Evaluation of Leached Heavy Metals Using Various Leaching Agents in Self-Produced Ceramics

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***Abstract*:- This study was aimed at evaluating the leaching ability of self-produced glazed ceramic food wares produced from locally available raw materials. The raw materials (rice husk, quartz and feldspar) were acquired from various locations within Bauchi, Bauchi State, Nigeria, while silt stone was obtained from Numan River Basin, Adamawa State, Nigeria. Leaching tests on the self-produced ceramics were conducted using various leaching agents (4.00 % acetic acid, boiled water, orange juice, boiled lipton tea and 1.00 % lactic acid). The levels of heavy metals leached in the self-produced ceramics/leaching agents were determined using Buck Scientific Atomic Absorption Spectrophotometer Model 210 VGP. The levels of nickel, chromium, manganese, cobalt, zinc, cadmium and lead (μg/cm^3) determined in the various leaching agents respectively ranged from 0.01 in lactic acid to 0.15 in lipton tea; not detected in lipton tea to 0.14 in boiled water; 0.09 in acetic acid to 2.70 in lipton tea; 0.06 in lactic acid to 1.47 in lipton tea; not detected in acetic acid, boiled water, orange juice and lipton tea to 119.20 in lactic acid; 0.01 in acetic acid to 0.26 in lactic acid and 0.04 in orange juice to 1.03 in lipton tea. The concentrations of the heavy metals leached in the self-produced ceramics are mostly below their safe permissible limits. Ceramics of low levels of heavy metals leaching ability are therefore produced and hence safe for human utilization and consumption.**

***Keywords:* Glazed, food wares, rice husk, quartz, feldspar, self-produced ceramics, leaching agents, heavy metals, permissible limits**

I. INTRODUCTION

The term “Ceramic” has been applied traditionally to earthenware objects produced by moulding and subsequent firing of moist clay at a low temperature to form hard and dense solids. Presently, ceramics are inorganic and non-metallic crystalline materials that are manufactured by heat treatment [1]. Ceramics are class of materials broadly defined as inorganic, non-metallic solids that provide the broadest range of functions of all known materials. They can be produced in the form by glasses, single crystals as well as poly-crystals [2]. They are also considered as solid compounds that are synthesized by the application of heat and in some cases heat and pressure, which is made up of at least one metal and a non-metallic elemental solid or a non-metal, a combination of at least two non-metallic elemental solids and a non-metal [3]. The earliest forms of ceramics were pots, vessels and figurines that were produced from clay alone or combined with some other materials like silica, hardened and sintered in fire. They were initially rough to touch, very porous and this brought about the discovery and the use of glazes in order to take care of these demerits [4].

Silica is the vital glaze component that occurs in nature as quartz. This enhances the fusion of clay and the glaze material. The melting point of silica is relatively high and therefore a flux is added in order to reduce the melting point. Naturally occurring feldspar as well as lead, magnesium, sodium and lithium are used as fluxes. Oxides of a number of metals are also used in glazes. When glazed ceramics are made to come in contact with acidic foods or liquids, leaching of the fused heavy metals may occur [5][6]. As good and aesthetic as glazed ceramics are, they can release toxic heavy metals or metals into food substances or edible substances that are placed or kept in them and therefore constituting health hazards to the users [7]. The aim of this research work is to embark on evaluation of leached heavy metals using various leaching agents in self-produced ceramics.

II. MATERIALS

Buck Scientific Atomic Absorption Spectrophotometer Model 210 VGP was used in this research. Analytical reagent (AnalaR) grade chemicals and distilled water were used in the study. All the glass and plastic wares used were thoroughly washed with detergent solution, 20.00 % (v/v) nitric acid, rinsed with tap water and finally with distilled water. The apparatus were then allowed to dry [8].

III. METHODS

Sampling and Sample pre-treatment

Clay, feldspar, rice husk and quartz were obtained from various locations in Bauchi, Bauchi State, Nigeria, whereas silt stone was obtained from Numan River Basin, Adamawa State, Nigeria. The clay was homogenized, soaked in water for three (3) days, sieved through a 250 μm sieve, allowed to dry and harden enough, then mould into cups and soup bowls. The ceramics produced were dried and bisque fired in a ceramic kiln at 900˚C. Quartz, feldspar and rice husk were also bisque fired at 900˚C in the ceramic kiln, crushed, ground, sieved using a 250 µm sieve, homogenized and weighed in different proportions to form glazes.

Quadiaxial test blends using quartx, rice husk ash, feldspar and Numan silt stone were produced and the best glazes obtained were applied to the self-produced ceramics. The self-produced ceramics were then subjected to leaching tests [9][10].

Leaching Tests

Freshly prepared acetic acid (4.00 %), lactic acid (1.00 %), boiled lipton tea (100˚C), orange juice and boiled water (100˚C) were respectively used as leaching agents in the self-produced ceramics. Each self-produced ceramic was separately filled with the leaching agent, covered with polythene, then topped with aluminium foil in order to avoid evaporation and also prevent reaction of some heavy metals such as cadmium with light [11]. Acetic acid was allowed to stand overnight (24 hours), while other leaching agents remained in the ceramic wares for 20 minutes.

Aliquot volume of each of the resulting leachate was measured into five separate 50 cm^3 volumetric flasks, water added to capacity and heavy metals of interest were determined at their respective wavelengths using Buck Scientific Atomic Absorption Spectrophotometer Model 210 VGP [9][11][12].

IV. RESULTS AND DISCUSSION

Results

Table 1 indicates the concentra-tions of nickel, chromium manganese and cobalt ($\mu g/cm^3$) leached in different leaching agents investigated in the self-produced ceramics, whilst Table 2 reveals the levels of zinc, cadmium and lead ($\mu g/cm^3$) leached in the various leaching agents studied in self-produced ceramics.

Table 1: Concentrations of Nickel, Chromium, Manganese and Cobalt ($\mu g/cm^3$) Leached in Different Leaching Agents

Leaching Agents	**Heavy metals**			
	Ni	**Cr**	**Mn**	**Co**
Acetic Acid (4.00 %)	0.02 ± 0.00	0.01 ± 0.00	0.09 ± 0.01	0.22 ± 0.00
Lactic Acid (1.00 %)	0.01 ± 0.00	0.09 ± 0.02	0.81 ± 0.10	0.06 ± 0.01
Boiled Lipton Tea	0.15 ± 0.01	ND	2.70 ± 0.16	1.47 ± 0.29
Orange Juice	0.03 ± 0.00	0.01 ± 0.00	2.52 ± 0.08	0.30 ± 0.03
Boiled Water	0.03 ± 0.01	0.14 ± 0.02	0.65 ± 0.02	0.73 ± 0.02

Values are mean ± standard deviation (n = 3), ND = Not detected.

Table 2: Levels of Zinc, Cadmium and Lead ($\mu g/cm^3$) Leached in Various Leaching Agents

Leaching Agents	**Heavy Metals**		
	Zn	**Cd**	**Pb**
Acetic Acid (4.00 %)	ND	0.01 ± 0.00	0.23 ± 0.02
Lactic Acid (1.00 %)	119.20 ± 2.14	0.26 ± 0.06	0.90 ± 0.08
Boiled Lipton Tea	ND	0.05 ± 0.00	1.03 ± 0.06
Orange Juice	ND	0.03 ± 0.00	0.04 ± 0.01
Boiled Water	ND	0.02 ± 0.00	0.62 ± 0.15

Values are mean ± standard deviation (n = 3), ND = Not detected.

Discussion

Concentrations of Nickel, Chromium, Manganese and Cobalt in Different Leaching Agents

The concentrations of nickel determined (μg/cm^3) in different leaching agents investigated ranged from 0.01 (1.00 % lactic acid) to 0.15 (boiled lipton tea) as illustrated in Table 1. Concentrations of 0.02 and 0.03 μg/cm^3 of nickel respectively determined in 4.00 % acetic acid and boiled water (orange juice) leaching agents were found to fall in between the observed extreme concentrations. The observed values of nickel determined in different leaching agents placed in the self-produced ceramics are lower than reported literature values of 70.00 to 134.00 μg/L (0.07 to 0.134 μg/cm^3) [13] as well as 0.20 mg/L (0.20 μg/cm^3) [14]. The concentration of nickel found in boiled lipton tea (0.15 μg/cm^3) compares fairly well with 0.07 to 0.134 μg/cm^3 [13]. All the observed values of nickel in different leaching agents are lower than 0.20 μg/cm^3 guideline for safe limits of nickel [15]. In extreme circumstances, nickel can cause degenerative respiratory disease that can kill [16].

The concentrations of chromium (μg/cm^3) leached in the self-produced ceramics ranged from not detected (boiled lipton tea) to 0.14 in boiled water. The levels of 0.01 μg/cm^3 respectively found in 4.00 % acetic acid and orange juice as well as 0.09 μg/cm^3 determined in 1.00 % lactic acid fell in between the extreme levels (not detected to 0.14 μg/cm^3) of the observed chromium. All the observed values of chromium with the exception of 0.14 μg/cm^3 are lower than reported literature value of 0.10 μg/cm^3 (0.10 mg/L) [17]. The determined values are also lower than 0.09 to 0.60 mg/dm^3 (0.09 to 0.60 μg/cm^3) [18]. All the chromium leached in the leaching agents investigated are lower than the maximum permissible limits of 2.30 mg/dm^3 (2.30 μg/cm^3) set by WHO, 2004 [19]. Chromium can accumulate in hair, skin, lungs, nails, liver, muscle fat, as well as placenta, where it is responsible for different health conditions in man [20].

The level of manganese (μg/cm^3) assayed in various leaching agents ranged from 0.09 (found in 4.00 % acetic acid) to 2.70 (determined in boiled lipton tea). Concentrations of 0.65 μg/cm^3 (found in boiled water), 0.81 μg/cm^3 (assayed in 1.00 % lactic acid) and 2.52 μg/cm^3 (found in orange juice) were found to fall in-between the range of manganese (0.09 to 2.70 μg/cm^3) determined. The observed values are mostly in good agreement with literature values of 0.01 to 2.14 mg/L (0.01 to 2.14 μg/cm^3) [18].

The levels of manganese in the present study are generally higher than the threshold limit of 0.50 μg/cm^3 set by WHO, 2004 [19]. The high level of manganese leached by various leaching agents in the self-produced ceramics may be due to the fact that the element naturally occurs in rocks as well as soil and might therefore be present in the parent rocks from which the raw materials were sourced [21]. Manganese is significant in the normal physiological functioning of both man and animals as well as in human nutrition [22].

The concentrations of cobalt leached in various leaching agents investigated ranged from 0.06 μg/cm^3 (found in 1.00 % lactic acid) to 1.47 μg/cm^3 (found in boiled lipton tea). The metal levels (μg/cm^3) of 0.22 (assayed in 4.00 % acetic acid), 0.30 (determined in orange juice) as well as 0.73 (found in boiled water) fell in between the range of cobalt concentrations determined. The experimental values (0.06 to 1.47 μg/cm^3) are generally lower than literature values of 0.39 to 41.46 ppm (0.39 to 41.46 μg/cm^3) [12]. Comparatively, the values obtained in the present study are lower than the threshold limit of 1.50 ppm (1.50 μg/cm^3) cobalt [12]. Cobalt is vital to man since it is part of vitamin B12, which is an essential vitamin and component for human health [23]. Cobalt can accumulate to toxic levels in the kidney, heart, liver, pancreas, skeleton and skeletal muscle. Cobalt can also produce tumours in animals and is a likely human carcinogen [24].

Levels of Zinc, Cadmium and Lead in Various Leaching Agents

Table 2 shows that the levels of zinc found (μg/cm^3) in various leaching agents studied ranged from not detected in all the leaching agents except in 1.00 % lactic acid, where a concentration of 119.20 μg/cm^3 zinc was determined. The experimental level of zinc is much greater than 2.00 μg/cm^3 permissible safe limit of zinc in water [25][26]. Zinc has the highest concentration among all the metals leached in the leaching agents studied. This might be because it is a common element found in soil, water, air and all forms of food. Zinc is an important element in human diet since it is needed in maintaining proper functions of the immune system. It is also useful for the normal brain activity and is fundamental in the growth and development of foetus. Its deficiency in the diet can be more detrimental to human health than too much of it [27]. Consumption of about 27.00 g of zinc per day can cause death.

The concentrations of cadmium determined in various leaching agents ranged from a 0.01 μg/cm^3 (found in 4.00 % acetic acid) to a 0.26 μg/cm^3 (determined in 1.00 % lactic acid). Cadmium values of 0.02, 0.03 and 0.05 μg/cm^3 were found to be in between the observed extreme values. The determined values are in good agreement with reported literature values of 0.01 to 0.28 mg/L (0.01 to 0.28 μg/cm^3) [18]. The observed cadmium values are much lower than 0.50 μg/cm^3 threshold limit established by NAFDAC [28]. The determined values are mostly greater than 0.01 μg/cm^3 permissible safe limit of cadmium in water [25][26]. Acute doses of 10.00 to 30.00 mg/kg/day of cadmium can cause severe gastrointestinal irritation, vomiting, diarrhea and excessive salivation [29].

The concentrations of lead (μg/cm^3) as determined in the various leaching agents placed in the self-produced ceramics investigated ranged from 0.04 (determined in orange juice) to 1.03 (assayed in boiled lipton tea). The experimental values are comparatively much lower than 5.00 μg/cm^3 safe limit of lead in water [15]. The toxic effect of lead is related to its accumulation in body organs like the brain and this can lead to poisoning referred to as Plumbism (Saturnism) which can also result into death among humans [30].

V. CONCLUSION

This research work was aimed at formulating heavy metals leaching resistant glazed ceramic food wares. Of all the leaching agents investigated, only lactic acid leached an appreciable level of zinc above its threshold value. Manganese was also leached by most of the leaching agents above its safe permissible limit. The levels of some heavy metals leached by other leaching agents investigated are generally below their safe permissible limits. Based on the present study, it is therefore evident that the use of most of the leaching agents in the self-produced or formulated ceramics will be safe. Ceramic wares of low levels of heavy metals leaching ability are therefore produced.

REFERENCES

[1] Eze, H. O. (2009). **"Introduction to Ceramic Technology"**. *Aku Graphic Press*, Chobo Uniport., Nigeria, pp. 20 – 30.

[2] Yet-Ming, C. and Karl, J. (1999). **"Fundamental Research Needs in Ceramics"**. *National Science Foundation*, Artington, pp. 1 – 54.

[3] Michael, W. B. (1997). **"Fundamentals of Ceramics"**. *The McGraw Hill Companies Inc.*, New York, pp. 1 – 19.

[4] Carter, C. B. and Norton, M. G. (2007). **"Ceramic Materials, Science and Engineering"**. *Springer Science and Business Media*, Berlin, Germany, pp. 20 – 21.

[5] Hight, S. C. (2001). Determination of Lead and Cadmium in Ceramic Ware Leach Solutions by Graphite Furnace Atomic Absorption Spect-roscopy: Method Development and Inter-Laboratory Trial. *Journal of Association of Official Analytical Chemists International*, **84**: 861 – 872.

[6] Demont, M., Boutakhrit, K., Fekete, V., Bolle, F. and Van Loco, J. (2012). Migration of 18 Trace Elements from Ceramic Food Contact Material: Influence of Pigment, pH, Nature of Acid and Temperature. *Food Chemical Toxi-cology*, **50**: 734 – 743.

[7] Omolaoye, J. A., Uzairu, A. and Gimba, C. E. (2010). Heavy Metals Assessment of Some Ceramic Products Imported into Nigeria from China. *Archieves of Applied Science Research*, **2**: 120 – 125.

[8] Hassan, U. F., Baba, H., Shibdawa, M. A., Mahmoud, A. A. and Ishaku, J. (2013). Assessment of Feed Quality Efficiency of Morin-ga oleifera Seeds. *Journal of Chemical Society of Nigeria*, **38** (2): 70 – 73.

[9] Henden, E., Cataloglu, R. and Aksuner, N. (2011). Determination of Arsenic Leaching from Glazed and Non-Glazed Turkish Tradition-al Earthenware. *Science of the Total Environment*, **409**: 2993 – 2996.

[10] Prajusha, V. V. (2013). **"Studies of Leaching of Metals from Food Ceramics"**. A Thesis submitted to the Department of Chemistry, University of Waikato, New Zealand.

[11] American Society for Testing Materials (2006). Standard Test Method for Lead and Cadmium Extracted from Glazed Ceramic Surfaces. C 398 – 34.

[12] Valadez-Vega, C., Zuniga-Perez, C., Quintanar-Gomez, S., Morales-Gonzalez, J.A., Madrigal-Santillan, E., Villagomez-Ibarra, J. R. *et al.* (2011). Lead, Cadmium and Cobalt (Pb, Cd and Co) Leaching of Glass-Clay Containers by pH Effect of Food. *International Journal of Molecular Sciences*, **12**: 2336 – 2350.

[13] Ajimal, M., Khan, A., Nomani, A. A. and Ahmed, S. (1997). Heavy Metals Leaching from Glazed Sur-faces. *Science Total Environment*, **207**: 49 – 54.

[14] Mohammed, N., Chin, Y. M. and Pok, F. W. (1995). Leaching of Lead from Local Ceramic Table Ware. *Food Chemistry*, **54**: 245 – 249.

[15] Singh, A., Sharma, R. K., Agrawal, M. and Marshall, F. M. (2010). Risk Assessment by Heavy Metal Toxicity through Contaminated Vegetables from Waste Water Irri-gated Area of Varanasi, India. *Tropical Ecology*, **51** (2S): 375 – 387.

[16] ATSDR (2005). Toxicological Profile for Nickel. Agency for Toxic Substances and Disease Reg-istry. G. A. Atlanta.

[17] Sheets, R. W. (1998). Release of Heavy Metals from European and Asian Porcelain Dinner Ware. *Science of the Total Environmnt*, **212**: 107 – 113.

[18] Aderemi, T. A., Adenuga, A. A., Oyekunle, J. A. O. and Ogunfowokan, A. O. (2017). High Level Leaching of Heavy Metals from Colourful Ceramic Fodd-wares: A potential Risk to Human. *Environmental Science and Pollu-tion Research International*, **24** (20): 17116 – 17126.

[19] World Health Organization (2004). Guidelines for Drinking-water Quality, Geneva. Retrieved on April, 2020 from: http://www.who.int/water_sanitation_health/publications/gdwq3rev/en/.

[20] Adelekan, B. and Abegunde, K. (2011). Heavy Metals Contamina-tion of Soil and Ground Water at Automobile Mechanic Village in Ibadan, Nigeria. *International Journal of the Physical Sciences*, **6**: 1045 – 1058.

[21] Gukas, H. J. and Datiri, Y. C. (2001). **"The Art of Pottery"**. *C. C. Communications*, Jos, Nigeria, pp. 3 – 10.

[22] Calkins, M. (2009). **"Materials for Sustainable Sites: A Complete Guide to the Evaluation"**. *John Wiley and Sons*, Hoboken, New Jersey, pp. 451.

[23] Bhagure, G. R. and Mirgane, S. R. (2011). Heavy Metal Contamina-tions in Groundwaters and Soils of Thane Region of Maharashtra, India. *Environmental Monitoring Assessment*, **173** (1 – 4): 643 - 652.

[24] Mendy, A., Gasana, J. and Vieira, E. R. (2012). Urinary Heavy Metal: An Associated Medical Condition in the US Adult Population. *Inter-national Journal of Environmental Health Research*, **22** (2): 105 – 108.

[25] FAO (2001). Cited in: Rilwan, M. Z., Adamu, H. M., Shibdawa, M. S. and Haladu, K. A. (2019). Deter-mination and Risk Assessment of Heavy Metals via Intake of *Allium cepa* from Wastewater used for Irrigation in Bauchi Suburb. *Chem-istry Research Journal*, **4** (2): 59 – 64.

[26] WHO (2001). Cited in: Rilwan, M. Z., Adamu, H. M., Shibdawa, M. S. and Haladu, K. A. (2019). Determination and Risk Assessm-ent of Heavy Metals via Intake of *Allium cepa* from Wastewater used for Irrigation in Bauchi Suburb. *Chemistry Research Journal*, **4** (2): 59 – 64.

[27] ATSDR (1994). Toxicological Profile for Zinc. US Department of Health and Human Services, Public Health Services, 205-93-0606.

[28] Ralph, W. S. (1997). Extraction of Lead, Cadmium and Zinc from Overglaze Decorations of Ceramic Dinner Wares by Acidic and Basic Food Substances. *The Science of the Total Environment*, **197** (1-3): 167 – 175.

[29] ATSDR (1999). Toxicological Profile for Cadmium. US Department of Health and Human Servi-ces, Public Health Service, 7440-47-3.

[30] Boadu, T. M. (2014). Heavy Metals Contaminations of Soil and Water at Agbogbloshie Scrap Market, Accra. Unpublished Project Report. Department of Theoretical and Applied Biology, Kwame Nkruwmah University of Science and Technology, pp. 1 – 59.