescence (**Table 1**) showed us the high contents of certain elements, magnesium, but especially zinc, the concentration of which is higher than that admitted by the standard, in the alloys in the form of solid solution, of intermetallic compounds, will go into solution in the food medium.

Examination of these results shows that both environments are aggressive on all three types of cookware. The mass loss increases steadily as a function of time for both media. The TF midfielder seems to be more aggressive than the OS midfielder. The loss of mass in both media is significant.

3.4. Loss of Mass of Samples

Figure 2 shows the results of the different mass loss measurements of the samples.

These results show that the OS and TF media are aggressive. The variations in the loss of mass of the samples in the two cooking media show the same general appearance whatever the concentration of the medium used.

These results reveal a first stage (0 to 15 min) during which the loss is rapid, followed by a second stage where the evolution of the change in mass is much smaller. This high rate of mass loss at the start of the test is mainly due to the detachment of certain solid particles coming from the foundry operation and not removed by the cleaning operations [13]. This high degradation of the samples can also be explained by the phenomenon of corrosions by intergranular pitting, these corrosions are favored by the presence of chloride ions in the solution. In addition, some authors report that the level of silicon and intermetallic compounds found between aluminum grain boundaries are activated by the presence of chloride ions found in OS media [20] [21]. This loss of mass appears to be related to the amount of silicon in intermetallic or needle form present in

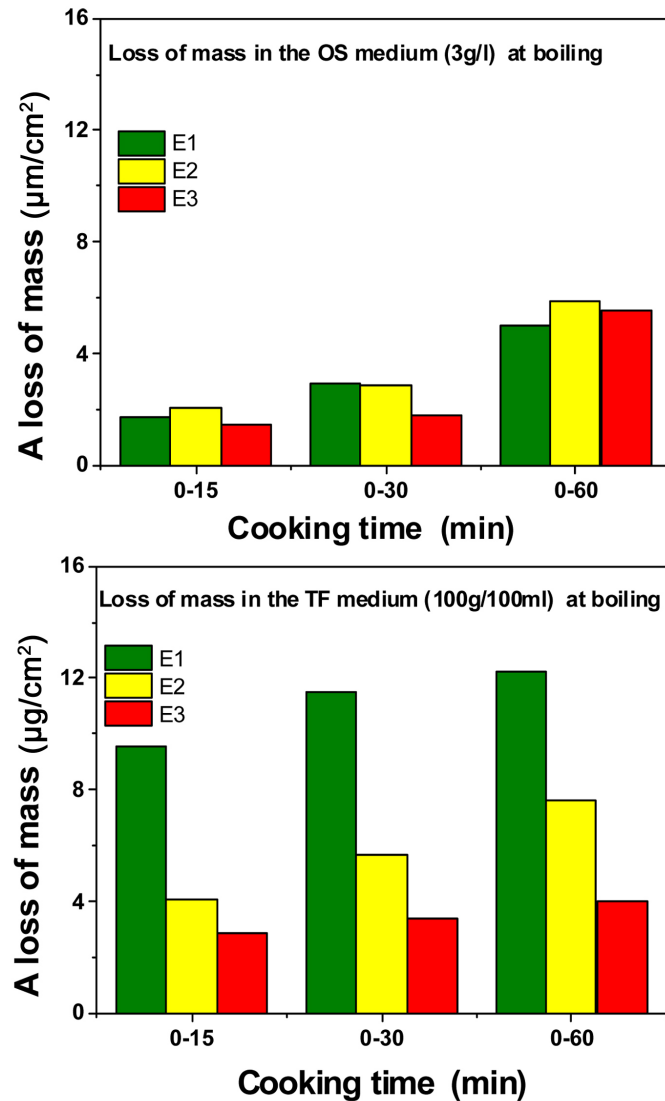


Figure 2. Evolution of mass losses as a function of cooking time in the different media chosen.

samples of local utensils [22].

The trend observed in the second stage in both study settings can be attributed to the combination of high conductivity and high solubility of oxygen [3]. Compared to E2 and E3, sample E1 has the best corrosion resistance, sample E2 has the greatest loss of mass in the OS cooking environment. Against all expectations in the TF culinary world, sample E3 exhibits the best corrosion resistance and sample E1 has the greatest loss in mass. At the end of this analysis, it can be seen that the corrosion resistance of the alloys obtained from recycling in Congo-Brazzaville depends on the one hand on the nature of the food environment and on the chemical composition of the alloys and on the other hand, this may be related to the surface morphology of the materials. On the other hand, it is the aqueous media which are the basis of the various major surface degradations. Aqueous corrosion seems to be at the origin of the loss of mass of the alloys be-

cause the dissolution of these alloys is therefore possible in contact with the culinary medium. Observation of this figure also shows a cumulative loss of mass at 60 min much more accentuated on the sample E2 (OS medium) and E1 (TF medium) and once again, better resistance to corrosion in culinary environments on E1 and E3. This confirms the role of certain firing media on the behavior of these samples and the role played by the nature of the intermetallic particles as a function of the firing time.

A significant increase in mass loss is also observed compared to the concentrations in the two media. Hydroxide ions (OH^-) are proportional to the loss of mass of aluminum [21]. As the TF medium consists of several organic and amino acids, the complexations of aluminum by these acids also justify the high mass loss because aluminum can form numerous complexes at acidic pH with monocarboxylic acids, dicarboxylic acids and amino acids [23].

3.5. Influence of the Chemical Composition of Aluminum Cookware

Figure 3 shows us the comparison of the cumulative mass losses after 60 min in the OS and TF media.

The influence of the chemical composition appears on the mass losses during the 60-minute curing tests, but also on the corrosion rates in the chosen media. Sample E1 has the best corrosion resistance overall in the OS medium, on the other hand in the TF medium it is sample E3 which seems to have better corrosion resistance. By taking into account the corrosion mechanism and the effect of the chemical composition of the samples, the role of additional elements and intermetallic particles can be shown: The level of silicon present in the alloys determines the number of corrosion sites of aluminum, the greater the silicon, the greater the loss of mass. This constant is demonstrated by comparing the results

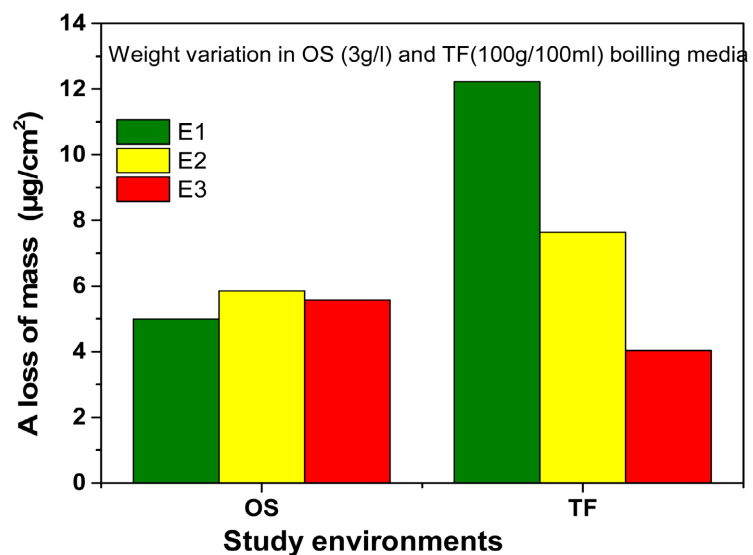


Figure 3. Comparison of cumulative mass losses after 60 min in OS and TF media.

of samples E1, E2 in the TF medium. Copper and iron play a secondary role here, masked by silicon.

On the other hand in the OS medium when the silicon content is low (case of sample E2 and E3), the role of iron becomes predominant and the numerous particles of iron-based intermetallic compounds constitute as many sites of corrosion in the material. In our alloys the various intermetallic compounds influence the loss of mass. Comparatively in the study environments after 60 minutes of cooking, the study shows that the mass loss of sample E1 is 27.3% (**Table 2**) less than that of sample E2 and E3 (**Figure 3**) in the middle OS. On the other hand, in the TF medium, the mass loss of sample E3 is 31.8% (**Table 2**) less than that of E1 and E2 (**Figure 3**). The amount of material lost by the pot has a great importance on longevity. Indeed, this loss results in a decrease in the thickness of the pot, which causes fragility and therefore deep corrosion.

The corrosion rates of the samples tested in the different culinary environments show fluctuations for the different environments considered. These results corroborate those of Ndiaye [3]. The results show that the mass loss of sample E3 in the TF medium is less than that of samples E1 and E2. The pH of the three samples is very varied, this variation in pH seems to be the cause of the mass losses observed in the samples. This is in accordance with the work of Shuping who showed that hydroxide ions (OH^-) significantly influence the loss of mass of aluminum [9] [24].

Table 3 gives us the results of the corrosion rate and the pH of the cookware samples at the different boiling exposure times.

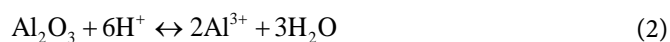
Table 2. Weight loss (mi/ Δ m) in cooking media at 60 min.

Backgrounds	Ratio (mi/ Δ m) 10^{-3} (%)	
	OS	TF
Ech		
E1	27.3	69.9
E2	39.1	61.8
E3	48.9	37.8

Table 3. Corrosion rate and pH of samples in boiling OS and TF media.

Media/pH	Corrosion rate $\mu\text{g}/(\text{j}\cdot\text{cm}^2)$ in boiling OS and TF media							
	OS			pH before/pH after	TF			pH before/pH after
	t (min)	0 - 15	15 - 30		30 - 60	0 - 15	15 - 30	
Ech								
E1	167.84	278.37	239.48	7.56/9.02	915.05	1103.80	586.78	4.35/4.45
E2	195.65	277.18	281.25	7.48/9.12	391.66	541.66	366.66	4.33/4.4
E3	139.84	168.64	267.35	7.81/9.12	275.99	329.54	193.61	4.31/4.43

These results show that the weight loss of kitchen utensils in culinary environments is a function of the chemical composition of the alloys, since samples E1 and E2 which have practically the same chemical composition lose more weight in the TF compared to the OS medium. On the other hand E3 loses more in the middle OS. Indeed, aluminum has a passive behavior in aqueous solution, it forms a compact protective film Al_2O_3 on its surface. However, the solubility of this protective film increases in acidic and alkaline medium. According to Sawadogo the release of aluminum in aqueous solutions can be explained by the chemical reaction that occurs on the surface of samples of aluminum cookware according to Equation (2) [21]:



where Al_2O_3 is the protective film (alumina) on the entire surface (anode and cathode). Aluminum ions released in solution react with organic acids found in foods such as oxalic acid, citric acid and other complexing agents such as hydroxyls.

3.6. Effect of Environment

The effect of pH on the aluminum lexivation of cookware in saline and tomato sauce is shown in **Table 2**. The losses of mass measured show a significant aggressiveness of the culinary environments. Cooking salt has a strong influence on tap water, tomato puree shows a strong aggressiveness compared to salt water. Because this fruit has a strong acidity (pH = 4.7). The place of cultivation and the period can be decisive for the acidity of the fruit. Chloride ions contained in the mains water used in the TF medium also contribute increase in this aggressiveness. **Table 3** shows the high corrosion rates at low pH, it decreases to a minimum pH of 4.31. As a result of high values pH = 7.81, the corrosion rate appears to be high. The authors report that aluminum is very sensitive to high pH and exhibits increased corrosion in alkaline environments. This result is in agreement with the work of Shuping Bi who states that leaching increased considerably in the ranges of pH < 4 or pH > 8 [22]. Pitting corrosion is a mechanism that can be activated, among others, by pH as has been presented by several authors [23]. From its results kitchen utensils are susceptible to being weakened and even destroyed over time by acidic and salty foods. Rim Karbouj also makes the same conclusion [25].

4. Conclusion

This work aimed to understand the effect of food condiments on artisanal kitchen utensils, from three samples collected in different workshops in Congo (Brazzaville). The chemical characterization by X fluorescence shows a diversity of chemical composition of materials, X-ray diffraction revealed the presence of secondary phases consisting of silicon and intermetallic particles which are preferential sites of corrosion. Aluminum and its alloys are widely used in industry, transportation, construction and automobiles, as well as in the manufacture of

kitchen utensils and food packaging. The great utility of aluminum is mainly due to its remarkable properties (relatively low cost, ease of shaping, good thermal conductivity, low specific and electrical weight, etc.). In developing countries, artisans use the properties of aluminum to make kitchen utensils for cooking and packaging food. Sample E1 showed good efficacy in OS medium compared to sample E2 and E3. In TF medium, sample E3 was found to be more resistant compared to E2 and E1. The measurement of the mass losses observed in culinary environments, reveals that the corrosion resistance of the material appears in the coherent order with respect to the silicon content in the samples (E1 and E2) and with respect to the culinary environment except in the middle TF. The mass losses observed in our study environments are mainly due to the presence of chloride ions (Cl^-) which promote the degradation of the aluminum surface by destruction of the protective layer. Alloy samples react differently in TF and OS media. The pH of the study media is between 7.56 and 7.81 for the OS medium and for the TF medium between 4.31 and 4.35. The effect of media on cookware alloys is due to the acidity of the corrosive medium. From these results, kitchen utensils are susceptible to being weakened and even destroyed over time by sour and salty foods. Preliminary tests do not allow us to conclude on the nutritional value of local kitchen utensils made in Congo Brazzaville. This study made it possible to update the data in the literature and should underline the agro-food and socio-economic interest of the manufacture of kitchen utensils by artisans. These tests must be supplemented by electrochemical measurements in order to evaluate the various corrosion parameters as well as the evolution mechanism for the various media tested on the one hand. On the other hand, carry out chemical tests on the corrosion substrates in order to be able to conclude on the food quality of its kitchen utensils.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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