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Leaching of Lead, Chromium and Copper into Drinks Placed in Plastic Cups at Different Conditions

Oliver Ahimbisibwe[1*], Denis Byamugisha[1], Paul Mukasa[1], Timothy Omara[2,3], Emmanuel Ntambi[1]

[1]Department of Chemistry, Faculty of Science, Mbarara University of Science and Technology, Mbarara, Uganda
[2]Department of Chemistry and Biochemistry, School of Sciences and Aerospace Studies, Moi University, Eldoret, Kenya
[3]Africa Center of Excellence II in Phytochemicals, Textile and Renewable Energy (ACE II PTRE), Moi University, Eldoret, Kenya
Email: *oliverahimbisibwe@gmail.com

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Abstract

Plastic cups commonly used by communities of Uganda lose color with time as they are used. The purpose of this study was to determine the concentrations of copper, lead and chromium in drinks placed in blue and green plastic cups from two companies (anonymized as A and B) in Uganda. New blue and green polyethylene plastic cups produced from unrecycled material were purchased from company A and company B while control cups of Luminarc white (Saudi Arabia) and porcelain white glazed (China) were purchased from shops in Mbarara city. The ash from the burnt blue and green plastic cups was digested using *Aqua Regia*. After a holding time of 30 and 60 minutes, milk and tea samples in the four different cups were also digested using *Aqua Regia*. Sample analysis for the supernatants was done in triplicates using an atomic absorption spectrophotometer. The ash of blue and green cups from company A was found to contain the highest concentration of heavy metals which ranged from 0.73 to 1.86 ppm for lead, 8.05 to 38.32 ppm of copper, and 655.80 to 756.57 ppm for chromium. For company B, the heavy metal concentrations ranged from 0.76 to 1.43 ppm, 5.51 to 22.85 ppm, and 756.67 to 815.25 ppm, respectively. The concentration of the leached metals ranged from 0.001 to 5.75 ppm for copper, 0.001 to 0.60 ppm for chromium, and 0.001 to 0.41 ppm for lead for the cup of company A. For the cup from company B, the concentration ranges for the leached metals were from 0.001 to 3.59 ppm, 0.001 to 1.08 ppm, and 0.001 to 0.29 ppm, respectively. Most of the values obtained for the leached concentrations of the metals studied were above the respective drinking water in WHO and USEPA permissible limits of 1 ppm (copper), 0.05 ppm (lead) and 0.1 ppm (chromium). The blue and green plastic cups from both companies had leached concentrations of copper, lead and chromium below the permissible limits at 25˚C, pH 6.2 (milk) and a holding time of 30 minutes. The control cups did not leach detectable amounts of copper, chromium and lead under the study conditions, hence it may be safe for holding the hot drinks at the temperatures experimented in this study.

Keywords

Plastic Cups, Leaching, Metal Concentration

1. Introduction

Since the 1950s, global demand for plastics from synthetic polymers have surged to over 368 million metric tons [1], due to their numerous advantages in modern life such as easy to mold and recycle into different desired plastic products. The commonly used form of plastics includes polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polyurethane (PU), polyvinyl chloride (PVC), polystyrene (PS), polycarbonate (PC), low-density polyethylene (LDPE), and high-density polyethylene (HDPE) [2] [3].

The different forms of plastics contain several materials (additives and catalysts) which are added for a specific purpose during their synthesis. The additives and/or catalysts in plastics are basically hazardous metal compounds which keep circulating in the environment due to the pervasiveness of plastic products utilized frequently. Unfortunately, the presence and the effects of the hazardous metals in plastics are less perceived once released in nature [3].

The most common hazardous metal compound additives and catalysts used in plastics include inorganic compounds like zinc oxide (ZnO) and antimony (III) oxide (Sb_2O_3) in PET, both used as white pigments, and also as fungicide and flame retardant synergist respectively. Chromium (VI) trioxide (CrO_3) in PE, lead sulphate ($PbSO_4$), lead chromate ($PbCrO_4$), lead molybdate ($PbMoO_4$), tetralead trioxide sulphate ($3PbO \cdot PbSO_4$), trilead bis(carbonate) dihydroxide ($Pb_3(CO_3)_2(OH)_4$), trilead dioxide phosphonate ($PbHPO_3 \cdot 2PbO$), lead oxide (PbO), are added in all plastics for pigmentation, fastness, and heat stability [4]. Basic copper carbonate hydroxide is added for pigmentation [5], copper oxide is added as an antimicrobial agent [6] during the synthesis of plastics. The principal uses of metal-based additives in plastics are inert fillers, pigmentation for colour, and stabilization [7]. Other additives may be biocides, lubricants, stiffness and hardness [3].

The different forms of plastics are used to manufacture vast products which serve various roles globally. The demand for a given product determines the type of plastic to be used for the production of that product to the market. For instance, PET is mainly used to produce packaging bottles of several drinks; PVC is used to make cooking oil cans, baby bottles nipples, coffee cups; PS is used in the making of hot beverage cups; while HDPE is used to make jerricans, milk bottles, plastic plates, mugs [2], among others. The plastic products manufactured are frequently used by humans, and modern daily life has become unthinkable without plastics [8].

Significant health concerns over human exposure to hazardous metals from plastics have increased over years. This is because research has significantly linked human diseases to metal contamination [9] from almost all plastics used for carrying, bottling and storage of foodstuffs in daily life. The metal contamination from plastics is attributed to leaching out of the hazardous metals into foodstuffs placed and kept in plastic products at certain conditions [2]. The influential factors of metal leaching from plastics into food materials reported include pH, sunlight, contact time and temperature [2] [10]. Some of the metals which leach from plastics have been classified with health risks [9], for example, lead is known to cause alteration of genetic material, delayed mental development, cancer, headaches, insomnia and nervousness, and tooth decay; chromium causes lung cancer, kidney and liver damage, while copper may cause anemia, liver and kidney failure [11]. Due to the above health effects to humans exposed to hazardous metals leached from plastics, quality control agencies in several countries like those of Europe have developed regulations on the use of metal additives in plastics [3], and many of the regulations are adopted by a wider international community [3] [12], so as to mitigate health risks on humans using plastic products. For example, the European regulatory directive on hazardous metals in plastics intended to come into contact with foodstuffs indicates that the limit values (in $mg \cdot kg^{-1}$) should be 2, 1 and 5 for lead, chromium (VI) and copper [3] respectively.

Hazardous metal leaching from plastics resulting in metal contamination of foodstuffs such as bottled water [2] [9] [13] [14] and packed food [15] have been evaluated by many researchers. However, evaluation of leaching of metals from other plastic products like jugs, cups, mugs intended to handle foodstuffs such as dry coffee, tea, milk tea, porridge (cold or hot drinks) that are frequently used for human consumption at different conditions has been given minimal attention.

Uganda consumes up to 600 metric tons of plastics per day of which 6% is collected as waste [16], much of which is PET [17], while the rest of the plastics contribute to environmental pollution [18]. Plastics used in Uganda from over 30 companies are basically recycled plastics of which 22% is HDPE (used to make jerricans, plates, buckets, jugs, cups, mugs). The low percentage of HDPE recycled plastic products is due to their long-lasting nature [18]. This fact creates an implication that humans interact with HDPE plastic products such as plastic cups for a long time, and consequently longtime exposure to metal contamination most likely occurs as metals leach from such plastics into foodstuffs like cold and hot drinks consumed on a daily basis by an individual. Therefore, a study aimed at generating information about the leaching of metals from some plastic cups used in several homes in Uganda was worth undertaking. The plastic cups used in a home setting can hold both cold and hot drinks such as cold or hot water, hot dry (African) tea/coffee and cold or hot milk, among other drinks. During the study, plastic cups without any drink and the drinks placed in the plastic mugs at varying conditions of pH, temperature and time were evaluated for metal concentrations. The information obtained will help quality control regulation bodies to take action based on the generated information and to make the population be aware of the possible food contamination resulting from the use of plastics.

2. Methods and Materials

2.1. Collection of Cups for the Study

Twenty-four plastic cups (12 blue and 12 green) of volume 0.5 L, made from recycled HDPE plastics which may hold both cold and hot drinks were purchased and selected randomly from the available batches at the premises of two companies (coded A and B) in Kampala, Uganda in November, 2018. For comparison during analysis, twelve control cups (six luminarc white and six glazed all-white porcelain) of volume 0.3 L, made in Saudi Arabia and China respectively were purchased from a supermarket in Mbarara city, Uganda. All the cups purchased were transported to the Mbarara University of Science and Technology (MUST) analytical laboratory, thoroughly rinsed three times with deionized water to remove any metal contamination stems produced during their manufacturing [2] and air dried.

2.2. Determination of Metal Concentration in Cups

One randomly chosen blue plastic cup from company A (BA) and another blue cup from company B (BB) were cut into small pieces using a stainless steel laboratory knife, separately placed in a crucible and ashed in muffle furnace at 600˚C. All the ash for each cup obtained was separately digested using concentrated nitric acid (AG 71% w/w, UNILAB), concentrated hydrochloric acid (AG 36.5% w/w, UNILAB) and deionized water in the ratio of 3:1:10, at 300˚C for 30 minutes [19], mixture was cooled, filtered, and the filtrates made to 100 mL mark in a volumetric flask with deionized water. Separate filtrates from one green plastic cup from company A (GA), then from company B (GB) were obtained using the same method. Absorbance of each filtrate was measured by using a graphite furnace atomic absorption spectrophotometer, GF-AAS.

2.3. Collection and Preparation of Drinks

Two kinds of drinks were used in the study; pure milk and dry tea. The milk (5 L, lactometer density of 1.03 g/cm^3) was purchased from a farmer in Kiruhura District, Uganda, placed in a clean stainless-steel milk container and transported to MUST analytical laboratory. Three liters of the milk were measured into a clean stainless-steel saucepan, boiled on a hot plate, and pH value for the milk (6.2 ± 0.1) was measured using pH meter (Hanna 991300). The procedure was repeated using deionized water instead of milk in another stainless-steel saucepan and 6 g of tea leaf powder (Igara tea brand, purchased from a supermarket in Mbarara city, Uganda), was added in the boiling deionized water to form dry tea and its pH (4.2 ± 0.1) was determined. The two drinks prepared were kept at boiling point for further experiments because the two drinks are taken by adult humans when hot and after boiling.

2.4. Determination of Metal Concentrations Leached in Drinks at Different Conditions

To determine the concentration of Pb, Cr and Cu that may have leached from the stainless saucepan into drinks, 10 mL of the boiling milk was pipetted directly from the saucepan into a 250 mL glass beaker without allowing any contact with the plastic cups, and digested using 6 mL concentrated nitric acid, 2 mL concentrated hydrochloric acid on a hot plate at 300˚C for 30 minutes with agitation, a clear solution mixture obtained was cooled, filtered and the filtrates made to 100 mL mark in a volumetric flask with deionized water. The procedure was repeated using dry tea to obtain the filtrates for analysis.

To determine the concentration of Pb, Cr and Cu that may have leached from the plastic cups into drinks, the following setups were made. Using a 500 mL glass beaker, one blue plastic cup from company A and one blue plastic cup from company B were each half filled with boiling milk and 10 mL were pipetted and digested as described before at different time intervals of 30 and 60 minutes to obtain the filtrates for analysis. Three blue plastic cups from company A were each half filled with cooling boiled milk at 95˚C, 65˚C and 25˚C, each cup placed in the respective water bath maintained at 95˚C, 65˚C and 25˚C, 10 mL pipetted from each cup after 30 minutes, and digested to obtain the filtrates for analysis. Three blue plastic cups from company B were similarly prepared at the same varying temperature.

The experimental set up was repeated using green plastic cups from company A and B; and then using dry tea at varying contact time and temperature for the plastic cups from company A and B and later for control cups.

Each of the filtrates obtained were analyzed in triplicate using Agilent 240z graphite furnace atomic absorption spectrophotometer (GFAAS) at the Department of Geology, Makerere University, Uganda. The metal concentrations were determined using the oxy-acetylene-argon gas flame at the appropriate wavelength of each metal (283.3, 357.9, 324.8) nm for lead, chromium and copper respectively. The instrument was calibrated using 0.5 ppm, 1 ppm, 2 ppm and 5 ppm working standards prepared by dilution from 1000 ppm analytical grade stock solutions of lead, copper and chromium salts. The instrument readings in μg/l for each metal were recorded, converted to ppm reported as mean ± SD metal concentrations leached into the drinks by using Equation (1).

$$\text{Metal concentration}\left(\text{ppm}\right)=\frac{\text{Vol}\times\alpha\times df}{1000\times wt} \qquad (1)$$

where α is the metal instrument reading in μg/L, vol is volume of sample used, df is the dilution factor, and wt is the weight of the sample. Microsoft excel (2016) was employed for statistical analysis and a one tail t-test was used to compare the means of different cases, $P < 0.05$ was taken significant statistically.

3. Results and Discussions

3.1. Total Concentration of Lead, Chromium and Copper in Plastic Cups

A representative sample of plastic cups collected for the study was analyzed for metal concentration using GFAAS. Table 1 shows the results obtained.

All the blue and green cups contained Pb, Cr and Cu metals in varying ranges (Table 1), Cr had the highest concentration (815.25 ± 0.13 ppm) while Pb had the least concentration (0.73 ± 0.04 ppm). The presence of the three metals studied in the plastic cups is evident of the inorganic compounds like oxides, carbonates and hydroxides of Pb, Cr and Cu [4] which are used in the manufacture of plastics. The high concentration of Cr in the cups may be due to the frequent use of chromium compounds which are intensely coloured, and the metal being resistant to corrosion and discoloration [20] hence applied in large concentrations during synthesis of plastics. Lead was of least concentration probably because of its limited use in plastic making due to its high toxicity [21] despite the fact that lead compounds are efficient heat stabilizers for plastics [22]. The results shows that the total mean concentrations of Pb, Cr and Cu in the plastic cups are above the European regulatory directive adopted by many countries on hazardous metals in plastics made but intended to come into contact with foodstuffs which should be 2, 1 and 5 for lead, chromium and copper (in $mg \cdot kg^{-1}$) [3] respectively. This calls for quality control institutions like Uganda National Bureau of Standards and policy makers in Uganda to formulate heavy metal regulatory policies and ensure that industries implement quality control measures at the production stage before products like cups are brought to market. The results in the current study agree with other studies which report high concentrations of metals in plastics made for foodstuff holding such as water bottles, food packaging as evidence that hazardous metal compounds are used in the synthesis of plastics [3] [9] [14].

3.2. Concentration of Pb, Cr and Cu that Leached from Plastic Cups into Drinks at Varying Temperature

Two different forms of drinks; milk and dry/African tea were used in this study and brought into contact with the plastic cups at varying temperatures. Table 2

Table 1. Concentration of Pb, Cr and Cu in plastic cups collected.

Type of mug	Metal concentration (ppm), n = 3		
	Pb	Cr	Cu
BA	1.86 ± 0.07	756.57 ± 0.28	38.32 ± 0.16
BB	1.43 ± 0.09	815.25 ± 0.13	22.85 ± 0.12
GA	0.73 ± 0.04	655.80 ± 0.15	8.05 ± 0.11
GB	0.76 ± 0.08	756.67 ± 0.31	5.51 ± 0.15

BA, GA and BB, GB = Blue, Green mug from company A and B.

Table 2. Concentration (in ppm) of Pb, Cr and Cu that leached into drinks from plastic and control cups at varying temperature.

Drink	Cup type	Temperature (°C), n = 3								
		25			65			95		
		Pb	Cr	Cu	Pb	Cr	Cu	Pb	Cr	Cu
Milk	BA	nd	0.021 ± 0.004	0.024 ± 0.001	0.1 ± 0.007	0.125 ± 0.003	0.17 ± 0.003	0.439 ± 0.01	0.24 ± 0.005	0.335 ± 0.001
	BB	nd	0.004 ± 0.004	0.09 ± 0.001	0.35 ± 0.005	0.23 ± 0.009	0.43 ± 0.013	0.089 ± 0.005	0.39 ± 0.007	0.555 ± 0.005
	GA	nd	0.021 ± 0.001	0.024 ± 0.001	0.044 ± 0.005	0.089 ± 0.007	0.156 ± 0.009	0.069 ± 0.003	0.21 ± 0.003	0.325 ± 0.005
	GB	nd	0.025 ± 0.001	0.075 ± 0.001	0.026 ± 0.001	0.195 ± 0.003	0.365 ± 0.006	0.074 ± 0.002	0.35 ± 0.004	0.5 ± 0.006
	Ctl	nd	nd	nd	nd	nd	nd	nd	nd	nd
Dry tea	BA	nd	0.095 ± 0.001	0.346 ± 0.006	0.056 ± 0.002	0.24 ± 0.012	2.495 ± 0.028	0.165 ± 0.001	0.485 ± 0.008	3.28 ± 0.004
	BB	nd	0.175 ± 0.002	1.995 ± 0.039	0.055 ± 0.003	0.555 ± 0.007	3.08 ± 0.02	0.22 ± 0.002	1.02 ± 0.002	3.47 ± 0.03
	GA	nd	nd	0.325 ± 0.003	0.083 ± 0.009	0.183 ± 0.003	1.915 ± 0.008	0.295 ± 0.002	0.4 ± 0.006	2.61 ± 0.004
	GB	nd	0.055 ± 0.001	1.895 ± 0.02	0.099 ± 0.002	0.15 ± 0.002	2.755 ± 0.03	0.21 ± 0.002	0.38 ± 0.004	3.29 ± 0.022
	Ctl	nd	nd	nd	nd	nd	nd	nd	nd	nd

BA, GA and BB, GB = Blue, Green cups from company A and B, Ctl = control, nd = not detected.

shows the concentration Pb, Cr and Cu that leached into the drinks.

The concentrations of the leached lead, chromium, and copper into the milk and dry tea from the plastic cups increased with increase in temperature (Table 2). The heat facilitates the dissolution there by providing energy to break bonds of the additives used in making the plastics. The increase in kinetic energy associated with higher temperatures allows the solvent molecules to more effectively break apart the additives' molecules that are held together by intermolecular attractions [23].

At all temperatures, there was leaching of metal concentration ($P<0.05$, especially for Cu) into dry tea than milk. This observation may be supported by the fact that carbonates, oxides and hydroxides of lead, chromium, and copper used as additives in the making of plastic cups react and leach most into more acidic media of dry tea (pH 4.2) than in less acidic media of milk (pH = 6.2) [24].

Generally, the concentrations of leached copper in the cups were greater than those of lead and chromium in both dry tea and milk. The high concentration of copper in both dry tea and milk may be due to the use of strongly basic copper carbonate hydroxide ($Cu_2(OH)_2CO_3$) to make blue and green pigments used in the making of the cups, which are easily neutralized by acidic condition in dry tea than in milk (Equations (2) and (3)) than the additives of chromium and lead, which are oxides, hydroxides and chromates [25].

$$\mathrm{CuCO_3(s)+2H^+(aq)\rightarrow Cu^{2+}(aq)+CO_2(g)+H_2O(l)} \quad (2)$$

$$\mathrm{Cu(OH)_2(s)+2H^+(aq)\rightarrow Cu^{2+}(aq)+2H_2O(l)} \quad (3)$$

Blue and green cups from company A and company B generally leached concentrations of the metals above the WHO and USEPA permissible limits in drinking water of 0.05 ppm for lead, 0.1 ppm for chromium and 1 ppm for copper [26] except at 25˚C, the lowest temperature used in this study.

All the luminarc white and porcelain white cups gave the concentrations of leached lead, chromium and copper into the milk and dry tea below detection level at the temperatures 25˚C, 65˚C and 95˚C**.** The results agree with the study findings in Turkey that the extent of leaching from porcelain depends on glaze composition and firing conditions [27] and hence control cups used in this study were probably glazed using material free from lead, chromium and copper and prepared using appropriate firing conditions.

3.3. Concentration of Pb, Cr and Cu that Leached from Plastic Cups into Drinks at Varying Contact Times

Time of which the drinks were allowed to be in contact with the plastic cups varied and the results in **Table 3** were obtained.

The plastic cups leached significant ($P < 0.05$) concentrations of lead, chromium and copper into the milk and dry tea at contact times of 60 than 30 minutes (**Table 3**). This is because the leaching of the metals from the polymer matrix occurs by diffusion process. The diffusion flux that causes the accumulation of lead, chromium and copper in the drinks from the plastic depends on the amount of time the milk or the dry tea spends in the cup. The more the time of contact between the plastic and the drink, the more the concentration leached. This is in agreement with similar studies that investigated the effect of contact time on metal migration from PET bottles into water and significant temperature influence was reported with increase in contact time [9] [14].

All cups from company A and company B generally leached concentrations of

Table 3. Concentration (in ppm) of Pb, Cr and Cu that leached into drinks from plastic and control cups at varying contact time.

Drink	Cup type	Contact time (minutes), n = 3					
		30			60		
		Pb	Cr	Cu	Pb	Cr	Cu
Milk	BA	0.02 ± 0.004	0.11 ± 0.004	0.11 ± 0.002	0.337 ± 0.008	0.147 ± 0.004	0.24 ± 0.001
	BB	0.022 ± 0.004	0.117 ± 0.006	0.14 ± 0.001	0.273 ± 0.003	0.34 ± 0.007	0.58 ± 0.007
	GA	0.032 ± 0.003	0.066 ± 0.004	0.10 ± 0.008	0.043 ± 0.003	0.147 ± 0.003	0.707 ± 0.011
	GB	0.028 ± 0.003	0.097 ± 0.002	0.12 ± 0.007	0.038 ± 0.001	0.239 ± 0.003	0.503 ± 0.001
	Ctl	nd	nd	nd	nd	nd	nd
Dry tea	BA	0.053 ± 0.001	0.193 ± 0.002	0.47 ± 0.003	0.093 ± 0.001	0.354 ± 0.005	3.61 ± 0.022
	BB	0.06 ± 0.001	0.523 ± 0.005	2.77 ± 0.023	0.123 ± 0.002	0.64 ± 0.003	2.92 ± 0.035
	GA	0.313 ± 0.003	0.112 ± 0.002	0.917 ± 0.005	0.166 ± 0.004	0.277 ± 0.004	2.32 ± 0.0053
	GB	0.089 ± 0.001	0.107 ± 0.003	2.41 ± 0.044	0.117 ± 0.002	0.283 ± 0.002	2.90 ± 0.002
	Ctl	nd	nd	nd	nd	nd	nd

BA, GA and BB, GB = Blue, Green cup from company A and B;Ctl = control, nd = not detected.

lead, chromium and copper above the WHO and USEPA permissible limits in drinking water. In all Luminarc white and porcelain white cups, the concentration of the metals investigated were below detection limit in the milk and tea solutions at the two contact times.

4. Conclusions

All the blue and green plastic cups from companies A and B were found to contain concentrations of Pb, Cr and Cu metals above the regulatory directives for plastics made intended for foodstuffs contact. The study revealed that blue and green cups of the two companies A and B leached into the two drinks managed at varying temperatures and contact times. The concentration of Pb, Cr and Cu that was only below the permissible limits was at 25˚C when using milk. Ugandans using plastic cups might, therefore, be consuming hazardous metals in the contaminated drinks which over time may exert health risks associated with these metals. The blue and green cups of the two companies A and B should consciously be used for handling drinks especially acidic drinks at temperatures above 25˚C, instead, Luminarc white and all-white porcelain cups may be a good alternative.

The National standards agency should ensure that all companies processing plastic containers in Uganda use plastic additive compounds that meet the requirements for making colored plastics and it should ensure that proper quality assurance is done during the production line/process. Regular examination of the leaching of metals and organic additives from plastics made in Uganda is worth doing under different conditions at all times.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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