# Aluminium leaching during tribocorrosion in acidic, alkaline and neutral solutions: cooking simulation

Giovanni U. Bruno[a], Álvaro Meneguzzi[b], Jane Zoppas Ferreira[c]

## Abstract

Some studies suspect that aluminium, among other elements, could cause dementia or some cognitive impairment in human beings as consequence from long exposures to the environment. This survey intended to measure how much aluminium is lixiviated during the cooking process with aluminium pots. The study considered pH variation, working in acidic, alkaline and neutral solutions. A tribometer equipment simulated the wear made by a kitchen spoon, often used during food preparation, and these simulations were inside compartments containing these solutions. After the simulation, the solutions were collected and analyzed by atomic absorption spectroscopy to verify the present aluminium quantity. It's possible to state from this study that the use of aluminium pots should be restricted. If used deliberately, it liberates high amounts of aluminium into the food, above the provisional tolerable weekly intake (WHO). This study didn't consider high temperatures as parameters. It's known that temperature raise makes the environment more aggressive, having more dissolution power and solubilization capacity.

keywords: aluminium, aluminum, tribometer, tribocorrosion.

## Introduction

Some studies suspect that aluminium, among other elements, could cause dementia or some cognitive impairment in human beings as consequence from long exposures to the environment (1) (2). As aluminium is the most abundant metallic element in the earth's crust (around 8%), it's common to find it in many different ways in nature: Silicates, oxides and hydroxides, combined with other elements, and complexes with organic matter. It's correct, therefore, to state that we are well adapted to live in an environment rich in aluminium. The aluminium entry into the human body can occur in many different ways, between food, medications and even drinking water (3). According to the World Health Organization (WHO) (4), the provisional tolerable weekly intake (PTWI) for aluminium from all sources, established by the Joint FAO/WHO Expert Committee on Food Additives (JECFA), is 1 mg/kg of body weight, including additives.

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[a] Engenheiro de Materiais – Laboratório de Corrosão, Proteção e Reciclagem de Materiais/UFRGS
[b] PHD; Professor – Laboratório de Corrosão, Proteção e Reciclagem de Materiais/UFRGS
[c] PHD; Professora – Laboratório de Corrosão, Proteção e Reciclagem de Materiais/UFRGS

The use of pots and equipments made by aluminium to cook and store food has been questioned for being a possible source of aluminium absorption by the organism. There are many ways to lixiviate aluminium and/or its derivates during the use of a pot made of this material, for example: wear by abrasion, dissolution, ions and complex formation and even acidic or alkaline corrosion.  In a simplified way, cooking with an aluminium pot involves variables that may render the process more or less aggressive to the metal. The most consumed food in Brazil has a variable pH (the table 1 below shows some examples), which leads to aluminium's amphoteric behavior. The metal reacts differently face to pH variation. In addition to alkalinity, there are many chemical compounds naturally inside food, others can be absorbed during plantation and/or processing, and still those compounds added during cook process, all of this influences the corrosion process in the metal. Finally, there is the wear made by some kitchen tool (spoon, fork, etc) used during the cooking process and by the very food in movement inside the pot. Considering the importance of the suspicion that the lixiviated aluminium quantity during food preparation is relevant, in comparison to the recommended values, this research analyses aluminium behavior face to different working conditions.

**Table 1 - Examples of foods and their pH values (5) (6)**

| Food | pH |
| --- | --- |
| Rice | 6,26 |
| Steak | 5,77 |
| Tomato Sauce | 4,36 |
| Eggs | 7,96 |
| Corn | 7,8 |
| Conch | 8,4 |
| Grapes, Concord | 2,8 |

**Methodology**

The samples were made from two pizza baking trays of 35 cm diameter, produced by Alumínio Royal S/A, laminated and conformed in Porto Alegre (Brazil).

The baking trays were first cut in rectangles (2 cm x 4 cm) to obtain samples for polarization curves analyses and posteriorly in small squares (2 cm x 2 cm) to the others tests. Finally, x-ray fluorescence analysis was performed with an Fluox portable *Thermo Scientific Niton xl 3t* to identify the present aluminium alloy.

**The Solutions**

Three solutions were made to the analysis as shown at table 2. The objective is to simulate three situations that make the material behave differently. An acidic, an alkaline and a neutral solution with chlorides.

**Table 2 - The solutions used to simulate different food environments**

| Solution | Concentration (mol/L) | pH |
|----------|----------------------|-----|
| NaCl | 0,1 | 7 |
| $CH_3COOH$ | 0,1 | 2,6 |
| NaOH | 0,1 | 12,5 |

**Polarization Curves**

The curves were performed in the three different solutions, using an Autolab equipment, suiting up parameters for each case. To the acidic and the neutral solutions the curves ranged from -1,6 V to 1,0 V and to the alkaline solution ranged from -1,9 to -1,0 V. The analysis were performed with a scan rate of 0,02 V/s.

For each experiment a rectangular sample was degreased uniformly with detergent and a soft sponge for five minutes. An electrochemical cell was prepared for each sample, remaining in contact with the solution during ten minutes before the analysis. The cell kept exposed to the solution a round surface of the sample of 0,62 cm².

**Tribology**

The abrasion tests were performed following ASTM G 133 standards, with a tribometer *CETR – Test Equipment Tribology*, with *ball on plate* method. The wear procedure were performed by an alumina sphere (4,7 mm of diameter), with a constant force of 1 N, in a frequency of 2 Hz, covering a 2 mm track repeatedly during 20 minutes. The open circuit potentials (OCP) were also measured 20 minutes before, during and 20 minutes after the abrasion analysis.

The samples were immersed and rubbed inside 30 ml of each solution previously announced, varying the coefficient of friction in each essay. At the end, the solutions were collected to further tests.

**Atomic Absorption Spectroscopy (AAS)**

The solutions collected in the end of tribology tests were submitted to atomic absorption spectroscopy analysis at the Ecology Center of Biosciences Institute at the Universidade Federal do Rio Grande do Sul (UFRGS).

The tests were performed with AAS/nitrous oxide-acetylene flame methodology and the collected sample from the alkaline solution was neutralized with nitric acid before the spectroscopy essay. The samples were submitted to aluminium detection and the used method had a detection limit of 0,077 mg/L.

**Results and Discussions**

The samples were first washed and taken to x-ray spectroscopy analysis to know what elements were present in that material. The equipment approached the alloy composition of all samples to the 5005. Vargel (8) states that the 5000 series has a strong resistance to corrosion. This is one of the reasons because this alloy could be used in kitchen tools, for example. This alloy has the following composition:

**Table 3 - 5005 alloy composition in % (7)**

| Alloy | Si | Fe | Cu | Mn | Mg | Cr | Ni | Zn | Ga | V | Ti | Al |
|-------|------|-----|------|------|---------|------|-----|------|-----|-----|-----|-------------|
| 5005 | 0,30 | 0,7 | 0,20 | 0,20 | 0,50-1,1 | 0,10 | --- | 0,25 | --- | --- | --- | Reminiscent |

**Polarization Curves**

To know better about the behavior of this aluminium alloy in the different chosen environments, three polarization curves were performed and are displayed below:
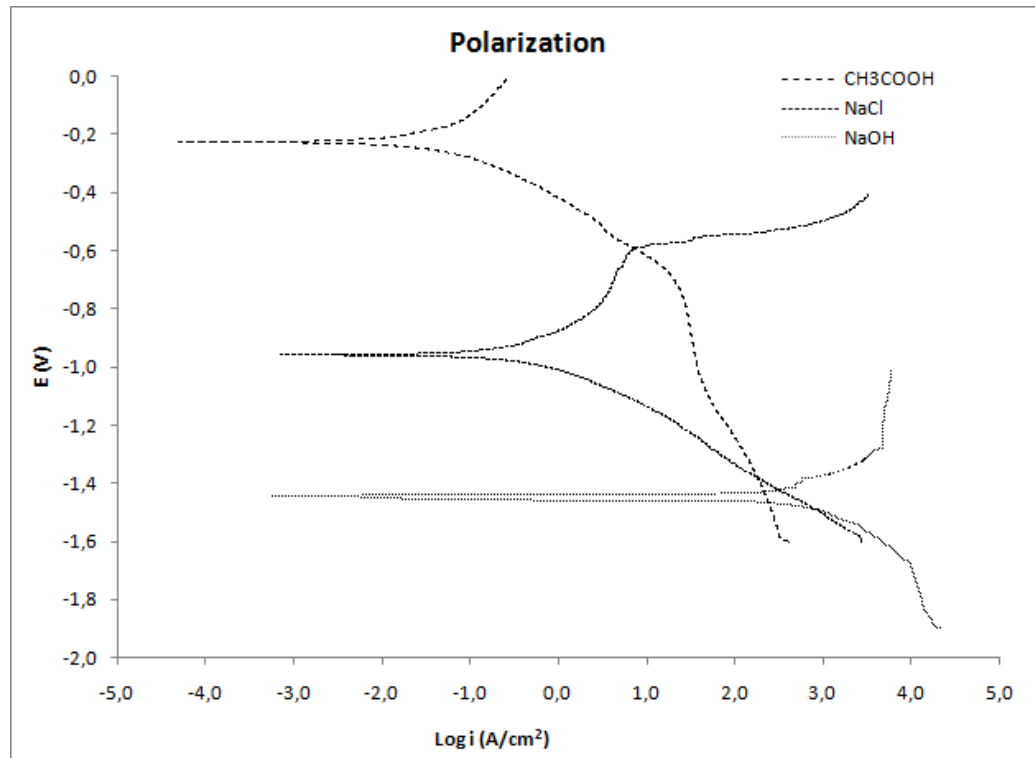
**Figure 1 –Polarization curves of the samples in the solutions**

The figure shows a general behavior of the alloy in each solution. It's possible to state about each curve's corrosion potential, that the most aggressive solution is the alkaline, followed by the saline and the less aggressive is the acid one. By analyzing the curves separately, using Tafel lines, it's possible to calculate the corrosion current to each case. The figure below shows the Tafel lines to the polarization curve for the acidic solution.
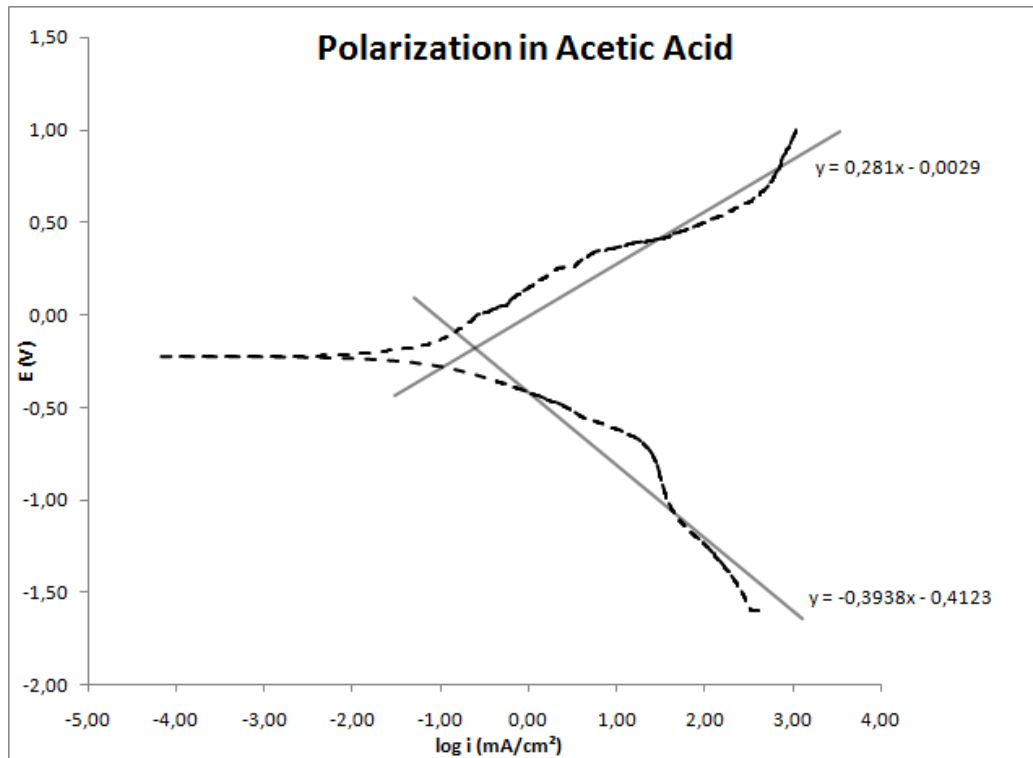
**Figure 2 - Tafel lines of the anodic and cathodic curves of the metal in acetic acid**

The intersection point of the two Tafel lines indicates the logarithm of the corrosion current, **log $i_{corr}$ = -0,5584 mA/cm²**. The figure below shows the intersection of the Tafel lines to the saline solution.
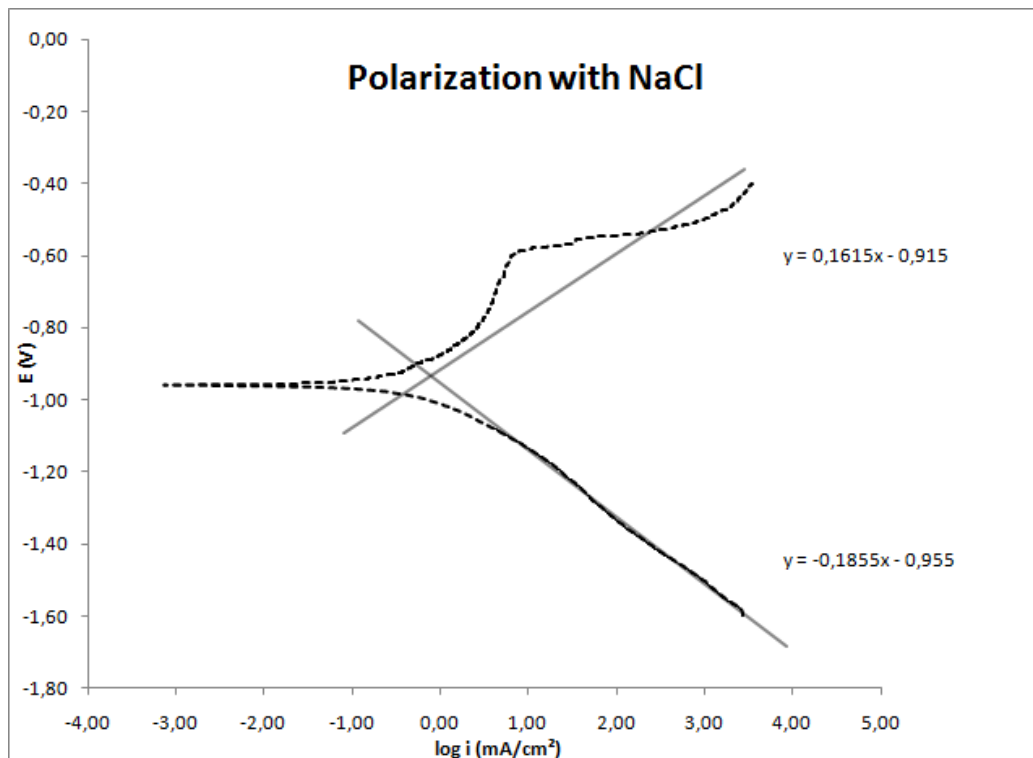
**Figure 3 - Tafel lines of the anodic and cathodic curves of the metal in a neutral solution**

The intersection point of the two Tafel lines indicates the logarithm of the corrosion current, **log $i_{corr}$ = -0,2772 mA/cm²**. The figure below shows the intersection of the Tafel lines to the alkaline solution.
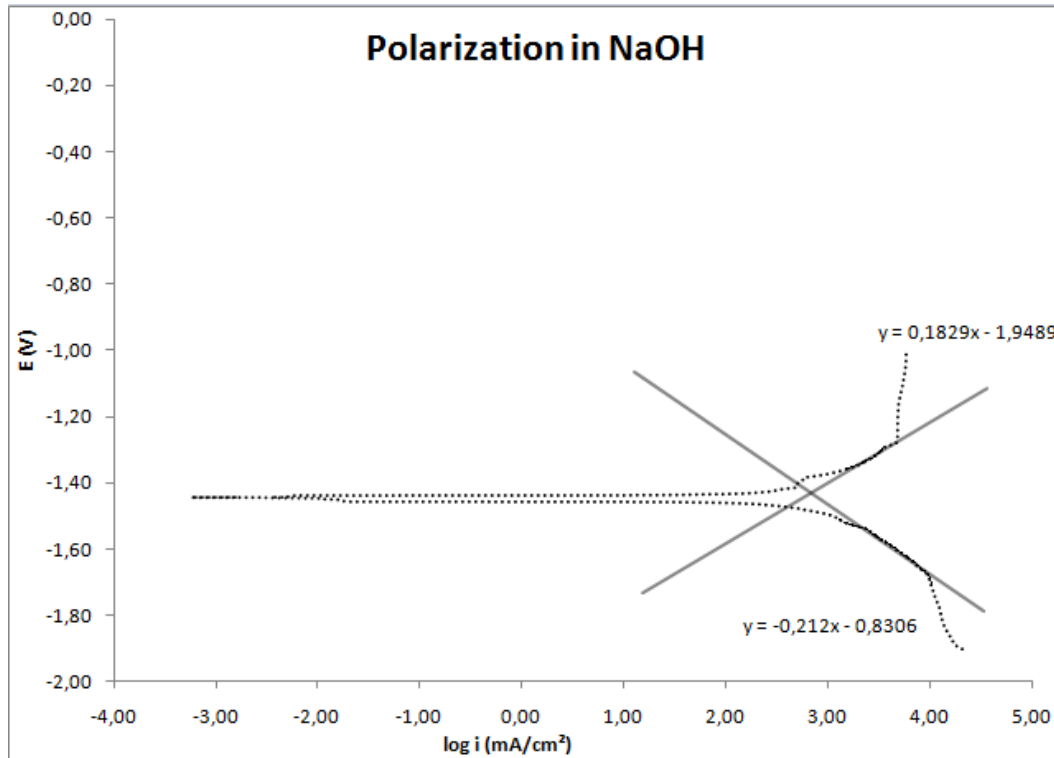


**Figure 4 - Tafel lines of the anodic and cathodic curves of the metal in NaOH**

The intersection point of the two Tafel lines indicates the logarithm of the corrosion current, **log $i_{corr}$ = 2,7860 mA/cm²**. The table below compares the results obtained with the polarization curves.

**Table 4 - Comparison between the current corrosions in the studied solutions**

| Solution | Characteristic | $i_{corr}$ (mA/cm²) |
|---|---|---|
| $CH_3COOH$ | Weak Acid | 0,27 |
| NaCl | Neutral | 0,52 |
| NaOH | Strong Base | 610,9 |

As the acetic acid is a weak organic acid, it has no power to dissolve the natural oxide layer of aluminium, even with its amphoteric behavior. It leads to a low corrosion current and consequently to a low corrosion rate. The reaction would form aluminium acetate and water. But as the acetic acid is weak and very diluted, in general it doesn't attack the metal.

In the case of the saline solution, the present ions make the environment more aggressive. However, among the common metals, aluminium presents the best resistance to sodium chloride. Salt favors pitting corrosion. The pitting density decreases with the raise of salt concentration in the solution. As well, as expected to a saline solution, it was evidenced in the curve a protrusion relative to pitting corrosion.

In the third case, the alkaline solution, the natural aluminium oxide layer is dissolved. It leads to a considerable raise of corrosion current and consequently at corrosion rate of the metal, which remains exposed to corrosive environment.

**Tribology**

The tribology measures were analyzed according to the potential variation before, during and after the abrasion procedure. First the sample was in contact with the solution for 20 minutes, allowing the potential measure to be generated by the combination alloy/solution. After this stage, the next 20 minutes was a continuous abrasion at 2 Hz, in a 2 mm track. Finally, after this second stage, a new potential measure was made during 20 minutes. A fourth sample in acetic acid was analyzed with an abrasion time of 10 minutes and 1,5 N, just for comparison purposes. This comparison is shown below:
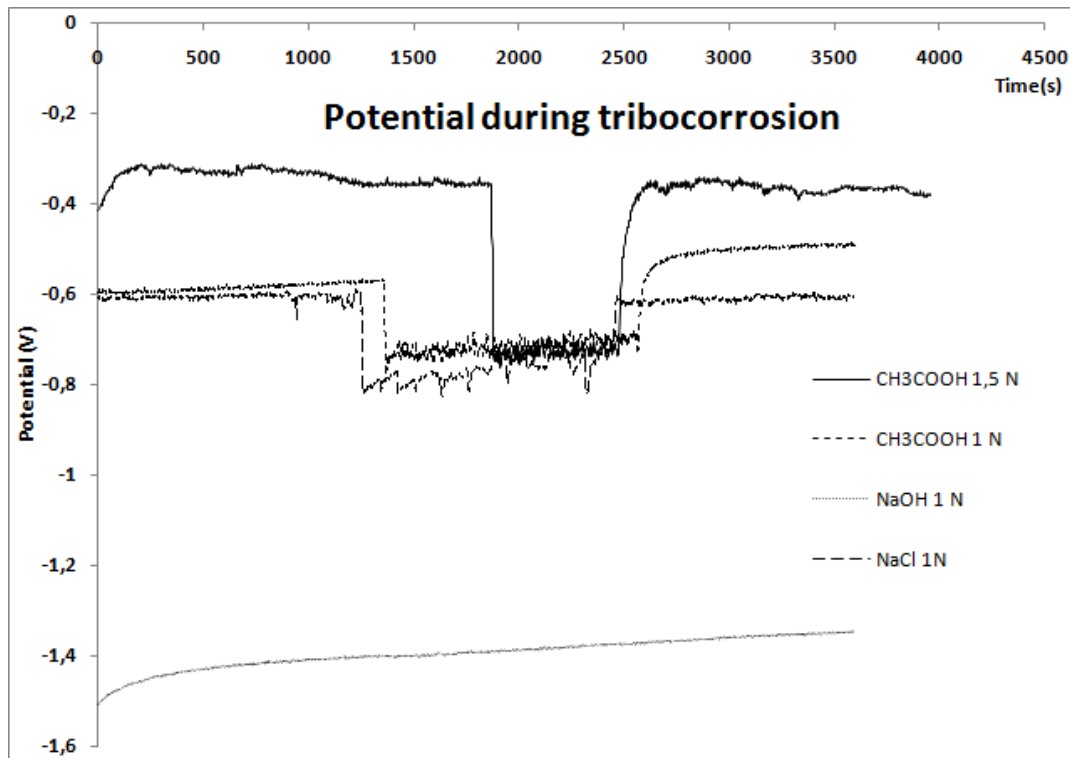


**Figure 5 - Open circuit potential curves of all samples**

It's possible to notice that in both cases with acetic acid, there is a potential decrease during the abrasion. That's because the protector aluminium oxide layer was broken. At the beginning of the analysis, the oxide thickness was different, but over time the curves are headed to same value on balance with the solution.

In contact with the saline solution, the behavior was very similar to the acid solution. However, the measures during the abrasion were more unstable, indicating a quicker formation of the oxide layer. In this way, each time the track was made, a little oxide layer was formed.

In the alkaline solution, the environment even allows the oxide layer to form. In a sufficiently alkaline solution it's dissolved and the metal becomes exposed to corrosion. It's noticed that the abrasion doesn't chance the sample's potential.

**Coefficient of Friction**

During the essays of tribology, the coefficients of friction were extracted between the alumina sphere and the analyzed samples. A comparison of the values is found at the graphic below:
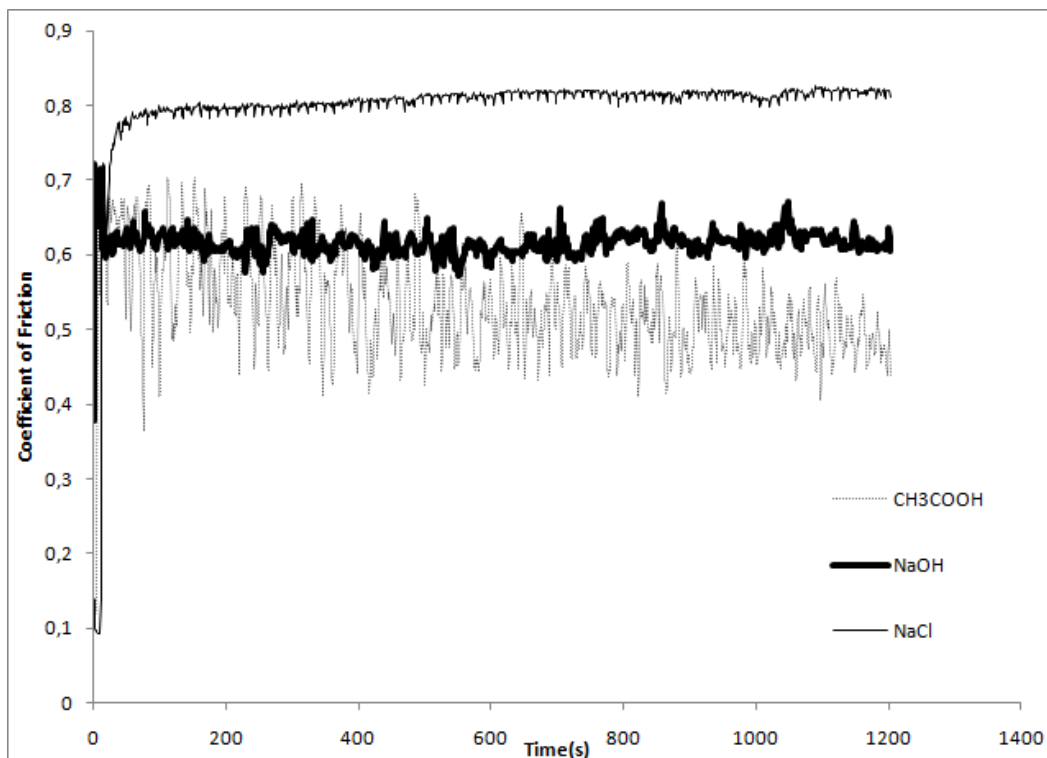


**Figure 6 - Comparison between the coefficients of friction**

The applied force by the equipment influences directly at the coefficient of friction and at the curve stability for each solution. In acidic solution, the curve is very unstable, what shows the

influence of the constant formation of the oxide layer during the abrasion process. In alkaline solution, as the oxide layer is dissolved, the coefficient of friction varies a lot less. Finally, in the saline solution there is certain stability. This may occur because the environment is less aggressive and there is a quicker formation of the oxide layer.

**Atomic Absorption Spectroscopy**

This analysis objective is to determinate if the quantity of lixiviated aluminium, during the contact with the solution and the abrasion, exceeds the limits established by the OMS. The table below shows the values found at the spectroscopy analysis.

**Table 5 - The concentrations obtained after tribocorrosion analysis**

| Parameter | Unit | Result | | | | DL* |
|---|---|---|---|---|---|---|
| | | CH³COOH (10 min.) | CH³COOH (20 min.) | NaOH (20 min.) | NaCl (20 min.) | |
| aluminium | mg/L | 1,62 | 2,58 | 150 | ND | 0,077 |

*Detection limit.

As stated in this word introduction, the limit established by the OMS to PTWI of aluminium is 1 mg/kg of body weight. Considering a 70 kg individual, he would have a 70 mg intake for week and the following hypotheses:

- Thirty minutes of cooking time;
- One tool harder than the aluminium to use;
- 1 L of liquids for meal;
- One meal a day;
- An aluminium pot with 20 cm of diameter;
- To stir eleven times with a 1 N minimal force.

The table below shows the weekly intake if the hypotheses above are followed:

**Table 6 - Concentration values for the hypotheses in each solution**

| Parameter | Unit | Result | | |
|---|---|---|---|---|
| | | CH³COOH | NaOH | NaCl |
| aluminium | mg | 102,06 | 9450 | < 4,851 |

The table shows that, considering the hypotheses, the quantity of aluminium ingested with an acidic solution and with an alkaline solution exceeds the PTWI established by the OMS. In the acidic case, the limit is exceeded around 46% the limit. In the alkaline case, the limit is absurdly exceeded in 12.500%. In the saline solution, the lixiviation of aluminium is minimal,

so much that it doesn't reach the limit of detection of the equipment. So, the maximum it could achieve in this case is less than 10% of the established limit, therefore the best environment to use to cook in aluminium pots.

## Conclusion

It's possible to state from this study that the use of aluminium pots must be restricted and made in a few specific cases. If used deliberately, the aluminium pot has huge chances of lixiviating in the solution (food) high quantities of particles, above the limit established by the OMS. This study didn't consider high temperatures as parameter, which alarms to a more dangerous situation, because found values at low temperature are already high and it's known that with temperature raise the solutions become more aggressive, have more dissolution power and solubilization capacity, and in this conditions the issued vapor increases the severity of wear in the pot.

According to the values found, foods which leads the environment alkaline or acidic should not be cooked in aluminum pans. If done, with one meal it overcomes the PTWI. The use of tools harder than aluminium (steel spoon, for example) should be avoided, because it helps to remove the protector oxide layer and exposes the metal. The neutral solution with chlorides was the only one to stay under the limit. Considering the use of some soft tool to cook, in ambient temperature, the lowest aluminium quantity ingested is in this case.

## Bibliographic References

(1) KAWAHARA, M.; KATO-NEGISHI, M.; Link **between Aluminum and the Pathogenesis of Alzheimer's Disease**: The Integration of the Aluminum and Amyloid Cascade Hypotheses, 2011.

(2) MCLACHLAN, D. R.; KRUCK, T. P,; LUKIW, W. J.; KRISHNAN, S. S.; Would decreased **aluminum ingestion** reduce the incidence of Alzheimer's disease?, 1991.

(3) ASSOCIAÇÃO BRASILEIRA DO ALUMÍNIO, **alumínio e saúde**, www.abal.org.br.

(4) ORGANIZAÇÃO MUNDIAL DA SAÚDE, **aluminium in drinking water**, 2010.

(5) U.S. FOOD AND DRUG ADMINISTRATION, **approximate pH of foods and food products**, 2007.

(6) DANTAS, S. T.; SARON, E. S.; DANTAS, F. B. H.; YAMASHITA, D, M.; KIYATAKA, P. H. M.; **determinação da dissolução de alumínio durante cozimento de alimentos em panelas de alumínio**, 2007.

(7) ASM HANDBOOK, **properties and selection: nonferrous alloys and special-purpose materials**, vol. 2, p. 53.

(8) VARGEL, C.; JACQUES, M.; SCHMIDT, M. P.; **corrosion of aluminium**, 2004.