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Risk Assessment of Using Aluminum Foil in Food Preparation

Ghada Bassioni[1,2,*], *Fathia S. Mohammed*[3] , *Essam Al Zubaidy*[3] *and Issam Kobrsi*[2]

[1] Faculty of Engineering, Ain Shams University, Cairo, Egypt.
[2] Chemical Engineering Department, The Petroleum Institute, Abu Dhabi, UAE;
[3] Chemical Engineering Department, American University of Sharjah, Sharjah, UAE.
[*]E-mail: gbassioni@pi.ac.ae

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The contamination of food is a major concern not only for developing countries but also for the entire world. In the present work, leaching of aluminum from aluminum foil in different food solutions was studied. Minced meat was used to prepare six different food solutions using tomato juice, citric acid, apple vinegar, salt, and spices. Three techniques for analysis were used, weight loss (W_L) measurements, Environmental Scanning Electron Microscopy (ESEM), and Inductively Coupled Plasma – Mass Spectrometry (ICP-MS). The results clearly indicate that the use of aluminum foil for cooking contributes significantly to the daily intake of aluminum through the cooked foods. The amount of leaching was found to be high in acidic solutions, and even higher with the addition of spices. According to the World Health Organization (WHO), the obtained values considered to be unacceptable. Finally, excessive consumption of aluminum from leaching aluminum foil has an extreme health risk effects. Aluminum foil may be used for packing but not for cooking.

Keywords: Aluminum foil; aluminum leaching; food preparation; aluminum level; toxicity

1. INTRODUCTION

Minimal exposure of aluminum to our bodies is not a problem. Human bodies can excrete small amounts very efficiently; an aluminum tolerable daily intake of 1 mg/kg body weight /day has been established by the World Health Organization (WHO) of the United Nation (UN) [1]. But unfortunately due to many reasons, most of us get exposed to and ingest more than what our bodies can handle [2]. It is reported that aluminum salts can be absorbed by the gut and concentrated in various human tissues including bone, parathyroid, and brain. High concentrations of aluminum have been detected in brain tissues of patients with Alzheimer's disease. Various reports have suggested that high aluminum intakes may be harmful to some patients with bone diseases or renal impairments. The aluminum health effects are far too vast to even being summarized [3]. Aluminum reduces the growth rate of human brain cells. Growth rate decrease becomes more pronounced at higher aluminum concentration [4].

Nowadays, it is a common practice to wrap meat and fish prior to oven cooking. Due to the possible relation between aluminum uptake and the specific diseases mentioned in many literatures, it is important to determine the aluminum concentration in the food wrapped with aluminum foil. The levels of aluminum content in different types of meat (beef, water buffalo, mutton, chicken, and turkey) packed with aluminum foil and cooked in an oven at three different temperatures/time periods (150 $^{\circ}$C for 60 minutes, 200 $^{\circ}$C for 40 minutes, and 250 $^{\circ}$C for 20 minutes) have been studied. The results indicate that cooking increases the aluminum concentration in both white and red meats by 89 – 378 % in red meats and 76 – 215 % in poultry. The least increase (76 – 115 %) is observed in the samples baked for 60 min at 150 $^{\circ}$C, while the highest increase (153 – 378 %) is in the samples baked for 20 min at 250 $^{\circ}$C. It is determined that the fat content in meat, in addition to the cooking process, affected the migration of aluminum. It has been also found that raw chicken and turkey breast meat contained higher amounts of aluminum than the raw chicken and turkey leg meat, respectively. However, meals prepared in aluminum foil may carry a health risk in addition to other aluminum sources. The aluminum contents in grilled and baked fish fillets, with and without ingredients, wrapped in aluminum foil have been also studied [5]. The aluminum migration seems to depend on several factors, e.g. grilling duration, heating temperature, composition, food pH value, and presence of any other substances (such as organic acids and salt) [6]. Other studies indicate that the aluminum leaching for 1 – 3 hours beef baked samples in aluminum foil ranged between 59.83 – 220.20 mg/kg [7].

Aluminum is found to leach out from the foil in different stimulants; particularly in distilled water as well as acidic and alkaline media. The migration is found to be above the permissible limit. Leaching of aluminum is found significantly higher in acidic and aqueous media in comparison to alcoholic and saline media. Higher temperature conditions also enhance the rate of migration of aluminum in acidic and aqueous media [8].

In recent years, there was a continuous use of aluminum foil in cooking process despite the fact that aluminum is associated with some diseases. The aim of this research is to detect leaching levels of aluminum from aluminum foil in different food solutions.

2. EXPERIMENTAL PART

2.1. Materials

Aluminum foil is taken from the local UAE market. Specimens of aluminum foil are cut into rectangular dimensional shapes of 8 x 4 cm, each with a 1 mm diameter hole at one end in order to hang them while inserting them into the different food solutions. In addition to that, other samples are cut into 2.5 x 2 cm to study the leaching process in the vapor phase. Besides that, other samples, with exposed diameters (D) of 22, 30, and 40 cm, are cut to cover the cooking pan that is used in the oven. Minced beef, tomato juice, citric acid, salt, apple vinegar and spices are used in different concentrations.

If not otherwise indicated, the experiments are performed at boiling point using drinking water. Some tests are performed with tap water, while others are performed at a temperature of 185 $^{\circ}$C.

2.2. Methods

The experiments of aluminum leaching from aluminum foil are performed in liquid and vapor phases at both boiling temperature and in the oven.

2.2.1. Weight loss measurement (W_L)

2.2.1.1. Liquid phase

The aluminum foil specimens of 8 x 4 cm are weighed using a four-digit sensitive weight balance and immersed completely in different food solutions. The meat solutions in drinking and tap water are boiled for one hour, followed by filtration to recover the meat extract. The volume is adjusted to give a 40 % of extract concentration. Different additives are used to prepare the following food solutions:

Solution (1): 250 ml of 40 % meat extract + 250 ml of tomato juice + 10 g of citric acid + 5 g of salt.

Solution (2): 250 ml of 40 % meat extract + 250 ml of tomato juice + 20 g of citric acid + 5 g of salt.

Solution (3): 250 ml of 40 % meat extract + 250 ml of tomato juice + 20 g of citric acid + 5 g of salt (tap water).

Solution (4): 250 ml of 40 % meat extract + 250 ml of tomato juice + 30 ml of apple vinegar + 5 g of salt.

Solution (5): 250 ml of 40 % meat extract + 250 ml of tomato juice + 20 g of citric acid + 5 g of salt + 3 g of spices.

Solution (6): 500 ml of 40% meat extract + 20 g of citric acid + 5 g of salt.

The pH of each solution is measured before the experiment. The aluminum foil samples are immersed in the boiling solution for two hours. After the experiment, the samples are cleaned by distilled water followed by acetone and finally are weighed.

2.2.1.2. Vapor phase

The 2.5 x 2 cm aluminum foil samples are exposed to the following food solutions:

Solution (1): 250 ml of 40% meat extract + 250 ml of tomato juice + 3 g of citric acid + 5 g of salt.

Solution (2): 250 ml of 40% meat extract + 250 ml of tomato juice + 20 g of citric acid + 5g of salt.

The pH of each solution is measured before the experiment. Then the samples are hung in the vapor phase of the boiling solution for two hours. After the experiment, the samples are cleaned and weighed.

2.2.1.3. Oven work

The following food solution is added to a 22 cm diameter Indian aluminum pan: 375 ml of 40 % meat extract + 375 ml of tomato juice + 30 g of citric acid + 7.5 g of salt and is covered by an aluminum foil. The exposed part of the foil is circular of the same diameter. The food is cooked in an oven at 185 $^{\circ}$C for 1.5 hours. The aluminum pan and the aluminum foil are cleaned and weighed before and after the cooking process. This procedure is repeated using 30 and 40 cm diameter pans.

To assure consistency, all the experiments are performed in duplicates.

2.2.2. Environmental scanning electron microscopy (ESEM)

Environmental scanning electron microscopy (ESEM) coupled with energy dispersion x-ray (EDX) measurement is used to perform the analysis of the aluminum foil samples before and after the cooking process. The results of the analysis help in indicating any leaching of metals from the original samples to the food.

2.2.3. Inductively coupled plasma – mass spectrometry (ICP – MS)

The concentration of aluminum dissolved in the solutions after cooking is analyzed using inductively coupled plasma – mass spectrometry (ICP – MS).

3. RESULTS AND DISCUSSION

3.1. Weight loss measurement – liquid and vapor phase

It is getting more common to use aluminum foil to wrap meat, chicken, potatoes, and some vegetables prior to oven cooking. The result of this work aids in determining whether or not aluminum foil leaches aluminum to the food. Related to the type of experiment performed, the aluminum foil sheets that are used have different shapes and dimensions. The corrosion rate ($\boldsymbol{CR}$) in mg.cm^{-2}.hr^{-1} is calculated using the following equation:

$$\boldsymbol{CR} = \frac{\mathrm{WL}}{\mathrm{A\,.\,T}}$$

WL is the weight loss in mg, A is the surface area of the specimen in cm^{2}, and T is the exposure time in hr.

The calculated aluminum intake per person is based on the assumption that the meals prepared are for a family composed of three members. The weight loss is found by using a sample ($10cm^2$) extrapolated to the area of the samples of 22, 30 & 40 cm diameter. The total weight loss divided by 3 gives the aluminum intake/person.

The calculation of aluminum leaching in liquid phase of solution (1) can be calculated as follows:

The weight loss from the aluminum foil sample of a total surface area of 10 cm^2 equals 1.8 mg in two hours exposure. For an aluminum foil sample of 22cm diameter, the total surface area is equal to $\pi/4*d^2 = 380cm^2$. The weight loss is equal to 68.4 mg. The aluminum Intake/person is equal to 22.8 mg.

Considering using aluminum foil for oven cooking, aluminum foil sample with dimensions of 60 x 40 cm is used with exposed surface area of 2400 cm^2. The detailed experimental results are summarized in Table (1). The variation of leaching of aluminum in two different food solutions is reported in Figure (1).

Table 1. Effect of different food solutions at boiling temperature on the leaching of aluminum foil in liquid and vapor phases using drinking water with exposure time of 2 hours (exposed area = 10 cm^2) [1]

Solution Number / Exposure Type	pH	Weight Loss (mg)	Corrosion Rate (mg/cm^2 * hr x 10^{-2})	Intake (mg/person)		
				D = 22 cm	D = 30 cm	D = 40 cm
1 / Vapor	3.7	1.7	8.5	21.1	39.4	69.6
1 / Liquid	3.7	1.8	9.0	22.8	42.4	75.4
2 / Vapor	2.8	3.3	16.6	42.1	78.1	139.2
2 / Liquid	2.8	3.2	15.8	40.0	74.4	132.4

[1] Results calculated on the basis of 22, 30, and 40 cm diameters, respectively.
Solution (1): 250ml 40% meat extract + 250ml tomato juice + 3g citric acid + 5g salt
Solution (2): 250ml 40% meat extract + 250ml tomato juice + 20g citric acid + 5g salt

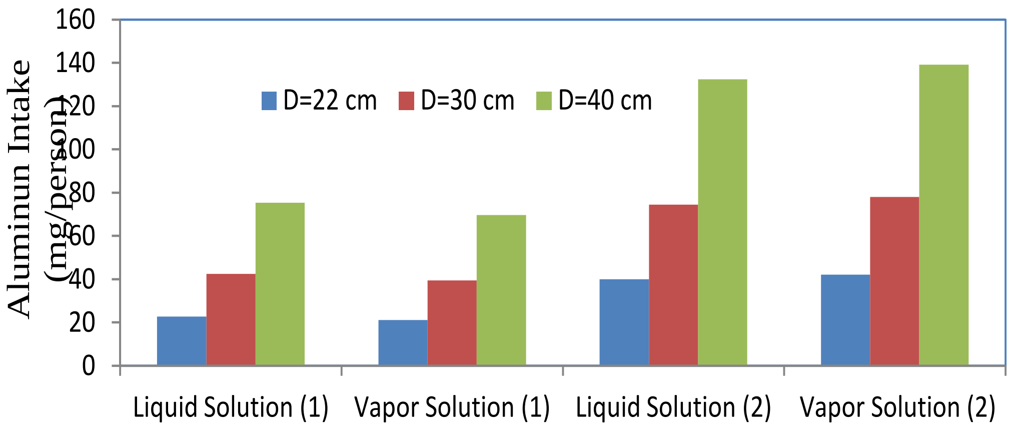


Figure 1. Effect of different food solution on the leaching of aluminum foil in liquid and vapor phase

From the above results, it is clear that the leaching of aluminum foil in solution (2), which contains higher acid concentration, is about double the leaching in solution (1). The aluminum intake for solution (1) is 132.4 mg/person in the liquid phase compared to 75.4 mg/person for solution (2) in the liquid phase (sample of 40 cm diameter). The pH of solution (2) is 2.8 while that of solution (1) is 3.7. These results emphasize the previous work which reported that the amount of aluminum, accumulated in food during the preparation process, depends on the pH value and the cooking time [6]. For the same solutions above, there is no considerable difference between the leaching of aluminum foil in both vapor and liquid phases. The leaching in solution (1) is 75.4 mg/person in the liquid phase and 69.6 mg/person in the vapor phase.

So, one can use the leaching value in any available phase to reflect the other phase. In the above experiment, an aluminum foil sample with a surface area of 10 cm^2 was left in boiling food solution for 2 hours. On the other hand, another aluminum foil sample with the same area is exposed to the vapor of the same solution for the same period of time.

The results presented in Table (2) and Figure (2) with the contact surface area of 64 cm^2 indicates the following behavior:

Table 2. Effect of different food solutions at boiling temperature on the leaching of aluminum foil (assumption that the diameter of the foil used is 22 cm (area = 380 cm^2))

Solution Number [2]	pH	Water Type	Weight Loss (mg)	Corrosion Rate (mg/cm^2 * hr x 10^{-2})	Intake (mg/person)		
					D = 22 cm	D = 30 cm	D = 40 cm
1	3.2	Drinking	16.7	13.1	33.05	61.5	109.4
2	2.8	Drinking	20.2	15.8	40.0	74.4	132.4
3	3.2	Tap	9.9	7.75	19.7	36.7	65.2
4	4.2	Drinking	70.9	55.5	140.3	261	465.0
5	3.0	Drinking	82.0	64.1	162.3	302.0	537.2
6	-	Drinking	18.7	14.61	37.0	68.8	122.2

[2] Solution (1): 250ml 40% meat extract + 250ml tomato juice + 10g citric acid + 5g salt.
Solution (2): 250ml 40% meat extract + 250ml tomato juice + 20g citric acid + 5g salt.
Solution (3): 250ml 40% meat extract + 250ml tomato juice + 20g citric acid + 5g salt (tap water).
Solution (4): 250ml 40% meat extract + 250ml tomato juice + 30ml apple vinegar + 5g salt.
Solution (5): 250ml 40% meat extract + 250ml tomato juice + 20g citric acid + 5g salt + 3g spices.
Solution (6): 500ml 40% meat extract + 20g citric acid + 5 g salt.

Comparing the behavior of aluminum foil in solutions (1) and (2), it is clear that by doubling the amount of citric acid added an increase of 20% in weight loss is observed. The leaching values, on the basis of D = 40 cm, are 109 mg/person for solution (1) and 132 mg/person for solution (2). This result shows the same trend regarding the pH which is 3.2 and 2.8 for solutions (1) and (2), respectively. Some experimental results obtained by previous works demonstrate that the complexation effect takes a very important role in the process of aluminum leaching from cooking utensils. Increased concentrations of complexing ions (organic acid, fluoride ions, or OH^-, etc) significantly enhance the release of aluminum [9].

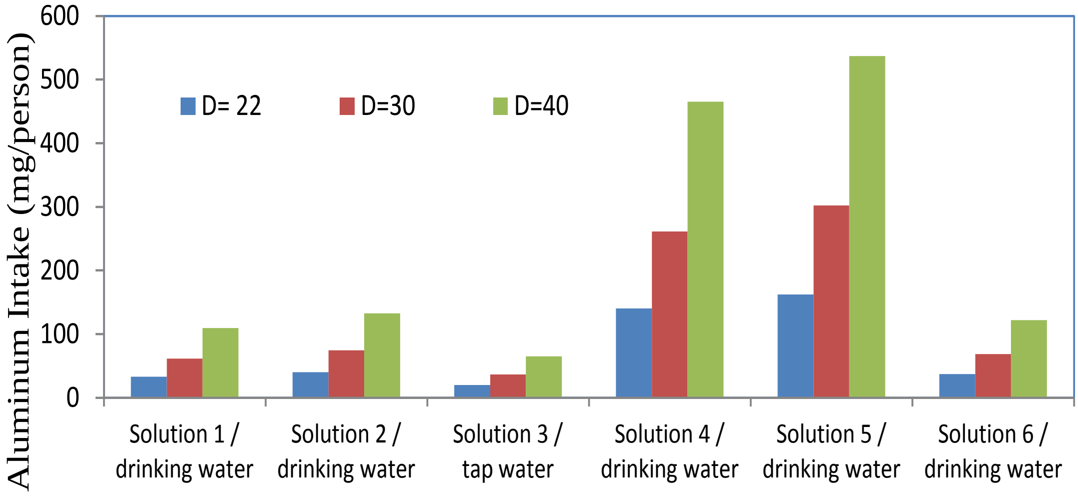


Figure 2. Effect of food solution, type of water, and diameter of the aluminum foil on the aluminum intake/person

The results for solution (2) and solution (3), which have the same composition but are prepared with different types of water, show that the weight loss from aluminum foil in solution (3) is half of that in solution (2). The rate of leaching in solution (2) is 132.4 mg/person while that in solution (3) is 65.2 mg/person. These results are in agreement with the discussion regarding the value of pH in the previous section.

The pH of solution (3) is 3.2 while that of solution (2) is 2.8. The migration of aluminum from aluminum foil into food-simulating solvents has been reported to be higher than in case of tap water [10]. This is in agreement with our findings.

Solution (2) and solution (4) differ only in the acid used. Solution (2) contains citric acid, while solution (4) contains apple vinegar. The pH of solutions (2) and (4) are 2.8 and 4.2, respectively. The weight loss in aluminum foil immersed for 2 hours at boiling temperature in solution (2) is 20.2 mg and that in solution (4) is 70.9 mg. All other parameters such as the temperature of the solution, the type of water used for preparation, the duration time, and the amount of salt used remains the same. The aluminum leaching intake is 465 m/person using for solution (4) compared with the value of 132.4 mg/person for solution (2). This means that a person in this case might ingest in one meal what he/she is allowed to take in 7 days, which resembles a very high health risk.

A reasonable cause can be related to the availability of OH^- ions in solution (4) and the increase in aluminum leaching. Moreover, it has been reported that the composition of food determines the amount of aluminum leaching [11]. It is also well established that aluminum dissolution is highly dependent on the pH, the temperature, and the presence of complexing agents. Aluminum exhibits a passive behavior in aqueous solutions due to the protective compact of Al_2O_3 film on its surface. However, the solubility of this protective film increases in different acidic and alkaline media [12].

Aluminum leaching in aqueous solutions may be explained by the following chemical reaction occurring on the surface of the aluminum cookware [9]:

$$Al_2O_3 + 6H^+ \rightarrow 2Al^{+3} + 3\ H_2O$$

The free Al^{+3} ions in solution react with the organic acids and the other complexion agents found in the food. The present result could be explained by the fact that the dissolution of aluminum may change the local pH on the aluminum surface, but it will not affect the measured pH of the solution [13]. Aluminum migration into acidic solvent when heated for 30 minutes at 95 °C was higher than that in the tap water and ranged from a factor of 60 for 0.5 % aqueous citric acid to a factor of 200 for 4 % aqueous acetic acid concentration. The migration levels in all heated samples (30 minutes at 95 °C) and acidic foods were less than that in 4 % acetic acid concentration. The apple vinegar, used in the current work, had a 5 % acetic acid concentration. In addition to that, it was reported that citric acid inhibits the corrosion of aluminum in sodium chloride solutions and the inhibition efficiency depends on its concentration. The protection efficiency increases with citric acid concentration up to a critical value of 1.0×10^{-5} M. At higher concentrations, the inhibition efficiency is reduced again with increasing acid concentration [14]. They attributed the mechanism of inhibition to the adsorption of citric acid onto the metal surface [15]. The above explanation could justify the values reported in this work.

Solution (5) contains 3 g spices compared to solution (2), the pH of these solutions are 3 and 2.8, respectively. Leaching of the aluminum foil in solution (5) is 4 times the leaching found in solution (2). Compared to 132.4 mg/person for solution (2), the intake values from solution (5) in food is found to be 537.2 mg/person. This value is also considered high and indicates a very high health risk to the consumers. The cause of this big increased leaching value is the presence of spices. This is in agreement with the previous work [15-17]. In cooking process, people are used to add tomato paste, lemon juice, table salt, and other spices which cause more leaching of aluminum from the cook ware.

Analysis and comparison of the results regarding solution (6) with the reference solution (2) showed that the addition of tomato juice has no effect on the leaching process of aluminum from the aluminum foil in solutions already containing citric acid and salt. This is probably because the addition of tomato juice does not change the pH value of the solution. The intake value is found to be 122.2 mg/person in solution (6) compared with 132.4 mg/person in solution (2). Adding tomato juice to the pure meat extract solution with no acid and salt caused a 100 % increment in the leaching of aluminum.

The pH value of the 40 % meat extract solution is 6.9 while the pH value of the solution of the meat extract and tomato juice is 4.9 [16].

3.1.1. ESEM Results

Figures (3) and (4) show a magnified picture of the blank surface of aluminum foil and the surface of aluminum foil after cooking for 2 hours in solution (2) (Table 2), respectively. The treated sample showed some holes due to the dissolution of aluminum into the food.

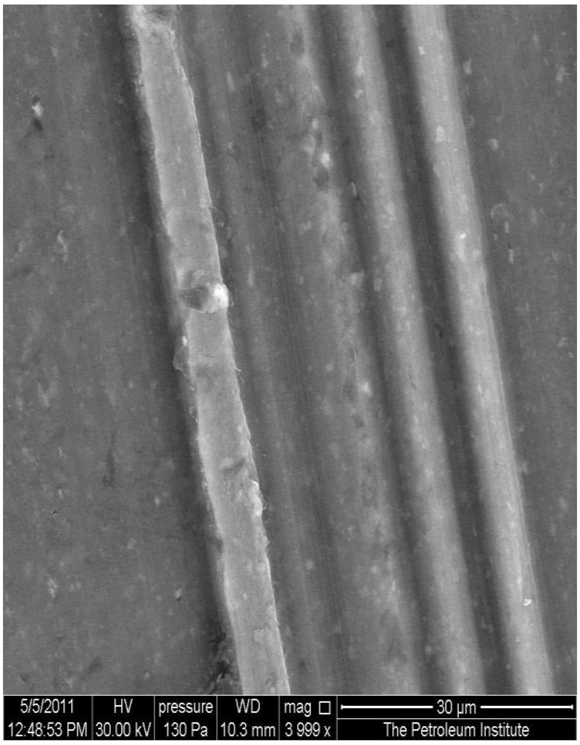


Figure 3. ESEM image of aluminum foil without treatment (blank).

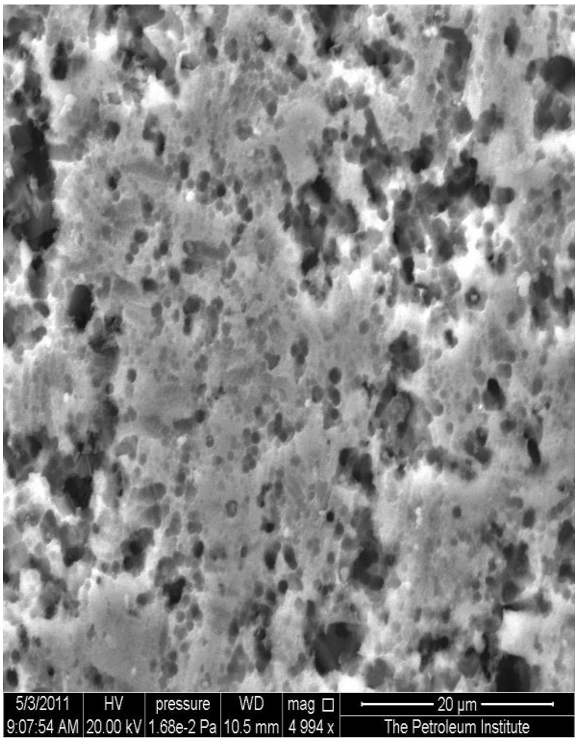


Figure 4. ESEM of an aluminum foil immersion in solution (2) for 2 hrs at boiling temperature

3.2. Weight loss measurement - oven work

As mentioned earlier in the experimental part, few experiments were conducted using aluminum pan, half filled with food solutions and covered by aluminum foil. The pan was introduced in oven for 1.5 hours at a temperature equal to 185 °C.

The leaching from the leaching foil into the food is increasing linearly with the exposed surface area to the food vapor (Figure 5).

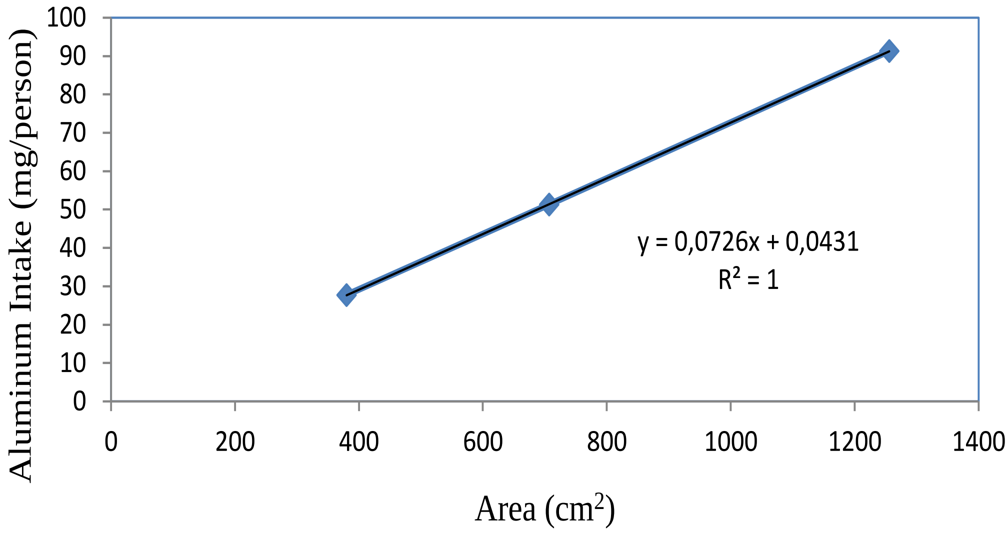


Figure 5. Relation between the aluminum circular pan area with aluminum intake/person

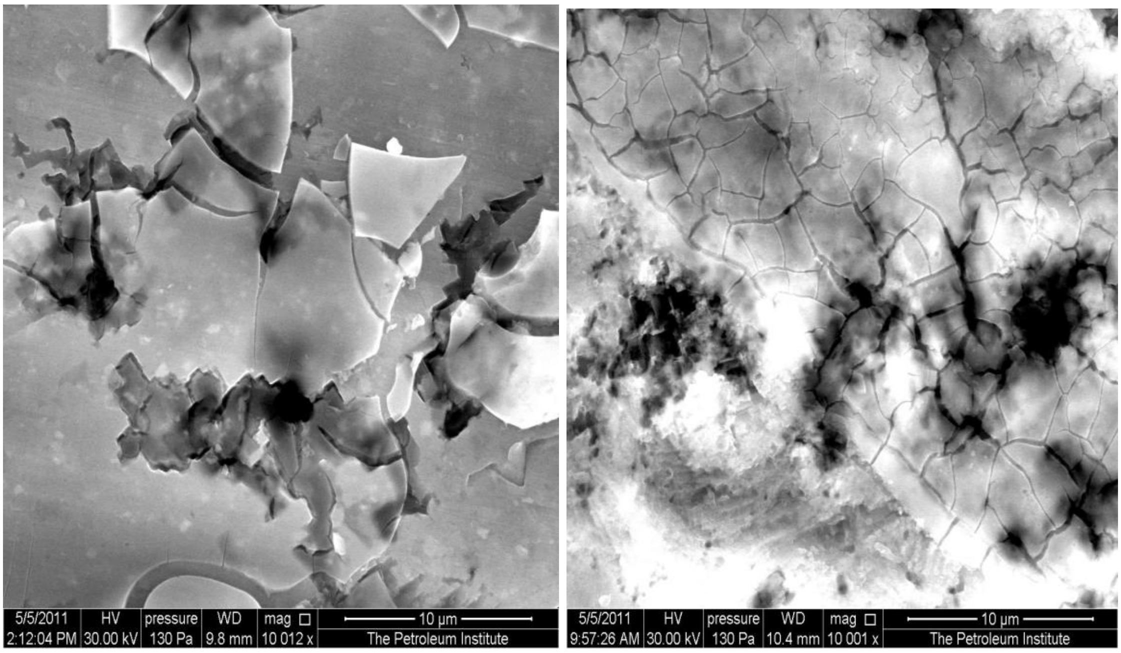
In a situation where we have rectangular piece of 60x40 cm, the leaching is 174.3 mg/person from the foil only. While using a circular foil with a surface area of 22 cm^2, the leaching is only 27.7 mg/person.

In both situations the leaching in such process is not only resulting from the aluminum foil covering the pan, but also from the pan itself. The leaching resulted from using a pan of 22 cm diameter in such experiment is 1000 mg, dividing this value by 3 as mentioned earlier gives a result of 333.3 mg/person. Adding of this value to the leaching from the aluminum foil covering the pan results a total value of 361 mg/person which can be regarded as a very high value. The values would be even higher when using a larger pan which requires a larger diameter of aluminum foil to cover it. Also, the leaching becomes even more when the aluminum foil is covering some of the above earlier mentioned food solutions and then introduced to oven at temperature of 220 – 250 °C for 1 – 1.5 hours. The aluminum leached from the foil wrapped grilled meat was higher than the foil covering the food cooked in the oven at 200 °C. It was reported that cooking temperature is more important in aluminum leaching than cooking time [5]. Higher cooking temperatures stimulated the leaching of aluminum from foil to meat because, at elevated temperatures, the oxide layer becomes thicker and changes from an amorphous to a crystalline structure. Using the different sides of aluminum foil (shiny and dull) showed that the differences in the aluminum leaching were not significant.

Aluminum foil is not suitable for cooking, and especially with acidic food. It is also possible that excessive consumption of food baked with aluminum foil may carry a serious health risk.

3.2.1. ESEM results

After the exposure of aluminum foil to the food solution in the oven at 185 °C for 1.5 hours, two different samples are examined as shown in Figures (6 A) and (6B). The damages are clearly shown on these two samples and especially at higher magnification. The figures also show that the aluminum foil is almost completely damaged. Higher cooking temperature stimulates the leaching of aluminum foil into the food and this is in agreement with the weight loss results reported earlier.



A **B**

Figures (6-A) and (6-B). ESEM images of an aluminum foil after cooking in an oven for 1.5 hrs at 185 °C (two different areas of the same sample with different magnifications).

3.3. ICP-MS results

The detailed metal analysis of the two food solutions, after using aluminum foil in the cooking process, using the ICP – MS shows that the aluminum content is 21160.9 and 19915.8 ppb for both solutions (1) & (2) respectively. The aluminum contents in both solutions are very high and are consistence with weight loss measurement results. These results also reflect the reason for the damage shown in the ESEM images.

Other details need to be considered, such as the pollution from heavy metals which has been increased recently and become an important environmental concern. These heavy metals are copper, mercury, lead, and other trace elements which may carry some risk to the human health.

4. CONCLUSION

The above work shows that the aluminum leaching from the foil into the food solution is the same in liquid and vapor phases. Aluminum foil used in cooking provides an easy channel for the metal to enter the human body. The increase in cooking temperature causes more leaching. The leaching is also highly dependent on the pH value of the food solution, salt, and spices added to the food solutions. Aluminum foil is not suitable for cooking specially with acidic food. It is also possible that excessive consumption of food baked with aluminum foil may carry a serious health risk.

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References

1. WHO (World Health Organization), "Safety evaluation of certain food additives and contaminants", WHO Food additives Series 46. Joint FAO/WHO Expert Committee on Food Additives (2001).
2. J. Damond, "A report on Alzheimer's disease and current research", Alzheimer Society of Canada (2005).
3. H. Gitelman, "Aluminum and health critical review", Marcel Dekker, Inc. New York (1989).
4. M. S. Kim, *Sci. Tot. Environ.*, 278 (2001) 127.
5. S. Turhan, *Meat Sci.*, 74 (2006) 644.
6. R. Ranaua, J. Oehlenschlägera and H. Steinhartb, *Food Chem.*, 73 (2001) 1.
7. E. J. Ekanem, J. A. Lori, F. G. Okibe, G. A. Shallangwa, B. A. Anhwange, M. Haliru and A. A. Moyosore, *J. Appl. Sci. Res.*, 5 (2009) 944.
8. P. Ojha, and P. Sharma, *J. Environ. Sci. Eng.*, 49 (2007) 62.
9. S. Bi, *Environ. Poll.*, 92 (1996) 85.
10. Y. T. Kawamura and T. Yamada, *J. Food Hygienic Soc. Japan*, 39 (1998 b) 178.
11. L. A. Al Jahaiman, *J. Saudi Chem. Soc.*, 14 (2010) 131.
12. H. Lenderink and M. J. de Wit, *Electrochem. Acta*, 38 (1993) 1989.
13. M. I. S. Verissimo, J. Oliveria and M. Gomes, *Sensors Actuators B* 118 (2006) 192.
14. G. Solmaz, B. Kardas and M. Erbil, *Corr. Eng. Sci. Tech.*, 43 (2008) 186.
15. F. S. Mohammed, E. A. H. Al Zubaidy, G. Bassioni, *Int. J. Electrochem. Sci.*, 6 (2011) 6424.
16. F. S. Mohammed, E. A. H. Al Zubaidy, and G. Bassioni, *Int. J. Electrochem. Sci.*, 6 (2011) 222.
17. F. S. Mohammed, E. A. H. Al Zubaidy, G. Yacoub, G. Bassioni, *Int. J. Electrochem. Sci.*, 7 (2012) 363.