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**ORIGINAL ARTICLE**

# Estimating Aluminum leaching from Aluminum cook wares in different meat extracts and milk

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Polarization methods

**Abstract** A method of estimating Al leaching from Al cook ware in some meat extracts and liquid milk was investigated. In the present work four kinds of Al cook wares from four countries were chosen from the local market. Extracts of boiled meat (lamb, chicken, and fish) were used to make 10–50% (w/v) concentrations. In addition liquid fresh milk and long life milk were diluted to make 10–50% (v/v) concentrations. Weight loss (WL) method, atomic absorption, polarization method, and surface study were applied. The "estimated" Al intake per person from WL in 30% meat extract and milk range from 8.16 to 12.75 mg/h with fish extract having the highest leaching and chicken extract having the lowest leaching. Atomic absorption gave comparable results to that of WL. Comparing the present results with the Provisional Tolerance Weekly Intake of Al approved by the FDA/WHO shows that Al leaching from Al cook wares may add high doses of Al into the diet.



## 1. Introduction

Aluminum is a non-essential metal to which humans are frequently exposed. Al can be toxic when injected directly to animals (Slanina and Falkeborn, 1984) or accidentally to humans as in the case of dialysis (Gitelman, 1989). Aluminum was regarded a neurotoxin agent since 1980 (Gitelman, 1989). Due to its accumulation in brain, bones, and liver Al was associated with some diseases like dialysis encephalopathy and bone disorder (Winship, 1993; Yokel, 1994). There is a continuing interest of Al content in food and its possible relation to Al toxicity especially to the elderly and to people with kidney failure (Winship, 1993; Fimreite et al., 1997; Soni et al., 2001; Ščančar et al., 2004).

Sources of Al entering the human body include food, water, beverages, cosmetics, medicines, food additives, and Aluminum leaching from Al cook wares. There is some disagreement about using Al cook wares in cooking. Some studies regard using Al utensil and Al foils safe for cooking (Ranau et al., 2001; Soni et al., 2001; Verissimo et al., 2006). Other studies regard them hazardous and do not advice using them especially in acidic food (Ščančar et al., 2004; Al-Mayouf et al., 2008; Fimreite et al., 1997). In Saudi Arabia there are a few studies which relate the daily habits of Al consumption with

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Al toxicity. An earlier study correlates Aluminum in blood serum for elementary school girls with the daily habits (Al-Saleh and Shinwari, 1996).

It is well established that Al dissolution is highly dependent on pH, temperature, and the presence of complexing agents. Al exhibits a passive behavior in aqueous solutions due to the protective compact $Al_2O_3$ film on its surface. However, the solubility of this protective film increases in acidic and alkaline medium. According to Bi (1996), Al leaching in aqueous solutions may be explained by the following chemical reaction occurring on the surface of the Al cookware:

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

The free Al ions in solution react with organic acids and other complexing agents found in food.

There is a continuous use of Al cook wares in different countries including Saudi Arabia despite its association with serious health problems. Thus the aim of this work is to shed some light on Al leaching in aqueous solutions containing meat extracts and liquid milk using different kinds of Al cook wares brought from the local market.

## 2. Materials and methods

### 2.1. Materials

Four kinds of Al cook wares were chosen from the local market from four different countries China, India, Syria, and Saudi Arabia. The samples were named S, Y, I, and C to avoid any misconceptions. In addition pure Al of 99.999% (from Good fellow, England) was used for comparison. Ground meat of lamb, chicken and fish were used from the local market. The liquid full cream milk (fresh and long life) was diluted to the desired concentrations to make 10–50% (v/v) solutions.

### 2.2. Weight loss method

In the present work weight loss method (WL) at 90 °C was used to study Al leaching in aerated food extract solutions as follows:

1. The four different Al cook wares were cut into rectangular specimens of dimensions 2 × 2.5 cm and 2 mm thickness. The average exposed area was 11.80-cm$^2$ ± 0.20. These specimens were treated in the following method prior to each experiment. Wet polishing with SiC up to 1200 grade, washing with distilled water, then ultrasonic cleaning in acetone (ultrasonic cleaner from Cole Palmer) for 2 min before drying.
2. Ground meat of lamb, chicken, and fish were weighed to make stock solutions of 50% (w/v). Then they were boiled in distilled water at 90 °C for 1 h to get the meat extract. Filtration of the meat extract solutions was followed by dilution to obtain the 50% (w/v) which was kept in the fridge for a maximum of 1 week before use.
3. The Al samples were weighed in a balance (± 0.0001 g from Mettler Toledo) then immersed in the hot meat extract at 90 °C for 1 h. All experiments were performed in aerated solutions and maintained at 90 °C ± 1.
4. After the leaching experiments the Al samples were immersed in a hot cleaning solution of $CrO_3$ + $H_3PO_4$ at 80 °C for 7 min to remove the reaction products from the surface. Finally, it was washed generously with distilled water then acetone. The dry Al sample was weighed again to determine the weight loss. All WL experiments were performed in duplicates. The pH of the meat extract was measured before and after the experiments.

### 2.3. Atomic absorption method (AA)

The remaining solutions after WL experiments were analyzed by AA technique using an instrument from Shimadzu (A.A. 6701F) to determine the amount of dissolved $Al^{3+}$.

### 2.4. Electrochemical method

This method was applied to estimate small changes of Al leaching using a sample holder (from Radiometer) connected to a galvanostat/potentiostat (from ACM). The Al cook wares were cut into circular disks of 1.4 cm diameter and an exposed area of 1.13 cm$^2$. The Al disk (as the working electrode) was fitted into a thermo stated sample holder cell. The reference electrode was SCE and the auxiliary electrode was Platinum. All electrochemical experiments were performed in aerated solutions and maintained at 60 °C ± 1 using a circulating water bath (from Haak). After performing open circuit potential for 1 h, polarization measurements were done to obtain corrosion current density from Tafel method.

### 2.5. Surface study

The surface morphology and surface analysis of Al samples were studied using scanning electron microscope (SEM) from (Joel-JSM-6060LV) connected with energy dispersion X-ray (EDX).

## 3. Results and discussion

### 3.1. Weight loss method

Although WL is an old technique, it is used to simulate Al leaching from Al cook wares. It is cost effective, quick, and environmentally friendly. The remaining of meat (mostly fibers) had little effect on Al leaching. An experiment of the WL method of the whole meat at 50% concentration was compared to the meat extract at 50% concentration and the difference was 2.7% only.

Corrosion rate (CR) from WL method was calculated using the following equation:

$$\text{Corrosionrate} = \frac{\Delta w}{A \times t}$$

where $\Delta w$ is the weight loss of Al alloy (mg) or the leaching amounts; $A$ is the surface area (cm$^2$), and $t$ is the immersion time (h).

CR in meat extracts and milk are listed in Tables 1 and 2. It is clear that there is a difference in Al leaching depending upon the composition of Al alloy which reflects the effect of alloying elements. It is shown from Tables 1 and 2 that the amount of Al leaching generally increased with increasing the concentration of meat extract.

**Table 1** CR of Al from WL after immersion for 1 h at 90 °C in different concentrations of meat extracts.

| Soln. extract | Al alloy | Conc. CR × $10^2$ (mg/cm$^2$ h) | | | | |
|---|---|---|---|---|---|---|
| | | 10% pH 6.73 pH 6.6 | 20% (w/v) pH 6.58 | 30% (w/v) pH 6.49 | 40% (w/v) pH 6.44 | 50% (w/v) pH 6.39 |
| Lamb | C | 2.43 | 3.28 | 3.25 | 3.31 | 3.45 |
| | I | 3.33 | 3.63 | 4.17 | 4.95 | 5.58 |
| | S | 1.91 | 2.53 | 2.85 | 3.35 | 4.53 |
| | Y | 2.33 | 2.73 | 3.01 | 3.21 | 3.43 |
| | | 10% pH 6.55 pH 6.6 | 20% (w/v) pH 6.40 | 30% (w/v) pH 6.31 | 40% (w/v) pH 6.27 | 50% (w/v) pH 6.22 |
| Chicken | C | 2.23 | 2.65 | 2.72 | 2.68 | 2.71 |
| | I | 2.73 | 2.66 | 3.25 | 3.26 | 3.25 |
| | S | 2.41 | 2.31 | 2.78 | 2.71 | 2.72 |
| | Y | 2.53 | 2.28 | 3.00 | 2.86 | 3.11 |
| | | 10% pH 6.92 pH 6.6 | 20% (w/v) pH 6.79 | 30% (w/v) pH 6.71 | 40% (w/v) pH 6.66 | 50% (w/v) pH 6.62 |
| Fish | C | 3.35 | 4.25 | 4.25 | 4.58 | 4.88 |
| | I | 3.45 | 4.09 | 4.14 | 3.86 | 4.16 |
| | S | 3.35 | 3.11 | 3.68 | 3.82 | 4.66 |
| | Y | 2.87 | 2.70 | 2.73 | 2.94 | 3.63 |

**Table 2** CR of Al from WL after immersion for 1 h at 90 °C in different concentration of milk.

| Soln. | Al alloy | Conc. CR × $10^2$ (mg/cm$^2$ h) | | | | |
|---|---|---|---|---|---|---|
| | | 10% pH 7.3 | 20% (w/v) pH 7.2 | 30% (v/v) pH 7.1 | 40% (w/v) pH 6.9 | 50% (w/v) pH 6.8 |
| Fresh milk (F. Milk) | C | 3.16 | 3.03 | 4.08 | 4.04 | 5.08 |
| | I | 2.30 | 3.26 | 3.43 | 3.81 | 5.49 |
| | S | 1.64 | 2.44 | 2.40 | 2.60 | 3.65 |
| | Y | 2.17 | 2.24 | 2.12 | 2.26 | 2.34 |
| Long life milk (L. Milk) | C | 3.72 | 3.55 | 3.62 | 3.58 | 3.86 |
| | I | 3.66 | 3.48 | 3.50 | 3.63 | 3.72 |
| | S | 2.33 | 2.44 | 2.60 | 2.78 | 2.95 |
| | Y | 2.31 | 2.97 | 3.25 | 3.16 | 3.73 |

To compare the present data with the Provisional Tolerance Weekly Intake (PTWI) of 7 mg of Al/kg body weight of adult (World Health Organization, 1989), some assumptions are made. Assuming a family of 4 persons using an Al utensil of medium size with a diameter 20 cm and height 15 cm, the internal area of the Al utensil exposed to leaching will be about 1200 cm$^2$. The concentration of 30% for lamb (alloy C) is taken as an example and Al leaching per hour will be equivalent to:

$$3.25 \times 10^{-2}\,\frac{\text{mg}}{\text{cm}^2\,\text{h}} \times 1200\,\text{cm}^2 = 39.0\,\frac{\text{mg}}{\text{h}}\ \text{Al per family}$$
$$= \frac{39.0\ \text{mg Al/h}}{4\ \text{persons}}$$
$$= 9.75\,\frac{\text{mg}}{\text{h}}\ \text{Al per person}$$

The "estimated" Al intake per person in mg/h from WL in 30% of meat extract and milk is shown in Figs. 1 and 2.

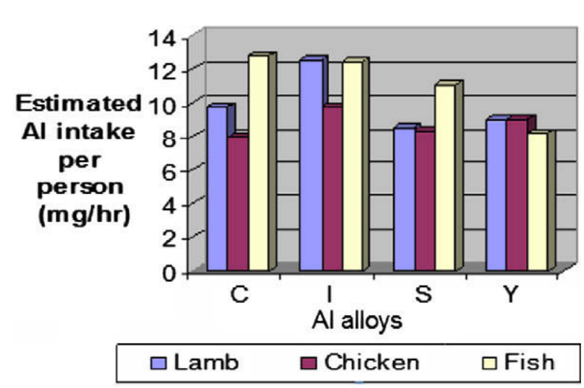


**Figure 1** Estimated Al intake per person from WL in 30% meat extracts solutions in different Al alloys.

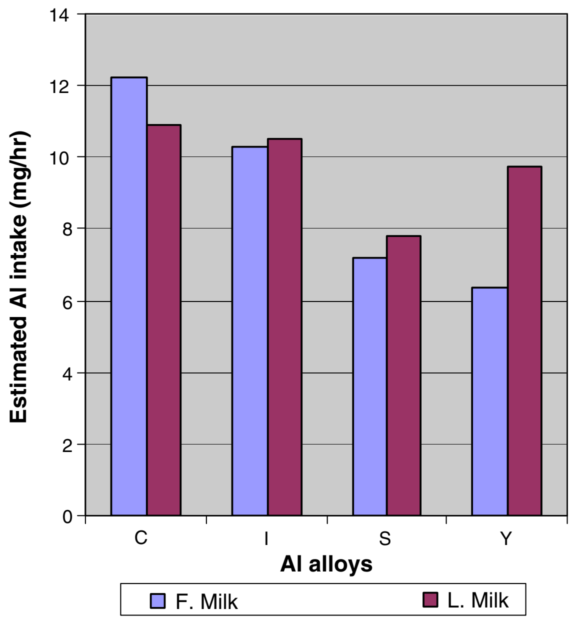


**Figure 2** Estimated Al intake per person from WL in 30% milk solutions in different Al alloys.

From these results the order of Al leaching in 30% solutions appears to be:

fish > lamb > fresh milk ~ long life milk > chicken

In general for 30% concentrations there is a difference in Al leaching among the four different alloys with alloy I having the highest leaching and alloy Y having the lowest leaching. There is a difference in Al leaching depending upon the composition of Al alloy which reflects the effect of alloying elements. It is found from this study that fish extract appears to leach more Al than lamb or chicken. This shows that the composition of food determine the amount of Al leaching which agrees with others (Ščančar et al., 2004).

WL method simulates the amounts of Al leached by food in conditions close to the real ones. Distilled water was used in this study but it is expected that Al leaching from Al utensil using tap water will leach more Al as found from a previous study (Al Juhaiman, 2000). In real life during cooking people may add tomato paste, lemon juice, table salt, and other spices which were shown by many researchers to leach more Al when using Al utensils (Verissimo et al., 2006; Joshi et al., 2003).

No significant differences in pH were found in the present work. This finding agrees with other studies (Fimreite et al., 1997; Neelam and Kaladhar, 2000). The amino acids in meat and lactic acid in milk have high susceptibility to react with $A^{3+}$ to form Aluminum complexes. The dissolution of Aluminum may change the local pH in Aluminum surface but it did not affect the pH of the solution which is measured by the pH meter. The pH range of the present study was almost neutral. In neutral solutions the protective $Al_2O_3$ film has low solubility. The solubility of the protective oxide film will increase as the pH decreases as was found by some researchers (Lenderink et al., 1993). Thus it is expected that Al leaching in meat and dairy products will be increased when the pH becomes low as in real cooking when adding lemon juice or tomato paste. This was also shown in a previous study of corrosion of Al in carboxylic and amino acids where the amount of Al leaching increased twice when the pH changed from 6.5 to 4.5 (Al Juhaiman et al., 2004; Al-Mayouf et al., 2008).

Even boiling water in Al cook wares resulted in leaching some Al into the medium, which was explained by the presence of fluoride (Neelam and Kaladhar, 2000). However this finding was completely opposed by another recent study where they showed that boiling water in Al utensils may reduce Al leaching in cooking by 60% (Karbouj and Nrtier, 2009).

The other controversial study reported that Al leaching was reduced by the presence of amino acids and that food stuff with neutral pH and low salt content like milk do not leach Al (Severus, 1989). Based on the present results there are appreciable amounts of Al leaching from milk ranging from 6.36 to 12.24 mg/h for 30% solutions for the different Al alloys. In addition, there was a significant Al leaching by pure amino acids (aspartic and glutamic acids) which was pH dependant (Al Juhaiman et al., 2004).

The safety of Al depends on whether it is absorbed or not and there are conflicting reports about its bioavailability (Prescott, 1989; Slanina and Falkeborn, 1984; Gitelman, 1989; Winship, 1993). A study of the average Al concentration in daily diet (Soni et al., 2001) showed that under normal circumstances the average dietary intake of Al (without water) is about 6–15 mg/day. In another report the total estimated average daily Al intake in USA was 26.5 mg/day (World Health Organization, WHO/FAO, 1989). The joint FAO/WHO Expert committee on food additives has established the Provisional Tolerance Weekly Intake (PTWI) of 7 mg of Al/kg body weight of adult (WHO/FAO, 1989). Based on the present results there are appreciable amounts of Al leaching from meat extract and milk ranging from 8.16 to 12.75 mg/h for 30% solutions for the different Al alloys. This amount will be increased with increasing concentration as shown in Tables 1 and 2. Using Al cook wares may leach significant amounts of Al into the food, which raise the amounts of Al to high levels and may be dangerous to children, the elderly and people with kidney problems.

### *3.2. Atomic absorption measurements*

Analysis of dissolved $Al^{3+}$ ions in the remaining solutions from WL experiments by AA are calculated in ppm (mg/L). As an example only 30% solutions of Fish and F. Milk were analyzed by AA. To compare the AA results with WL data the results from AA were multiplied by the volume of solution then divided by the area of Al sample and the time of leaching experiment of 1 h. The same earlier assumption was applied to calculate the "estimated" Al intake per person in mg/h from AA. The results of WL and AA are shown in Figs. 3 and 4. A good consistency was observed between the data from WL and AA. Using AA for analysis after WL is an uncommon method in the literature. Yet it was shown in this study and in a previous study that WL and AA gave comparable results (Al Juhaiman et al., 2004; Al-Mayouf et al., 2008).

### *3.3. Electrochemical method*

The purpose for using this method is to detect any small changes in Al leaching. However it does not reflect the natural tendency of Al dissolution as WL. In these experiments the concentrations of meat and milk extracts were kept. All

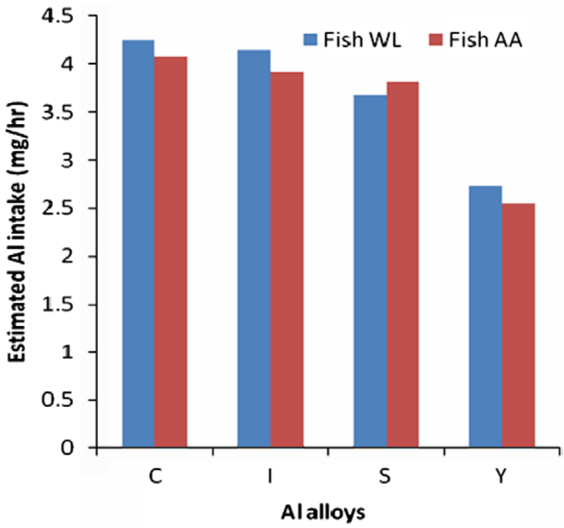


**Figure 3** Comparison of Al leaching from WL and AA in 30% fish extracts in different Al alloys.

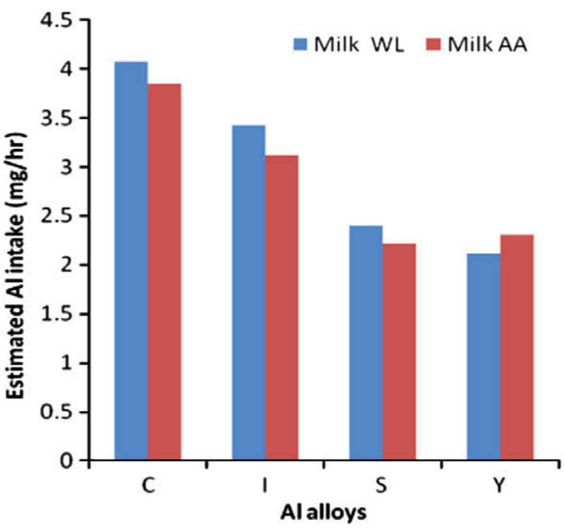


**Figure 4** Comparison of Al leaching from WL and AA in 30% fresh milk in different Al alloys.

experiments were performed in aerated solutions at 60 °C ± 1. As an example only the solutions of 30% fish extract and the fresh milk was used. After running open circuit potential for an hour to attain equilibrium (not shown), Tafel plots were performed. The results are shown in Figs. 5 and 6.

The electrochemical parameters of Al in these solutions are listed in Table 3. It is shown from Table 3 that values of current density ($I_{corr}$) are in general less than 0.50 μA/cm$^2$. These low values may be explained because of the low temperature which is the maximum allowed temperature for electrochemical measurements. Thus it is expected that these current density data will be doubled or tripled when the temperature is increased to 100 °C as in real cooking.

From Tafel results the order of Al leaching of Al alloys in fish and milk solutions appeared to be:

Y > C > I > P > S

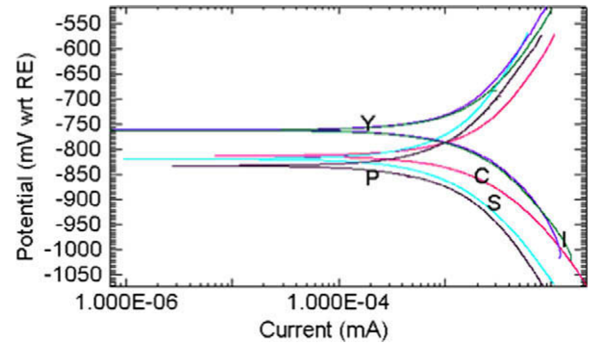


**Figure 5** Tafel plot of 30% fish extract in pure Al and in different Al alloys at 60 °C.

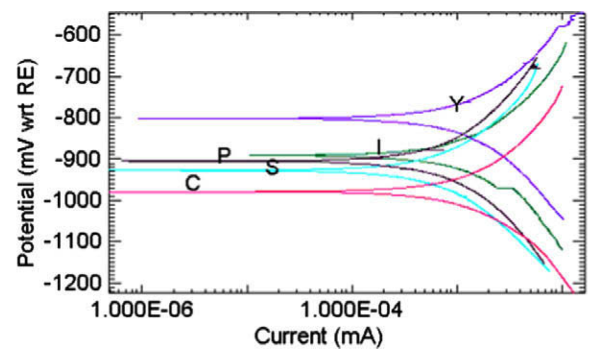


**Figure 6** Tafel plot of 30% fresh milk in pure Al (P) and in different Al alloys at 60 °C.

It is surprising that alloy S leach less than pure Al in fish extract and fresh milk. This clearly reflects the role of alloying and shows that some alloying element may reduce Al leaching.

### *3.4. Surface study*

Using EDX the Al composition was found to range between 97.2% and 98.6% Al with different alloying elements. Alloy S is chosen as an example of Al alloys after leaching experiment for 1 h. Washing the sample with distilled water and magnifying the surface in SEM in 30% fish extract (Fig. 7) showed some holes due to dissolution of Al by fish extract constituents. The same result was shown for 30% milk solution where the white area is partially milk constituent (Fig. 8). These results show that the amino acids in meat and lactic acid in milk have high susceptibility to react with $A^{3+}$ to form Aluminum complexes.

## 4. Conclusion

This method allows us to estimate Al leaching from Al cook wares in meat extracts and milk at any concentration; it is reliable and simulates Al leaching from Al cook wares. Leaching of Al depends on solution composition, concentration and Al composition (alloying elements). In the present study WL and AA gave comparable results. Based on the present results it is estimated that using Al cook wares may leach significant amounts of Al into the food. This raises the amounts of Al

**Table 3** Electrochemical parameters of Al alloys in 30% (w/w) fish extracts (pH 6.4) and 30% (v/w) milk (pH 7.0) at 60 °C.

| 30% Food extract | Al alloy | $-E_{corr}$ (mV/dec.) | $B_a$ (mV/dec.) | $-B_c$ (mV/dec.) | $I_{corr}$ (μA/cm$^2$) |
|---|---|---|---|---|---|
| Fish | Pure Al | 823 | 82.7 | 74.2 | 0.35 |
| | C | 822.4 | 86.7 | 69.5 | 0.41 |
| | I | 771.0 | 82.54 | 69.2 | 0.38 |
| | S | 819.3 | 89.67 | 69.2 | 0.23 |
| | Y | 767.4 | 103.3 | 83.9 | 0.47 |
| Fresh milk | Pure Al | 902.5 | 80.4 | 68.1 | 0.27 |
| | C | 980.2 | 77.5 | 65.6 | 0.28 |
| | I | 891.5 | 96.8 | 90.6 | 0.38 |
| | S | 835.2 | 66.4 | 57.5 | 0.22 |
| | Y | 796.4 | 70.8 | 59.6 | 0.26 |

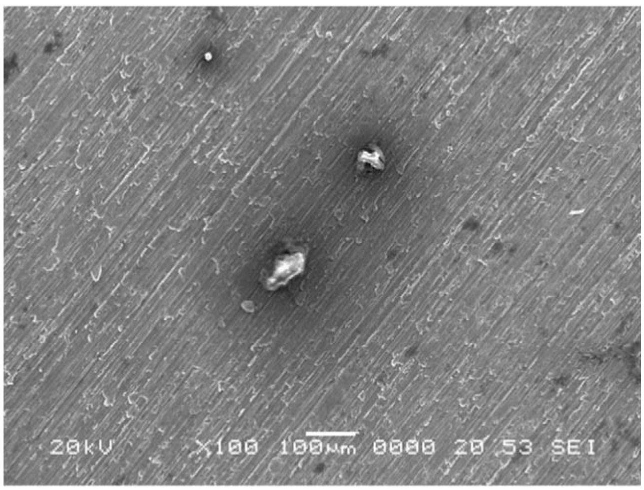


**Figure 7** SEM of alloy S after leaching in 30% fish extract at 90 °C, 1 h.

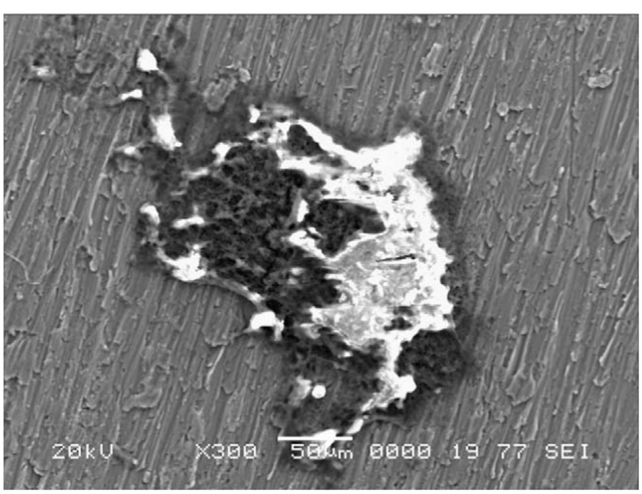


**Figure 8** SEM of alloy S after leaching in 30% fresh milk at 90 °C, 1 h.

to high levels and may exceed the recommended amounts set by WHO/FAO. Therefore, continuous monitoring of Al level in daily diet, water, medicines, and food additives is recommended.

## Conflict of interest statement

The author declares that there are no conflicts of interest.

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