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Release of lead from glaze-ceramicware into foods cooked by open flame and microwave

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Abstract

Samples of ceramicware (24 bowls), purchased from a single manufacturer lot and 36 bowls were collected randomly from a local market in Great Cairo Governorates, Egypt (six locations and six samples from each) and were subjected to different concentrations of acetic acid (2, 4, 6 and 8%) to investigate the release of lead. Results showed that the 4% acetic acid solution was the best concentration for lead extraction from ceramicware followed by 6, 8 and 2%. Release of lead from ceramicware by acetic acid caused a visible erosion of the surface glaze on each bowl. On the other hand, a great variability between the levels of leachable lead and their locations during the extraction time (20 extractions) was also observed. Transfer of leachable lead from ceramicware into foods (salsa and beans) cooked by traditional and microwave methods was also studied. Results revealed that the correlation between the cooking time of salsa by the traditional method and leachable lead was approximately linear. Moreover, the concentration of lead in the final product (salsa) cooked by the traditional method was higher (15.0 ppm) than in that cooked by a microwave method. However, there were no significant differences between the leachable lead levels into beans cooked either by traditional or microwave methods. Slightly visible erosion in the surface glaze of bowls after cooking was also observed. © 2001 Published by Elsevier Science Ltd.

1. Introduction

Since the late eighteenth century, certain ceramic glazes have been recognized as potential sources of dangerous lead levels when used with acidic or alkaline foods (Mellor, 1934). The US Food and Drug Administration (FDA) routinely monitors both domestic and imported ceramicware (FDA, 1987). Historically, lead has been the primary flux in silicate glazes because it imparts many desirable properties (Buildini, 1977; Haber & McCauley, 1983). Improperly formulated or fired lead glazes, however, allow hazardous levels of lead to be released from the surface of the ware (ILZRO, 1971). Lead glazes are more soluble at higher temperatures (Norris & Bennett, 1951). It has been reported that hot-oven leaching of specially glazed wares with 4% acetic acid for 2 h resulted in 1–8 times more lead being leached than at room temperature (Gould & Moss, 1982).

Acute and chronic toxic properties of lead are well known. Lead is an acute poison with a toxicity rating of 3–4 (moderately to very toxic), but intoxication from a single exposure is rare (Gosselin et al., 1984). Of greater concern are the cumulative effects of chronic exposure, especially on children (Gastonis & Needleman, 1992). These effects include damage to the kidney and liver; permanent neurological defects; encephalopathy and, possibly, death (Gosselin et al.). For the general population, diet is the major source of lead (DCA, 1982). It is estimated that a child up to 2-years-old ingests 100 μg of lead per day from food and water and that a child between 2- and 3-years-old ingests 150 μg/day. This latter value is the FDA recommended maximum daily intake for children under the age of two (DCA). The Joint Expert Committee on Food Additives, in 1972, established a permissible chronic intake level of lead for adults of 3.0 mg of lead per week (JECFA, 1972).

The great use of glazed ceramicware may create health problems. The release of lead from ceramicware in contact with food is regulated in Japan (Ministry of Health and Welfare, 1986). The International Organization for Standardization (IOS, 1981) has proposed the same limits for the migration of metal from ceramicware. Recently, the FDA, USA, proposed a regulatory limit for ceramic foodservice pitchers, excluding creamers,

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that would limit the leaching of lead from the glazes to not more than 0.1 μg/ml of the extraction solution (Federal Register, 1989). The FDA is also soliciting comments and information on the need to decrease leachable lead from ceramicware other than pitchers. The proposed limit, 0.1 ppm, is 1/25 to 1/50 of the current legal limit.

Most of the ceramic bowls used in Egyptian homes are made from common clay and fired at high temperatures. The bowls are then covered with wet glazing materials (lead silicate) by hand and refired again at high temperatures. When such bowls have been properly fired at high temperatures, they do not release lead into foods in hazardous concentrations. Thus the aim of this investigation was carried out to shed light on the levels of lead leached from ceramicware collected from different locations in Great Cairo Governorates using different concentrations of acetic acid. Also studied were the leachable levels of lead from ceramicware into high-acid foods, such as tomato paste (salsa), and non-acidic foods (beans) cooked by traditional (open flame) and microwave methods.

2. Materials and methods

2.1. Sample collection

In October and November 1999, samples of 36 bowls were collected randomly from local markets in Great Cairo Governorates, Egypt (six locations and six samples from each). The samples were checked to confirm the presence of leachable lead. In December 1999, an additional 24 bowls were purchased from a manufacturer in Misr El-Kadema, Cairo, Egypt, from a single manufactured lot to study the effect of various concentrations (2, 4, 6 and 8%) of acetic acid on leachable lead. The bowl samples were divided into four groups (six samples of each). Also, an additional 24 bowls were purchased from the same manufacture lot and divided into two groups (12 samples of each) to check the leachability of lead into foods [tomato paste (salsa, as acidic food) and beans (as a non-acidic food)] by open flame and microwave cooking (six bowls for each). The weight, diameter and depth of each bowl were measured. The mean weight was 396.6±47.2 g, the mean diameter was 14.09±0.96 cm, and the mean depth was 6.5±0.88 cm. All bowls were washed gently with a commercial dish-washing detergent and a sponge, rinsed with distilled water and air-dried.

2.2. Experimental design

Experiment [1] constituted 24 extractions every day, four concentrations (2, 4, 6 and 8% acetic acid) and six samples for each concentration, to examine the effect of acetic acid solutions on leachable lead from the bowls during 10 extractions. The same experiment was applied to the glaze material powder (lead silicate). One gram of material was dissolved in 100 ml of each concentration (2, 4, 6 and 8%) of acetic acid and extracted every 24 h, 10 times. The means of six samples of each concentration were recorded.

Experiment [2] constituted 36 extractions daily (six locations and six samples from each) using the US FDA regulatory method for leachable lead from glazed-ceramic bowls, 20 times, which involves filling each ceramic bowl with 4% acetic acid solution, and allowing it to stand for 24 h. The acetic acid solution is decanted, mixed and aliquots taken for atomic absorption analysis. In order to ensure uniformity, 400 ml was chosen as the standard volume for each extraction.

Experiment [3] constituted the effect of cooking tomato paste (salsa) and beans to determine leachable lead from ceramicware. 24 new bowls were divided into two groups (12 bowls each for tomatoes and beans). The bowls for tomatoes or beans were each also divided into two groups, the first group (six bowls) for cooking by the open flame method and the other (six bowls) for microwave cooking. In tomato paste (salsa), 400 ml of tomato juice was used for cooking, in a similar way to the acetic acid extract. For the traditional (open flame) method, the samples (six bowls) were boiled for 45 min. (time of conversion to salsa). However, in the microwave method, the samples were boiled using an energy level of 70% (700 W) for 20 min (conversion to salsa). To test for lead leaching into beans during cooking, 100 g of dried beans were cooked in 400 ml of distilled water by the two methods (open flame and microwave). In the first method (open flame) cooking was by boiling for 1 h. However, in the second method, the samples were boiled using an energy level of 70% (700 W) for 40 min. Salsa and bean samples (2 g) were taken every 5 min, and digested for atomic absorption analysis using 10 ml each of concentrated nitric and sulfuric acids and 5 ml of 70% perchloric acid at 121°C for 45 min, in an autoclave. Every procedure was repeated six times with the two methods and pH value was recorded.

2.3. Lead analysis

Lead was analyzed according to the method of AOAC (1990). Samples were diluted when necessary prior to lead measurement, using a Perkin-Elmer Atomic Absorption Spectrophotometer model 2380 set to 283.3 nm wavelength. The limit of detection of the method was 0.05 ppm. The mean relative standard deviations of all aliquots for the acetic acid, tomato paste (salsa) and bean extraction analysis were 2.8, 4.9 and 5.3%, respectively.

3. Results and discussion

3.1. Lead levels released from ceramicware using different concentrations of acetic acid solutions

The method of AOAC (1990) and the equivalent method of the American Society for Testing and Materials (ASTM Standards, 1987) are usually used for testing ceramicware for leachable (extractable) lead. Both methods require 24 h for leaching with 4% acetic acid at room temperature. This investigation determined the leachable levels of lead from 24 bowls of ceramicware purchased from a single manufacturer lot in Masr El Kadima, Cairo using different concentrations of acetic acid (2, 4, 6 and 8%) every 24 h, 10 times, to identify the efficiency of these concentrations in extracting lead at room temperature. Table 1 indicates that the extraction levels of lead were high after 24 h (first day) using different concentrations of acetic acid, then the extraction rate was decreased by increasing the extraction time up to 10 times. However, the highest extraction rate of lead was observed on using 4% acetic acid solution, followed by 6, 8 and 2%, respectively, during the extraction time at room temperature as shown in Table 1.

The previous concentrations of acetic acid (2, 4, 6 and 8%) were also used to test the levels of lead in silicate (lead silicate) glaze material (Table 2). Results confirmed that 4% acetic acid solution was the best concentration for extraction of lead from silicate glaze material. The differences in extractability of lead from ceramicware or silicate glaze material using the different concentration of acetic acid may be due to the differences of pH values of different concentrations of acetic acid. However, Sheets and Turpen (1997) reported that leached lead increased as acid strength increased. Acetic acid (4%) was the most effective concentration for extraction of lead from ceramicware or silicate glazes material. Moreover, the acetic acid extractions caused visible erosion of the surface glaze from each bowl.

3.2. Monitoring of lead in different glazed-ceramicware

Changes in the lead concentration released from lead-glazed ceramicware collected randomly from local markets in Great Cairo Governorates were studied using 4% acetic acid solution at room temperature (25–27°C), 20 times. Table 3, and Fig. 1 reveal that, during the extraction time (20 extractions), the total amounts of leached lead ranged from 149 to 3156 ppm. This means that a great variability was found between the levels of lead in ceramicware and their locations. Variability may be due to the glaze applied by hand and it may be fired in wood ovens where the temperature could vary considerably. These factors may account for some of the variability in leaching (Mejia & Craigmill, 1996). The exponential decrease of release indicates that the migration of lead occurs continuously, while the article is used and, hence, intake of metal may be cumulative. For comparison, the Joint International Expert Committee on Food Additives has recommended a maximum permissible chronic intake level of lead for adults of 3.0 mg lead per week (JECFA, 1972). These findings are in agreement with previous investigation of Mejia and Craigmill (1996), who showed that the total amount of lead leached from each vessel during the 20 extractions ranged from 365 to 2446 mg, with a mean of 1155 mg. Also, Romieu et al. (1994) indicated that lead levels of up to 3730 ppm are released by lead-glazed ceramic bowls. On the other hand, Sheets, Turpen and Hill (1996) reported that, total amount of lead released under standard conditions reached up to 61 000 (μg/bowl) in ceramic dinnerware. The US FDA indicated that, of 5222 imported lots of ceramicware examined, 46 exceeded FDA, 1991, guidelines, and, 676 lots of

Table 1
Lead concentrations released from glazed ceramic bowls (single manufactured lot) extracted by acetic acid solutions[a]

Extraction no. (time/day)	Mean lead concentration (ppm±S.D.) in acetic acid solutions			
	2%	4%	6%	8%
1	15.5±4.2	19.7±7.8	17.1±4.8	16.1±3.7
2	7.8±3.2	15.2±2.2	14.8±2.4	10.9±4.3
3	13.0±3.0	16.5±3.4	15.9±2.5	14.8±3.2
4	1.1±0.6	3.2±1.7	1.8±1.0	1.4±1.0
5	2.1±1.2	2.7±1.2	2.5±1.3	2.3±1.1
6	1.4±1.0	2.1±1.1	3.1±1.4	2.9±1.2
7	1.1±0.8	3.1±1.2	1.9±1.0	1.6±1.0
8	2.4±1.1	2.8±1.1	2.4±1.1	2.6±1.1
9	1.8±1.0	1.9±1.0	1.6±1.0	1.2±1.0
10	1.6±1.0	2.1±1.0	1.8±1.0	1.4±1.0
Total	47.8	69.3	62.9	55.2

[a] Mean, six samples for each concentration.

Table 2
Lead concentrations released from glazed material (lead silicate) extracted by acetic acid solutions[a]

Extraction no. (time/day)	Mean lead concentration (ppm±S.D.) in acetic acid solutions			
	2%	4%	6%	8%
1	592±20.2	850±17.9	753±36.1	730±26.9
2	133±7.7	179±8.5	108±6.3	103±6.1
3	28.1±3.2	51.0±4.6	21.0±3.1	28.5±3.4
4	29.4±3.6	13.2±3.0	20.6±3.0	26.5±3.6
5	12.6±3.1	15.8±2.9	8.6±2.8	3.9±2.1
6	9.1±2.5	6.1±2.1	4.5±2.4	3.9±1.8
7	7.3±2.8	5.1±2.3	4.5±2.6	6.2±2.4
8	8.0±3.1	3.1±1.8	5.5±2.4	3.6±1.8
9	2.9±1.1	3.1±1.4	3.3±1.6	4.2±2.0
10	2.6±1.0	2.2±1.2	3.4±1.5	1.8±1.0
Total	825	1129	932	912

[a] Mean, six samples for each concentration.

Table 3
Lead concentrations released from glazed ceramic bowls collected from great Cairo governorates (from six different locations)[a]

Extraction no. (time/day)	Mean lead concentration (ppm±S.D.) in 4% acetic acid					
	Location 1	Location 2	Location 3	Location 4	Location 5	Location 6
1	17.4±4.5	55.1±13.4	508±113.9	19.7±4.2	130±32.5	103±25.8
2	16.0±3.1	42.2±12.1	419±106.6	17.1±4.3	104±25.8	72.9±18.6
3	17.1±2.3	43.9±13.1	391±89.2	16.5±3.8	66.5±16.6	74.7±18.5
4	14.8±3.5	34.5±12.5	255±59.9	16.8±3.2	40.2±11.8	70.7±17.9
5	15.0±3.2	35.2±11.7	213±31.5	15.1±3.8	36.1±9.1	54.8±13.5
6	14.3±2.1	27.5±11.7	241±46.6	15.4±3.3	28.2±7.3	52.1±13.1
7	14.7±2.7	23.5±12.5	184±38.8	16.0±3.7	24.8±6.6	19.3±4.8
8	15.2±3.6	26.3±12.9	160±30.3	14.7±3.2	20.5±5.3	15.5±3.6
9	15.5±3.9	14.1±6.6	179±31.3	13.6±3.6	16.0±5.1	12.8±3.8
10	3.1±1.5	8.6±3.9	153±31.2	15.4±3.5	18.1±4.8	9.7±2.6
11	2.1±1.1	11.4±3.1	129±32.3	3.8±1.1	10.2±3.1	4.8±2.1
12	0.9±0.4	3.5±1.2	110±27.5	1.1±0.3	5.2±1.6	1.5±0.5
13	0.5±0.2	0.5±0.2	84.0±21.0	0.3±0.1	2.3±0.8	0.4±0.1
14	0.4±0.2	0.4±0.2	60.0±15.0	0.3±0.1	0.9±0.3	0.3±0.1
15	0.3±0.1	0.4±0.2	41.0±10.3	0.2±0.1	0.4±0.2	0.3±0.1
16	0.4±0.2	0.4±0.2	15.0±3.8	0.4±0.2	0.4±0.2	0.2±0.1
17	0.2±0.1	0.3±0.1	9.5±2.4	0.2±0.1	0.3±0.2	0.3±0.1
18	0.2±0.1	0.4±0.2	3.6±0.9	0.2±0.1	0.3±0.2	0.4±0.1
19	0.3±0.1	0.4±0.1	0.6±0.3	0.3±0.1	0.4±0.2	0.3±0.1
20	0.2±0.1	0.3±0.1	0.6±0.2	0.2±0.1	0.4±0.1	0.2±0.1
Total	149	329	3156	167	505	494

[a] Mean, six samples for each location.

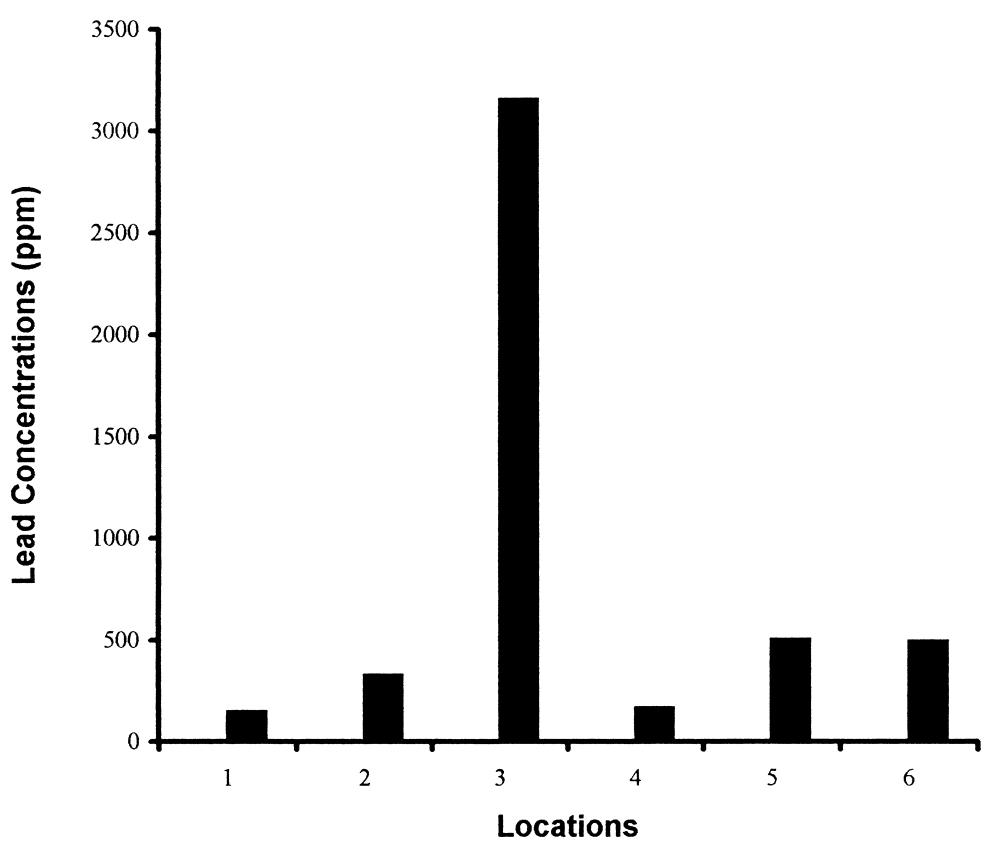

Fig. 1. Sum of lead concentrations released from glazed-ceramicware collected from six different locations and extracted by 4% acetic acid 20 times.

domestic ware examined; 17 lots exceeded (Boczynskyj & Yess, 1995). However, Mohamed, Chin and Pok (1995) showed that 54.7% of ceramic utensil items tested exceeded the US FDA maximum permitted lead release values.

3.3. Transfer of lead from glazed ceramicware to foods

Tomato fruits and beans are considered to be important crops used in different types of foods. The effects of temperature and time of cooking on the transfer of lead from glazed ceramicware to tomato paste (salsa) (as acidic food) and beans (a neutral food) cooked by traditional (open flame) and microwave methods were studied. Table 4 shows that a gradual increase in lead leached from ceramicware was noticed in tomato paste during cooking, either by traditional or microwave methods. Also, the extraction rate of lead was higher in the samples of tomato cooked by the traditional (open flame) method than by the microwave method. This may be due to the longer time of cooking by the traditional method (45 min) to convert tomato juice into salsa compared than by the microwave method (20 min). Moreover, a correlation between the time of cooking by traditional method and leachable lead was approximately linear. On the other hand, the concentration of lead in the final product (salsa) cooked by the traditional method was higher (15.0 ppm) than that cooked by the microwave method (6.92 ppm).

The use of the microwave for cooking, reduced the cooking time of beans (40 min) compared with the traditional method (60 min) as shown in Table 4. However, no significant differences were observed in the levels of leachable lead between the cooking methods (traditional or microwave). Generally, the leachable lead from ceramicware into tomato paste (salsa) was higher than the leachable lead into beans. This may be due to the higher acidity of salsa than beans; i.e., the higher the acidity values, the higher was the leachable lead. Moreover, a slightly visible erosion in the surface glaze of bowls was observed.

These findings are in accordance with a previous investigation of Mejia and Craigmill (1996). The authors reported that leaching of lead into salsa (pH = 4.8) was variable and ranged from 8 to more than 500 ppm. They further reported that cooking beans with water in the bowls did not cause substantial leaching. On the other hand, Mohamed et al. (1995) reported that acidic foods caused lead to be leached out from the ceramicware. Also, Romieu et al. (1994) reported that foods and beverages prepared and stored in traditional lead-glazed ceramicware can become contaminated with lead; lead levels of up to 3730 ppm are released by lead-glazed ceramicware. They recorded, also, that acidic substances stored in lead-glazed ceramicware are more likely to absorb lead, since the solubility of lead increases as pH decreases.

In a recent study on factors affecting blood lead levels in children in Los Angeles County, Gellert (1993) reported the lead levels in beans cooked in three lead-glazed ceramicware dishes used in food preparation for children with elevated blood lead levels. The mean lead level, they found was 1.18 ppm (0.03 to 2.2 ppm), which they characterized as ‘‘far exceeding biologically acceptable levels’’ which is slightly lower than what was found in this study; however, their pots had been used for some time for food preparation.

The widespread domestic use of microwave ovens increases the probability of lead ingestion from glazed ceramicware. The results obtained in this investigation

Table 4
Lead concentration (ppm) in tomatoes and beans cooked in lead-glazed ceramicware by traditional[a] and microwave methods[b]

Time min	Tomatoes				Beans			
	Traditional method		Microwave method		Traditional method		Microwave method	
	pH	Mean conc.±S.D.	pH	Mean conc. ±S.D.	pH	Mean conc. ±S.D.	pH	Mean conc. ±S.D.
0.0	5.11	0.38±0.13	5.11	0.38±0.13	6.11	0.09±0.03	6.11	0.09±0.03
5.0	5.10	0.98±0.33	5.00	0.44±0.15	6.11	1.23±0.46	6.11	2.05±0.68
10.0	5.01	2.46±0.82	4.92	2.90±0.97	6.11	1.62±0.55	6.21	2.10±0.80
15.0	5.00	5.95±1.98	4.90	3.24±1.08	6.22	2.37±0.81	6.30	2.55±0.94
20.0	4.90	7.19±2.40	4.56	6.92±2.36	6.22	2.43±0.84	6.32	3.00±1.20
25.0	4.82	8.39±2.80			6.23	2.49±0.84	6.42	3.04±1.40
30.0	4.81	9.68±3.23			6.31	2.56±0.88	6.48	3.21±1.07
35.0	4.61	11.46±3.82			6.31	2.72±0.94	6.64	3.26±1.60
40.0	4.56	13.24±4.41			6.42	2.88±0.96	6.88	3.30±1.10
45.0	4.54	15.02±5.60			6.46	3.03±1.14		
50.0					6.47	3.25±1.10		
55.0					6.66	3.47±1.16		
60.0					6.92	3.70±1.23		

[a] Cooking over an open flame.
[b] Mean, six samples for each method.

coincide with those reported by Sheets et al. (1996) who showed that eating a single microwaved meal could result in the ingestion of amounts of lead approaching or exceeding the 3.0-mg permissible weekly limit for adults.

It is concluded that regular use of such glazed ceramicware for food preparation could present a health hazard. Glazed ceramicware may be a potential source of toxic lead levels when used with acidic or alkaline foods. Results suggest that the use of such bowls for microwave heating of common foods could result in less lead than from the open flame method; this may be due to the short time of cooking.

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