Mobility of aluminum and mineral elements between aluminum foil and bean cake (Moimoi) mediated by pH and salinity during cooking

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Abstract

The dependence of pH and salinity on the leaching of aluminum and allied metals from aluminum foil into bean cake during cooking was studied. Concentration of metals in the foil samples and prepared bean cake were determined using energy dispersive X-ray fluorescence and atomic absorption spectrophotometers respectively. Analysis of aluminum foil brands sold within the study area showed that they all contained aluminum, magnesium, iron, zinc, manganese, chromium and nickel with mean concentrations ranging between 972.45–977.10 g/kg, 16.21–19.84 g/kg, 2280–5710 mg/kg, 1140–1260 mg/kg, 400–410 mg/kg, 80–90 mg/kg and 480–540 mg/kg respectively. Maximum leaching of aluminum into bean cake wrapped in aluminum foil was observed in the saline bean paste followed by alkaline and acidic pastes. The increase in the concentration of iron and zinc in the bean cake samples indicated that these metals coleached with aluminum into the food during cooking. The loss of magnesium, nickel, manganese and chromium from the bean cakes after cooking suggested that demineralization of this food material occurred simultaneously with the leaching of aluminum, iron and zinc into it. The hazard quotient model indicated that all the metals that leached into the bean cake were at concentrations that could pose serious health risks to consumers.

Keywords Aluminum foil · Bean cake · Salinity · Leaching · Demineralization

1 Introduction

Aluminum foil is a thin metal leaf with a thickness less than 0.2 mm, making it easy to bend and wrap around objects. The production of aluminum foil for use in wrapping household products to prevent them from light, moisture and aroma started in Switzerland [1]. It has been used to pack food and drinks for storage and holding food meant for cooking, baking or roasting. Recent studies have shown that aluminum can leach from aluminum foil and aluminum cooking utensils in different media [2–4]. The extent of the leaching is dependent on the acidity of the cooking medium and/or food material, cooking temperature, cooking time, presence of complex species and food composition [5, 6]. The type of aluminum foil used does

not significantly affect the amount of aluminum leached into the food material [2].

The Joint Food and Agriculture Organization/World Health Organization Expert Committee on Food Additives (JECFA) established a provisional tolerable weekly intake (PTWI) for aluminum of 1 mg/kg body weight in 2006. Five years later, the committee re-evaluated the safety of aluminum and proposed a PTWI of 2 mg/kg body weight [7]. These regulations notwithstanding, the use of aluminum foil and aluminum cooking utensils in cooking has increased in recent times. At high concentrations in the human system, aluminum has been reported to reduce the growth rate of human brain cells [8] and has been associated with rise in male infertility by reducing sperm count [9]. Other health problems as a result of aluminum

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toxicity include chronic renal failure [10], neurological disorders [11], Alzheimer's disease [12], skeletal diseases [13], hematopoietic diseases [14], Parkinson disease [15], immunologic health effects [16, 17] and multiple sclerosis [18].

Though current research have put paid to doubts of the possibility of aluminum leaching into food from aluminum foils and aluminum cooking utensils under favourable conditions, changes in the mineral element composition of the food items during the course of leaching and hazard level associated with regular consumption of such food items are yet to be reported.

In this study, the dependence of pH and sodium chloride concentration on the leaching of aluminum from aluminum foil into food vis a vis changes in the mineral element composition of the food material was studied. The health risk associated with the regular consumption of food cooked with aluminum foil was also assessed.

2 Experimental

2.1 Compositional study of locally available aluminum foil brands

The elemental compositions of popular aluminum foil brands within the study area were determined to ascertain the nature and quantities of elements present in them. The samples were collected from open markets and supermarkets in Owerri, Imo State Nigeria. Eight food grade aluminum foil products, three from each company were sampled. The different foil brands were labeled a–h as follows (Table 1):

A roll of each product was collected from a particular sales outlet bimonthly for a period of 6 months. Analyses were carried out on the 24 foil samples collected.

The elemental composition of the aluminum foils were determined using a compact multi-element benchtop Energy Dispersive X-ray Fluorescent Analyzer (EDXRF) EDX3600B by Skyray Instrument (Precision: 0.0% deviation; Detection limit: 0.0001% (1 ppm)–99.99%). The

aluminum foils were pulverized to a fine homogeneous size, pelletized and measurements were made at 40.0 kV and current of 200 μ A. The concentrations in g/kg of the metallic elements in the samples were obtained.

2.2 Food sample collection and preparation

Beans cake (Moimoi or Moin–Moin), a regular Nigerian steamed bean pudding taken as food or snack was used for this study. It is prepared from ground and spiced beans and often wrapped with aluminum foil before cooking. Iron bean seeds (*Phaseolus vulgaris*) of weight 200 g were soaked in deionized water for 5 h. The seed coat was removed by rubbing between the palms and the cotyledon was pounded in a wooden mortar to produce a smooth paste by adding 50 mL of deionized water. The paste was transferred into a plastic bowl and stored in a refrigerator.

2.3 Effect of pH on the leaching of aluminum

5 g bean paste was weighed separately into seven 100 mL beakers and the pH of the pastes in each beaker adjusted to 2, 4, 6, 7, 8, 10 and 12 using HCl and NaOH solutions. Aluminum foil was folded into sachets and the paste from each beaker transferred into a labeled sachet. The same weight of the initial bean paste (at pH 6.2) was wrapped in a polyethylene sachet and used as control. The wrapped pastes were steam-cooked in a non-stick pot for 90 min and allowed to cool under room conditions. Each of the prepared bean cakes was mashed to a homogenous paste in a plastic bowl and 0.5 g of each paste was transferred into a digestion tube. The samples were digested using the nitric acid-perchloric acid method [19]. The concentrations of aluminum and other minerals identified in the analyzed foil samples were determined using Agilent 240FS Fast Sequential Flame atomic absorption spectrophotometer.

2.4 Effect of salinity on the leaching of aluminum

5 g bean paste was weighed separately into four 100 mL beakers and their pH adjusted to 7. The paste in each beaker was thoroughly mixed with 0.2, 0.4, 0.6 and 0.8 g of NaCl respectively. Aluminum foil was folded into sachets and the paste from each beaker transferred into a labeled sachet. The wrapped pastes were steam-cooked in a non-stick pot for 90 min and allowed to cool under room conditions. The samples were digested and the elemental composition determined using an atomic absorption spectrophotometer.

Table 1 Sampled aluminum foil brands

Samples	Brand
a	Akpaka original aluminum foil
b	Omega quality aluminum foil
c	Xtra-thick premium quality aluminum foil
d	Merlin original aluminum foil
e	Queen foil
f	Royal wrap premium quality aluminum foil
g	Rose aluminum foil wrap
h	Golden Papel De Aluminio

2.5 Mineral element mobility studies

X-ray florescence was used to determine the changes in the mineral element composition of the foils used in cooking the bean cake samples in the different study media. The fluorescence spectra of foils used to wrap the bean cakes with the highest observed concentrations of aluminum in the pH and salinity studies were obtained. The spectrum of a fresh foil from the roll served as control.

The experiments in all the studies were performed in triplicate and average values of the results used for the data computation.

2.6 Human health risk assessment

The health risk associated with the daily consumption of the bean cakes cooked with aluminum foil with respect to their mineral element compositions was assessed using the Hazard Quotient (HQ) model [20, 21] and is shown in Eq. (1).

$$HQ = \frac{DIR}{R_f D_o} \quad (1)$$

DIR is the daily intake rate calculated using Eq. (2) [22].

$$DIR = C_{(\text{Metal conc.})} \times C_{(\text{Factor})} \times D_{(\text{Beans cake intake})} \quad (2)$$

The DIR for this study was modified to Eq. (3).

$$DIR = (C_{(\text{Metal conc.})} - C_{(\text{Metal in control})}) \times C_{(\text{Factor})} \times D_{(\text{Beans cake intake})} \quad (3)$$

where $C_{(\text{Metal conc.})}$ = metal concentration in beans cake after cooking with aluminum foil (mg/kg); $C_{(\text{Metal in control})}$ = the metal concentration in control beans cake (mg/kg); $C_{(\text{Factor})}$ = conversion factor (0.088); $D_{(\text{Beans cake intake})}$ = daily intake of beans cake ($0.1 \text{ kg person}^{-1} \text{ day}^{-1}$). The conversion factor of 0.088 was set to convert fresh beans cake

weight to dry weight and was obtained using Eq. (4) [23, 24].

$$IR_{dw} = IR_{ww} \left[\frac{100 - W}{100} \right] \quad (4)$$

where IR_{dw} = dry weight intake rate; IR_{ww} = wet weight intake rate ($0.1 \text{ kg person}^{-1} \text{ day}^{-1}$) and W = percent water content obtained by drying the bean cake to constant weight at 100°C and calculating the percentage of water lost (= 12%).

The $R_f D_o$ is the oral reference doses of the leached metals as given by DEFRA [25] and FAO/WHO [26].

If $HQ \geq 1$, the food is considered not safe for consumption and might have a potential health risk.

3 Results and discussion

The data obtained from the X-ray fluorescence studies showed that the aluminum foil samples did not contain only this element but other metals namely magnesium (Mg), chromium (Cr), manganese (Mn), iron (Fe), nickel (Ni), zinc (Zn), lead (Pb), tin (Sn) and antimony (Sb). The mean concentrations of the elements in the foil samples are shown in Table 2.

All the 24 foils sampled contained magnesium, iron, zinc, manganese, chromium and nickel in the order $Mg > Fe > Zn > Ni > Mn > Cr$. Despite the importance of these metals in living systems, their presence in high concentrations and sometimes their oxidation–reduction properties and chemical coordination enable them to escape control mechanisms. These result in their binding with protein sites not made for them in living systems. This is done by displacing original metals from their natural binding sites which leads to malfunctioning of cells and ultimately toxicity [27].

Table 2 Mean concentrations of metals in the different aluminum foil samples

Metals	Concentrations (g/kg)							
	a	b	c	d	e	f	G	h
Mg	17.33±0.05	16.21±0.14	18.52±0.22	16.91±0.19	19.60±1.24	17.92±0.33	19.84±1.61	18.21±1.50
Al	975.54±1.12	975.81±1.18	973.82±1.02	975.22±3.15	972.45 0.44	974.42±1.01	975.32±0.73	977.10±2.61
Cr	0.08±0.01	0.08±0.02	0.08±0.01	0.08±0.01	0.08±0.01	0.08±0.01	0.08±0.00	0.09±0.03
Mn	0.41±0.02	0.41±0.01	0.40±0.01	0.41±0.01	0.40±0.01	0.41±0.01	0.41±0.02	0.41±0.01
Fe	4.82±1.17	5.71±3.13	5.40±0.74	5.53±2.08	5.38±1.03	5.38±2.01	2.61±0.54	2.28±0.14
Ni	0.51±0.03	0.51±0.01	0.48±0.06	0.52±0.02	0.52±0.01	0.54±0.03	0.51±0.02	0.54±0.01
Zn	1.19±0.04	1.18±0.01	1.19±0.01	1.18±0.02	1.26±0.14	1.16±0.08	1.14±0.13	1.15±0.06
Pb	0.03±0.01	0.00±0.00	0.00±0.00	0.00±0.00	0.00±0.00	0.00±0.00	0.00±0.00	0.00±0.00
Sn	0.00±0.00	0.00±0.00	0.00±0.00	0.00±0.00	0.05±0.01	0.00±0.00	0.00±0.00	0.03±0.01
Sb	0.00±0.00	0.00±0.00	0.00±0.00	0.06±0.02	0.15±0.03	0.00±0.00	0.00±0.00	0.11±0.05

The mean concentrations of Al, Mg, Cr, Mn, Fe, Ni and Zn in the control bean cake and the samples wrapped in aluminum foil at different pH values and NaCl concentrations are shown in Table 3.

The mean concentrations of the mineral elements in the bean cake samples at different pH and NaCl concentrations are compared in Figs. 1 and 2.

The percentage changes in the concentration of the mineral elements in the bean cake samples cooked at different conditions is shown Table 4.

Aluminum concentration in the bean cakes increased from 13.2 to 38.2% at pH 2 to 4, and dropped to 26.2% at pH 6. At neutral pH, no leaching was observed but rather the initial aluminum content of the bean cake decreased by 3.4%. The bean cakes at alkaline pH showed a steady increase in the concentration of leached aluminum from 33.3 to 161.0% between pH 8 and 12 respectively. These results were similar to those by Essam et al. [3] in their study of effect of pH, salinity and temperature on aluminum cookware leaching during food preparation. They observed that the corrosion rate was rapid at low pH, decreased to a minimum at pH 6.4 and then increased sharply between pH 8 and 10. Bi [28] observed that leaching of aluminum from cooking utensils was enhanced dramatically in the ranges of pH < 4 or pH > 8. A similar observation was also reported by Wong et al. [29].

The concentration of aluminum was very high at 0.2 g of NaCl and increased to almost five times this amount at 0.4 g NaCl. Beyond this point, a sharp drop in aluminum concentration that maintained an almost constant value was observed at 0.6 g and 0.8 g of NaCl. These observations were exactly similar to earlier reports [3, 30]. They explained that the conductivity of the medium and solubility of oxygen were at a maximum at the peak point. At higher NaCl concentration, the solubility of oxygen reduced which resulted to the reduction in the leaching of aluminum. The highest concentrations of aluminum in the bean cake samples in this study were at pH 4, pH 12 and 0.4 g of NaCl respectively.

The increase in the concentrations of iron and zinc in the bean cake after cooking suggested that these metals co-leached with aluminum during the cooking process. The concentrations of magnesium, chromium, manganese and nickel in the bean cakes decreased after cooking. This observation was most prominent under saline conditions. This indicated that the leaching of aluminum, iron and zinc into bean cake was followed directly by the loss of essential minerals from the food material.

The X-ray spectra of the foil samples after the cooking of the bean cake at pH 4, pH 12 and 0.4 g NaCl are shown in Fig. 3.

Table 3 Mean concentration of metals in bean cake samples at different cooking conditions

Metals	Metal concentrations in bean cake samples at different study conditions (mg/kg)													
	Control	pH	NaCl concentration (g)											
			2	4	6	7	8	10	12	0.2	0.4	0.6	0.8	
Mg	455 ± 3.12	0.000 ± 0.00	230 ± 4.12	134 ± 3.70	30 ± 1.38	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	
Al	1758 ± 8.43	1990 ± 5.74	2430 ± 3.17	2218 ± 7.31	1699 ± 8.64	2343 ± 9.09	3255 ± 4.28	4589 ± 7.72	4274 ± 5.02	12286 ± 4.46	8382 ± 8.94	9126 ± 7.66		
Cr	26 ± 1.36	0.000 ± 0.00	8 ± 0.47	18 ± 1.22	0.000 ± 0.00	9 ± 0.61	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00		
Mn	78 ± 4.61	0.000 ± 0.00	38 ± 2.08	78 ± 1.25	30 ± 1.59	8 ± 0.39	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00	0.000 ± 0.00		
Fe	1387 ± 6.74	1778 ± 8.20	1496 ± 5.51	1555 ± 3.86	1468 ± 6.66	1430 ± 7.26	1489 ± 4.81	1479 ± 6.08	1424 ± 5.74	1489 ± 4.20	1484 ± 6.47	1493 ± 7.53		
Ni	808 ± 4.59	702 ± 4.44	701 ± 2.81	786 ± 5.93	895 ± 2.21	806 ± 4.38	758 ± 3.62	772 ± 2.79	722 ± 6.04	159 ± 3.16	410 ± 2.95	489 ± 3.07		
Zn	1422 ± 6.85	1425 ± 7.13	1481 ± 4.04	1426 ± 5.68	1334 ± 3.29	1401 ± 5.93	1446 ± 2.57	1466 ± 4.30	1491 ± 3.84	1593 ± 1.22	1970 ± 6.81	1799 ± 1.16		

Fig. 1 Variation in mineral element concentration in bean cake samples at different pH **Fig. 2** Variation in mineral element concentration in bean cake samples at different NaCl concentrations **Table 4** Changes in the concentration of the metals in bean cake after cooking

Metal	Change in concentration (%)										
	pH							NaCl			
	2	4	6	7	8	10	12	0.2	0.4	0.6	0.8
Mg	−100	−49.5	−70.5	−93.4	−100	−100	−100	−100	−100	−100	−100
Al	13.2	38.2	26.2	−3.4	33.3	85.2	161.0	143.1	598.9	376.8	419.1
Cr	−100	−100	−30.8	−100	−65.4	−100	−100	−100	−100	−100	−100
Mn	−100	−105.3	0.0	−61.5	−99.2	−100	−97.4	−100	−100	−85.9	−100
Fe	22.0	7.9	12.1	5.8	3.1	7.4	6.2	2.7	7.4	7.2	7.6
Ni	−13.1	−13.2	−2.7	10.8	−0.2	−6.2	−4.5	−10.6	−80.3	−49.3	−39.5
Zn	0.2	4.5	0.3	−6.2	−1.5	1.7	3.1	4.9	11.9	38.5	26.5

The average concentrations of the metals in the foil samples at optimum aluminum leaching conditions after the preparation of the bean cakes are given in Table 5.

The changes in the concentration of these metals on the foils are given in Table 6.

The results showed that the leaching of aluminum, iron and zinc into bean paste during cooking is accompanied by the deposition of magnesium, chromium, manganese and nickel on the aluminum foil. Aluminum foil during the cooking process behaved like an electrode for the release of aluminum, iron and zinc as well as providing a surface for

the deposition of magnesium, chromium, nickel and manganese, with the bean paste acting as an electrolyte for these reactions. A proposed mechanism of the reduction–oxidation reactions taking place in the bean paste during cooking under acidic, alkaline and saline conditions are illustrated in Fig. 4.

These reactions are summarized as:

Fig. 3 X-ray spectra of foils after cooking **a** Control **b** pH 4 **c** pH 12 **d** 0.4 g NaCl

Table 5 Average concentration of metals in foils after cooking

Element	Concentration in foil after cooking (g/kg)			
	Control	pH 4	pH 12	0.4 g NaCl
Mg	16.487 ± 0.48	16.875 ± 0.31	16.646 ± 0.39	17.256 ± 0.26
Al	975.888 ± 1.73	974.866 ± 0.94	974.114 ± 1.72	973.318 ± 1.30
Cr	0.082 ± 0.04	0.082 ± 0.11	0.083 ± 0.06	0.087 ± 0.17
Mn	0.498 ± 0.22	0.498 ± 0.41	0.498 ± 0.19	0.513 ± 0.53
Fe	5.387 ± 0.49	5.098 ± 0.61	5.188 ± 0.18	4.280 ± 0.87
Ni	0.587 ± 0.25	0.589 ± 0.57	0.587 ± 0.14	0.595 ± 0.29
Zn	1.145 ± 0.36	1.105 ± 0.48	1.118 ± 0.93	1.092 ± 0.74

Table 6 Percentage change in concentration of metals in foils after cooking

Element	pH 4	pH 12	0.4 g NaCl
Mg	2.4	1.0	4.7
Al	−0.1	−0.2	−0.3
Cr	0.0	1.2	6.1
Mn	0.0	0.0	3.0
Fe	−5.4	−3.7	−20.5
Ni	0.3	0.0	1.4
Zn	−3.5	−2.4	−4.6

Fig. 4 Mechanism of redox reactions in bean cake**Table 7** Hazard quotient of leached aluminum in beans cakes

Metal	Hazard Quotient											R _f D _o (mg kg ⁻¹ day ⁻¹)
	pH							NaCl concentration (g)				
	2	4	6	7	8	10	12	0.2	0.4	0.6	0.8	
Al	7.1	20.7	14.2	−1.8	18.0	46.1	87.1	77.4	323.9	203.8	227.8	0.286
Fe	4.9	1.4	2.1	1.0	0.5	1.3	1.2	0.5	1.3	1.2	1.3	0.700
Zn	0.1	1.7	0.1	−2.6	−0.6	0.7	1.3	2.0	5.0	16.1	11.0	0.300

where $M = \text{Fe, Zn}$; $N = \text{Mg, Cr, Mn, Ni}$ and $n = \text{oxidation number of the metal}$. The anodic processes leached aluminum, iron and zinc into the bean paste while the cathodic processes demineralized the bean paste thereby making the bean cake less nutritious.

The toxicity of the bean cakes arising from the amount of metals that leached into them under different cooking conditions was determined using the hazard quotient (HQ) model and the results are shown in Table 7.

The HQ values for aluminum were very high and increased with increase in NaCl concentration to a maximum value of 323.9 at 0.4 g of NaCl. In the alkaline bean paste media, it increased from 18 to 87.1 as pH increased from 10 to 12. The HQ values in the acidic paste media were much lower than the saline and alkaline media showing a maximum value of 20.7 at pH 4. The HQ gave a negative value at pH 7 and implied that only the bean cake cooked under this condition was safe for consumption. Though the HQ values for iron and zinc exceeded the recommended limit, they were much lower than those obtained for aluminum. Iron toxicity would be more likely in the acidic bean cake than alkaline and saline ones. Zinc toxicity was more likely in the saline bean cake and less likely in neutral and slightly alkaline types.

4 Conclusions

Food grade aluminum foils sold in Owerri municipal, Imo State Nigeria contained metal impurities in the order $\text{Mg} > \text{Fe} > \text{Zn} > \text{Ni} > \text{Mn} > \text{Cr}$. The wrapping of acidic, alkaline or saline bean paste in aluminum foil during bean

cake preparation resulted to the leaching of high levels of aluminum, iron and zinc into this food material from the foil. The leaching of these metals into bean cake was complimented by the loss of magnesium, manganese, nickel and chromium from the bean paste. These redox processes did not only demineralize the bean cake but also raised the concentration of the leached metals to toxic levels. Since it is difficult to control the pH and salinity of the bean paste during the preparation of this food material, the use of aluminum foil in wrapping this delicacy before cooking should be totally discouraged and the old time practice of using edible plant leaves for this purpose sustained.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

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