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Effect of sucrose and fructose on aluminium residues and quality of heat-treated milk

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Abstract
Aluminium is one of the toxic metal ions. Its utensils are used for transport, storage and boiling of milk. The aim of this work was conducted to evaluate the aluminium residues in raw and heat-treated milk in aluminium utensils. Then the work straight forward to solve this by adding sucrose (0.5% and 1%) and fructose (0.5% and 1%) as corrosive inhibitor and natural preservative to the milk samples during boiling and storage for 72 hours in refrigerator (4°C). Heat treated milk samples (at 100°C for 2 minute) and heated milk with sucrose (0.5% and 1%) & fructose (0.5% and 1%) were analysed for aluminium levels, protein%, fat% and calcium, chloride and sodium concentration at 0, 24, 48 and 72 hours of storage. Raw milk was analysed for the same parameters and revealed that, aluminium level was (8.76±0.66 ppm), protein% (3.49±0.03%), fat% (6.90±0.06%), calcium concentration (40.85±0.99mg/dl), chloride concentration (61.33±0.86mg/dl) and sodium concentration (52.67±0.78mg/dl). Analysed heated milk and sucrose & fructose treated one showed that leaching of aluminium into the milk increased by its boiling and storage in aluminium utensils. Meanwhile, its level was decreased significantly in the treated groups with sucrose (0.5% and 1%) and fructose (0.5% and 1%). Also, fat%, chloride and sodium content of heat-treated milk were changed significantly comparing with the treated one. The result clear that fructose followed by sucrose can be used to prevent aluminium leaching to milk and maintain its quality with increasing its shelf life.

Keywords: *Milk, aluminium migration, sucrose, fructose*

Introduction

Milk and dairy foods are healthy foods and considered a good source of calcium and vitamin D as well as protein and other essential nutrients. They provide phosphorus, potassium, magnesium, and vitamins A, B12, and riboflavin so milk should be free from contamination and residues.

Aluminium comprises 8.13% of the earth's crust, considering the third most abundant element in our environment. It is present in soil, rocks and minerals and even in water and food, it has no essential biological function to the human body. At last decades, aluminium was not considered to have any adverse impact on human health. But days, it generally use in water treatment, in manufacturing of food containers and cooking utensils, in medicines and cosmetics which increase its human risk **(Soni *et al.,* 2001 and Marta *et al.,* 2006).**

Aluminium comes in the milk and milk products from many sources. Milk gets contaminated before milking, from the feed and fodder fed to the dairy cows. Besides, it can be introduced into the milk and milk products during the production process or by contamination from tools, containers and equipment made of aluminium **(Soni *et al.,* 2001 and Deeb & Gomaa, 2011).** The use of aluminium utensils for processing and storage of milk can increase the level of this metal in milk and its products**.** Leaching of aluminium from cookware is an important source, increase aluminium residues in food is dependent on several factors such as pH of the food and/or cooking medium, duration of contact or heating, temperature, presence of sugar, salts, organic acids and other ions **(Ranau *et al.,* 2001).** Humans can be exposed to aluminium through water, foods, airborne dust and pharmaceuticals **(Semwal *et al.,* 2006).**

On the other hand, a high level of aluminium has been detected in the brain tissues of Alzheimer's patients. Various studies suggested that high aluminium intake can be harmful to some patients with bone diseases or renal impairments **(Gitelman, 1989)**. Also, a decrease of Growth rate at higher aluminium concentration becomes more obvious **(Kim, 2001).** In addition to**,** Aluminium toxicity is well known in patients with long standing chronic renal failure **(Meiri *et al.,* 1993)**, aluminium has also been associated with several skeletal osteomalacia and neurological failures **(Gupta *et al.* ,2005).** Regarding the suggested provisional tolerable daily intake of 1mg Al/Kg body weight per day of the FAO/WHO Expert Committee on food additives **(FAO/WHO,1994).**

Leaching of aluminium in the preparation of tomato sauce could be decreased by sugar, resulted in decrease aluminium intake (**Marta *et al.,* 2006)**. Recent studies revealed that metals corrosion inhibited using natural products obtained from plant or animal and considered them as a green corrosion inhibitor (**Devarayan *et al.,* 2012; Huang *et al.,* 2014 and Al-Mazaideh *et al.,* 2016).**

Sucrose is a common table sugar. It is a disaccharide, a molecule composed of the two monosaccharides, glucose and fructose. Sucrose is produced naturally in plants, from which table sugar is refined. As sucrose is biologically safe and is highly soluble in water, it might be used as corrosion inhibitor in the pipelines and water tanks **(Ali-Shattle *et al.,* 2009).** on the other hand, fructose, or fruit sugar is a simple ketonic monosaccharide found in honey, tree and vine fruits, flowers, berries, and most root vegetables **(Wolfgang, 2004).** Commercially, fructose is derived from sugar cane, sugar beets, and maize, fructose-sweetened food and beverage products cause less of a rise in blood glucose levels than do those manufactured with sucrose or glucose **(EFSA, 2011).** Fructose is often recommended for diabetics because it does not trigger the production of insulin by pancreatic β cells, probably because β cells have low levels of GLUT5 (facilitated-diffusion glucose transporters), moreover it considered 73% sweeter than sucrose at room temperature **(Sato *et al.,* 1996).** For a 50 gram reference amount, fructose has a glycemic index of 23, compared with 100 for glucose and 60 for sucrose **(Glycemic index 2017).**

Moreover, sugar is a good natural food preservative **(Seetaramaiah *et al.,* 2011),** which increase the shelf life of food and maintain its quality for longer time **(Sharma, 2015).** Sugar is capable of preserving food and extending storage time through reducing water activity and pathogenic microbial cells **(Duman *et al.,* 2007).**

This study was conducted to detect the levels of aluminium content in heated and stored milk in aluminium pan and trial to reduce the migration of aluminium to milk by using of sucrose and fructose and study their effect on its quality.

Materials and Methods

Sample collection

A total of 25 kg of freshly raw buffalo milk were collected randomly from dairy shops at Cairo - Egypt. Samples were transferred under strict hygienic measures to laboratory as soon as possible where they were divided into 5 parts according to the experimental design as follows: -

1st part raw buffalo milk was heated at 100ºC for 2 minutes in aluminium pan without additive.

2nd and 3rd parts milk was heated at 100ºC for 2 minutes in aluminium pan with addition of sucrose 0.5% and 1%.

4th and 5th parts milk was heated at 100ºC for 2 minutes in aluminium pan with addition of fructose 0.5% and 1%.

The experiment repeated for 5 trails.

All milk samples (heat treated and heated with sucrose & fructose) were subjected to the following analysis at 0, 24, 48, 72 hours of the experiment while the samples were stored in refrigerator.

Sucrose and fructose were obtained from Sigma-Aldrich with CAS Number 57-50-1 and CAS Number 57-48-7 respectively.

1-Aluminium determination

Each part was homogenized then analysed for aluminium content by wet oxidation method, according to **AOAC (1990)** by using a Perkins Elmer 2380 Atomic Absorption Spectrophotometer at wave length 309, temp. 2900-3000^{0}C with nitrous oxide and acetylene.

2-Biochemical examination: -

2-1-Preparation of whey milk: -

Milk serum (whey) was prepared by centrifugation of milk at 3000 rpm for 10 min. to remove cream and cells. Then, five ml of rennin was dissolved in 270 ml of normal saline and one ml. of this solution was added to 10 ml of defatted milk, after 30 min. of incubation at 37ºC the milk was centrifuged at 10000 rpm for 20 min. and then the supernatant (whey milk) was separated **(Frost and Tina, 1988).** Milk serum was used for chloride and sodium determination (**Schoenfeld and Lewellen, 1964)** and calcium **(Kramer and Tisdall, 1982)**.

2-2-Determination of protein and fat % in milk samples: -

Analysis of milk samples for determination of its fat, and protein was performed according to the techniques recommended by **(FSSAI, 2015).**

Raw milk samples before heating were analyzed for the same parameters.

3-Statistical Analysis

The data obtained from four groups were analysed by one –way ANOVA using the SPSS statistical package program, and difference among the individual means were compared using LSD range test

Result and Discussion

Table (1). Raw milk sample analysis data

Aluminium (ppm)	Protein%	Fat %	Calcium (mg/dl)	Chloride (mg/dl)	Sodium (mg/dl)
8.76 ± 0.66	3.49 ± 0.03	6.9 ± 0.06	40.85±0.99	61.33±0.86	52.67±0.78

Values are expressed as mean ± SD

Table (2). Levels of aluminium residue in heat treated and heated with additives milk samples (expressed as ppm):

Sample	Heat treated milk	Heated milk with sucrose 0.5%	Heated milk with sucrose 1%	Heated milk with fructose 0.5%	Heated milk with fructose 1%
0 hour	19.2^{A} ±0.86	17^{AB} ±0.35	11.17^{abC} ±0.6	12.9^{abCD} ±0.7	11.3^{abCD} ±0.38
24 hours	20.8^{A} ±0.9	17.9^{aB} ±0.57	11.83^{abC} ±0.16	13.17^{abCD} ±0.9	11.7^{abCD} ±0.26
48 hours	23.24^{A} ±0.92	18.13^{aB} ±0.94	12.05^{abC} ±0.98	13.48^{abCD} ±0.82	11.94^{abCD} ±0.58
72 hours	23.44^{A} ±0.83	18.87^{aB} ±0.13	12.08^{abC} ±0.84	13.63^{abCD} ±0.91	11.85^{abCD} ±0.75

Values are expressed as mean ± SD; there is a significant difference between means have the same capital and small letter in the same row (P< 0.05)

Table (3). Prevalence of protein% and fat% in heat treated and heated with additives milk samples:

Sample		Heat treated milk	Heated milk with sucrose 0.5%	Heated milk with sucrose 1%	Heated milk with fructose 0.5%	Heated milk with fructose 1%
Protein %	0 hour	3.5 ±0.1	3.57 ±0.06	3.59 ±0.05	3.54 ±0.05	3.58 ±0.05
	24 hours	3.55 ±0.05	3.64 ±0.08	3.66 ±0.02	3.57 ±0.02	3.58 ±0.04
	48 hours	3.6 ±0.05	3.65 ±0.05	3.64 ±0.05	3.63 ±0.04	3.57 ±0.04
	72 hours	3.57 ±0.04	3.63 ±0.04	3.58 ±0.04	3.55 ±0.05	3.60 ±0.05
Fat %	0 hour	6.95 ±0.05	6.93 ±0.04	6.87 ±0.07	6.85 ±0.02	6.90 ±0.08
	24 hours	6.46 ±0.04	6.45 ±0.13	6.57 ±0.07	6.45 ±0.07	6.46 ±0.08
	48 hours	6.52 ±0.04	6.42 ±0.12	6.4 ±0.08	6.44 ±0.05	6.43 ±0.06
	72 hours	6.15^{A} ±0.08	6.15^{AB} ±0.04	6.23^{abC} ±0.04	6.46^{abcD} ±0.04	6.42^{abcD} ±0.02

Values are expressed as mean ± SD; there is a significant difference between means have the same capital and small letter in the same row ($P < 0.05$).

Table (4). Calcium, chloride and sodium levels of heat treated and heated with additives milk samples (expressed as mg/dl):

Sample		Heat treated milk	Heated milk with sucrose 0.5%	Heated milk with sucrose 1%	Heated milk with fructose 0.5%	Heated milk with fructose 1%
Calcium	0 hour	38.86±1.2	39.76±0.98	39.5±0.96	39.55±0.9	39.02±0.6
	24 hours	40.3±1.01	39.73±0.67	39.83±0.62	38.6±0.7	39.97±1.02
	48 hours	40.47±0.8	39.6±0.9	39.6±0.8	40.7±0.89	39.7±0.96
	72 hours	39.5±0.75	38.85±0.29	39.27±0.49	39.31±0.59	39.8±0.49
Chloride	0 hour	55±1.15	55.33±1.2	58±0.88	56.67±1.2	58.67±0.98
	24 hours	52.33^{A}±0.97	54.67^{AB} ±1.65	58.67^{abC}±0.98	56.00^{ABCD} ±1.52	57.33^{aBCD} ±1.45
	48 hours	48^{A}±1.53	53.33^{aB} ±0.88	58^{abC}±1.15	56^{aBCD}±0.98	56.67^{aBCD} ±1.45
	72 hours	45^{A}±0.25	54^{aB}±0.14	57^{aBC} ±0.26	57.67^{aBCD} ±0.88	58^{aBCD} ±0.88
Sodium	0 hour	41.67±0.05	43.67±0.88	45±1.15	44±1.15	47±0.88
	24 hours	40.33^{A} ±0.88	42.67^{aB} ±1.2	45^{aBC} ±1.15	44.33^{aBCD} ±1.20	46.67^{abCD} ±0.88
	48 hours	36^{A}±1.15	39^{aB}±0.98	42^{abC}±1.15	40^{aBCD} ±0.88	44^{abCd} ±0.88
	72 hours	34.67^{A} ±0.76	39^{aB} ±0.88	42^{aBC} ±1.15	40^{aBCD} ±1.15	43.67^{abCD} ±0.98

Values are expressed as mean ± SD; there is a significant difference between means have the same capital and small letter in the same row ($P < 0.05$).

Data analysis of examined raw milk samples were recorded in table (1) showed that the mean value of aluminium was 8.76±0.66ppm, protein % was 3.49±0.03, fat% was 6.9±0.06, mean value of Calcium was 40.85±0.99 mg/dl, mean value of Chloride was 61.33±0.86 mg/dl and mean value of Sodium was 52.67±0.78 mg/dl.

Concerning the aluminium content in the examined samples, the present data in table (2) showed the mean values of aluminium content of heat treated milk in aluminium pan without additive, milk was heated in aluminium pan with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and 1% at 0 hours were 19.2±0.86, 17±0.35, 11.17±0.6, 12.9±0.7 and 11.3±0.38 ppm, respectively. At 24 hours the mean values were 20.8±0.9, 17.9±0.57, 11.83±0.16, 13.17±0.9 and 11.7±0.26 ppm, respectively. At 48 hours they were 23.24 ± 0.92, 18.13±0.94, 12.05±0.98, 13.48±0.82 and 11.94±0.58ppm, respectively. And at 72 hours were 23.44±0.83, 18.87±0.13, 12.08±0.84, 13.63±0.91, and 11.85±0.75ppm, respectively. There was a significant difference ($p<0.05$) of aluminium levels of heat-treated milk and heated with sucrose & fructose milk samples through the experimental time. Aluminium contamination of raw milk was recorded by **El-Mossalami and Noseir (2009), Al-Ashmawy (2011) and Meshref *et al.,* (2015)**. Market milk is the biggest contribution to the intake of aluminium. Its estimated total intake via consumption of milk and milk products was 246.72mg /week which represent 205.5% of the Provisional Tolerable Weekly Intake which may impact to humanity (**Meshref *et al.,* 2015).**

Moreover, increasing of aluminium content of milk samples after heat treatment was reported by **El-Mossalami and Noseir, (2009),** As a result of its migration from aluminium pans**.** Other studies reported increasing of aluminium content of red and white meats after cooking in aluminium foil **(Turhan, 2006)** as well as, aluminium migration from aluminium foil to chicken parts during cooking **(EL-Zeini and Hosny, 2003).** Presented data reveals that the treated samples with sucrose (0.5% and 1%) and fructose (0.5% and 1%) inhibit the aluminium migration to the milk during heating and storage. Nearly the same result was reported by (**Marta *et al.,* 2006)** who stated that white sugar could decrease the leaching of aluminium to the tomato sauce and consequently lower its intake through food, it forms a coating sort that reduce the contact between aluminium surface and the acids in food. The presence of sugar can decrease aluminium leaching in food, the sugar has no acid or alkaline neutralization properties **(Fimreite *et al.,* 1997)**. Also, (**Khalil *et al.,* 2016**) used Density functional theory calculation and reported that fructose followed by sucrose was a good aluminium corrosive inhibitor and discussed that by their ability to accept electrons from aluminium. They added that fructose exhibits higher electrophilicity value than sucrose, also the reactivity of inhibitor fructose towards the metallic surface adsorption is more than that of sucrose.

Table (3) showed the mean values of protein % of heat treated milk in aluminium pan without additive, milk was heated in aluminium pan with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and 1% at 0 hours were 3.5±0.1, 3.57±0.06, 3.59±0.05, 3.54±0.05 and 3.58±0.05%, respectively. At 24 hours such values were 3.55 ±0.05, 3.64±0.08, 3.66±0.02, 3.57±0.02 and 3.58±0.04%, respectively. At 48 hours they were 3.6±0.05, 3.65±0.05, 3.64±0.05, 3.63±0.04 and 3.57±0.04%, respectively. While at 72 hours were 3.57±0.04, 3.63±0.04, 3.58±0.04, 3.55±0.05 and 3.60±0.05%, respectively. While the mean values of fat % of heat treated milk in aluminium pan without additive, as well as heated milk in aluminium pan with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and1% at 0 hours were 6.95±0.05, 6.93±0.04, 6.87±0.07, 6.85±0.02 and 6.90±0.08%, respectively. At 24 hours were 6.46±0.04, 6.45±0.13, 6.57±0.07, 6.45±0.07 and 6.46±0.08%, respectively. At 48 hours were 6.52±0.04, 6.42±0.12, 6.4±0.08, 6.44±0.05 and 6.43±0.06%, respectively and at 72 hours were 6.15±0.08, 6.15±0.04, 6.23±0.04, 6.46±0.04 and 6.42±0.02%, respectively.

Regard the result of protein % and fat% of heat -treated milk in aluminium pan without additive, milk was heated in aluminium pan with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and1% at 0 hours. The recorded data revealed a non-significant change of protein% during the experiment. Meanwhile fat% showed a significant difference after 72 hours of storage between heat treated milk and the heated samples with sucrose 1% and fructose 0.5% and 1%. Sugar as a natural preservative **(Seetaramaiah *et al.,* 2011)** in such concentration are capable of preserving milk samples and delaying fatty acids oxidation and maintain the quality of milk then increase the shelf life of it. Chemical and physical alterations as auto oxidation and trans fatty acids formation of milk lipids during storage were stated by **(Semma, 2002).**

Table (4) showed the mean values of calcium, chloride and sodium levels of heat treated and heated milk with additives samples, the mean values of calcium levels of heat treated milk in aluminium pan without additive in which milk was heated in aluminium pan with addition of sucrose 0.5% and 1% as well as milk heated in aluminium pan with addition of fructose 0.5% and 1% at 0 hours were 38.86±1.2, 39.76±0.98, 39.5±0.96, 39.55±0.9 and 39.02±0.6mg/dl. At 24 hours they were 40.3±1.01, 39.73±0.67, 39.83±0.62, 38.6±0.7 and 39.97±1.02mg/dl. And at 48 hours they were 40.47±0.8, 39.6±0.9, 39.6±0.8, 40.7±0.89 and 39.7±0.96mg/dl. While 72 hours were 39.5±0.75, 38.85±0.29, 39.27±0.49, 39.31±0.59 and 39.8±0.49mg/dl, respectively. The mean values of chloride levels of heat treated milk in aluminium pan without additive as well as milk was heated in aluminium pan with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and 1% at 0 hours were 55±1.15, 55.33±1.2, 58±0.88, 56.67±1.2 and 58.67±0.98mg/dl. At 24 hours were 52.33±0.97, 54.67±1.65, 58.67±0.98, 56±1.52 and 57.33±1.45 mg/dl. And at 48 hours were 48±1.53, 53.33±0.88, 58±1.15, 56±0.98 and 56.67±1.45mg/dl. While at 72 hours were 45±0.25, 54±0.14, 57±0.26, 57.67±0.88 and 58±0.88mg/dl, respectively. Moreover, the mean values of sodium levels of heat treated milk with addition of sucrose 0.5% and 1% and milk was heated in aluminium pan with addition of fructose 0.5% and 1% at 0 hours were 41.67±0.05, 43.67±0.88, 45±1.15, 44±1.15 and 47±0.88mg/dl, respectively. At 24 hours were 40.33±0.88, 42.67±1.2, 45±1.15, 44.33±1.2 and 46.67±0.88mg/dl, respectively. While, at 48 hours were 36±1.15, 39±0.98, 42±1.15, 40±0.88 and 44±0.88 mg/dl, respectively. Also at 72 hours were 34.76±0.76, 39±0.88, 42±1.15, 40±1.15 and 43.67±0.98 mg/dl, respectively.

Regarding the result of milk whey electrolyte, its calcium levels recorded a non-significant change between concerned groups. Meanwhile chloride and sodium level showed a significant difference between them with obvious decrement their levels in raw boiled milk sample through the experiment, the treated groups maintain their levels. Decrease of chloride ions after boiling can be explained as the chloride ions incorporated into the growing aluminium oxide film **(Lee and Pyun, 1999)** meanwhile, sucrose 1% and fructose 0.5 and 1% can inhibit the aluminium corrosion by formation of coating sort **(Marta *et al.,* 2006)** and consequently maintain the chloride level. On the other hand, Sodium level change was parallel with that of chloride level. A direct correlation between chloride and sodium ions concentration in milk was stated **Fox and McSweeney, (1998).**

Conclusion and Recommendation

From the obtained results, it could be concluded after estimation of aluminium residue in each of heat treated milk in aluminium pan without additive, milk was heated in aluminium pan with addition of sucrose 0.5% and 1% and milk with addition of fructose 0.5% and 1% that milk with addition of fructose 1% was the best one concerning the aluminium residue followed by milk with addition of sucrose 1%. The heat-treated milk in aluminium pan without additive was the least one. Sucrose and fructose decrease aluminium migration into milk and can be used as natural preservative to maintain its quality and to increase its shelf life. On the other hand, it is recommended to heat milk in Copper-bottomed stainless-steel vessels instead of aluminium vessels because the copper bottom helps the vessel heat up fast and the stainless steel is easy to clean and not leave harmful residue.

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