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Contamination of Foods by Migration of Some Elements from Plastics Packaging

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1. Introduction

There are various types of packaging material including paper, board, plastic, metal, glass, wood and other materials. Paper and board packaging accounted for the largest share of global packaging sales in 2003 with 39% of the total. Plastic packaging accounted for 30% (rigid and flexible plastics) of the market, with metal packaging accounting for 18% and glass packaging a further 7%. Other packaging products accounted for the remaining 6% of the market. Rigid plastics was the fastest growing sector of the market during the period 1999-2003. Around 70% of overall consumer packaging consumption is used for food and beverage packaging (WPO, 2008). Many different types of plastics are being used as packaging materials. The key components in plastic materials are polymers which are made of units of organic material, and one or more of large molecular weight can be formed as desired. Most polymers are petrochemical compounds with additive materials to give them properties of flexibility, elasticity and resistance to fracture and transparency to light (Oi-Wah & Siu-Kay, 2000; Al-Dayel et. al. 2009)

The final plastic material thus is a mix of polymer, additives, manufacturing aids, and side products from the complex polymerization process that were not intentionally added (Bradley & Coulier 2007).

Low density Polyethylene (LDPE) is used in preparation of most of the hot food packaging. LDPE has high flexibility, and can be affected by organic solvents. It has melting temperature of 110 ° C (wikipedia.org 2012).

High Density Polyethylene (HDPE), has the same uses as those of the low density, but it is much flexible and resistant to organic solvents and to high temperatures. It is used in manufacture of some household appliances, pipes and hoses. It is also used in food packaging which are subject to sterilization temperatures. HDPE is characterized by its ability to isolate the humidity, and its flexibility even at freezing temperature.

Different types of additives, such as antioxidants, stabilizers, lubricants, anti-static and anti-blocking agents, have been developed to improve the performance of polymeric packaging materials (Achilias 2007; *S*usan1992).

The role of food and beverage packaging as a source of contaminants have raised many concerns after the widespread use of such containers; packaging. Any substance (monomers

and other starting substances, additives, residues) which migrates from the packaging into the food is of concern if it could be harmful to the health (Donatella et. al., 2010; Grob et. al., 2006).

The migration of additives or contaminants from polymeric food packaging to food may be separated into three different, but inter-related, stages: diffusion within the polymer, solvation at the polymer food interface, and dispersion into bulk food (Oi-Wah & Siu-Kay, 2000). The migration has been showed to increase with fat content and storage temperature (Sanches, et al.,2007).

Antimony does not bioaccumulation, so exposure to naturally occurring antimony through food is very low. Antimony is present in food, including vegetables grown on antimony-contaminated soils, mostly in the low µg/kg wet weight range or less (WHO, 2003).

Antimony toxicity is dependent on the exposure dose, duration, route (breathing, eating, drinking, or skin contact), other chemical exposures, age, sex, nutritional status, family traits, life style, and state of health (Ross and Adrian, 2009). Chronic exposure to antimony in the air at levels of 9 mg/m^3 may exacerbate irritation of the eyes, skin, and lungs (Roper, 1992). Long-term inhalation of antimony can potentiate pneumoconiosis, altered electrocardiograms, stomach pain, diarrhea, vomiting, and stomach ulcers, results which were confirmed in laboratory animals (Roper, 1992). Although there were investigations of the effect of antimony in sudden infant death syndrome, current findings suggest no link. Long-term exposure in experimental animals has shown an increase in the hepatic malfunction and blood changes (ATSDR, 1992). It is not clear yet whether antimony is a human carcinogen. Occupational epidemiology could not confirm evidence of lung carcinogenicity caused by antimony as detected in female rats (Gerhardsson et al. 1982; Jones, 1994; Groth et. al.1986). Furthermore, because the experimental results were not uniform, animal lung carcinogenicity by antimony is still a matter for debate (Jones, 1994; Newton et. Al., 1994; Ross and Adrian, 2009).

This work is an attempt to evaluate the migration of elements from packaging materials into food stuffs. A severe contact condition between food and packaging materials has been created to evaluate the maximum potential migration of elements (Grob et. al.1999).

2. Experimental

2.1 Samples

Two types of polyethylene samples have been selected from the Saudi market. One is called thermal bag which is commonly used for hot food, and the other one is called food bag which is commonly used for cold and freezing food. Both of these types are made of liner low density polyethylene. The samples elemental concentrations have been obtained using neutron activation analysis (NAA).

2.2 Neutron Activation Analysis (NAA)

Samples were irradiated at the McMaster University Reactor in Hamilton, Ontario, Canada. It is a 2MW open pool research reactor.

The Gamma ray spectrum acquisition was carried out by the use of a high resolution intrinsic germanium detector.

Two portions of each sample were weighed into a plastic irradiation container. One portion is for short-lived isotope analysis (half life < 24 hours). This portion is about 4 grams weighed into a 7 ml volume container, and it was irradiated for about 60 seconds using a thermal neutron flux of approximately $6x10^{12}$ n/ cm^2.s . After a short decay period of 6 minutes, the gamma ray spectrum was acquired. The sample was then allowed to decay for further period of 24 hours.

The other portion was for longer lived isotope analysis (half life >5 days). Approximately 24 grams of the sample was weighed into a 40 ml container, and was irradiated for 20 minutes using thermal neutron flux of approximately $8x10^{12}$ n/ cm^2.s. After 3 days of decay, the sample was counted for 60 minutes. The sample was again allowed to decay for further period of 18 days and then counted for 2 hours. Results were calculated for each of the four spectra.

2.3 Quality assurance for NAA analysis

To assess the analytical process and make a comparative analysis, Standard Reference Material coal sample (SARM-18) from South African Bureau of Standards and a trace element in coal sample from the USA National Institute of Standards and Technology (NIST) (SRM 1632c) were analyzed in the same manner as other samples (Anderson & Cunningham, 2000; Wang & Sakanishi, 2004). Table (1) gives the comparison of the certified values and these obtained in this work for each reference materials. The results are generally in a good agreement except for Sr, Zr, Ba, Sb, Ba, Ca, Eu, Na, and As in one or other standard reference material samples.

Element in SARM 18				NIST 1632c		
mg/kg (%) (µg/Kg)	This work	RSD	Range@	This work	RSD	Certified Value
Al (%)	1.34	*3*	*2.54-2.61*	0.94	*4*	0.915±0.0137
Sb	0.30	*5*	*0.3**	0.22	*5*	0.461±0.029
As	0.52	*4*		3.5	*3*	6.18±0.27
Ba	91	*6*	*71-82*	70	*5*	41.1±1.6
Br	3.5	*3*	*3**	20.9	*3*	18.7±0.4
Cd	0.7	*20*		<0.3		0.072±0.007
Ca (%)	0.138	*4*	*0.17-0.19*	0.21	*4*	0.145±0.03
Ce	20.6	*3*	*21-24*	8.2	*3*	11.9±0.2
Cs	1.17	*4*	*1**	0.473	*5*	0.594±0.010
Cl	54	*5*		1130	*3*	
Cr	17.3	*3*	*14-18*	10.4	*3*	13.73±0.20
Co	7.5	*3*	*5.5-7.2*	3.4	*3*	3.48±0.2
Cu	4.9	*9*		6.7	*6*	6.01±0.25
Dy	1.87	*3*		0.57	*3*	

Eu	0.31	*3*	*0.3**	0.17	*3*	0.124±0.003
Ga	7.1	*5*	*8**	2.6	*6*	3*
Au (µg/Kg)	0.65	*23*		<0.5		
Hf	1.9	*3*	*1.7-1.9*	0.44	*4*	0.585±0.010
In	0.028	*12*		0.02	*11*	
I	1.0	*11*		1.5	*6*	
Ir (µg/Kg)	<0.7			<0.5		
Fe (%)	0.22	3	0.28-0.29	0.77	*3*	0.735±0.011
La	10.6	*3*	*9.0-13*	4.7	*3*	
Lu	0.207	*3*		0.055	*4*	
Mg (%)	0.056	6.5	0.1-0.11	0.039	*8*	0.0384±0.0032
Mn	21.5	3.2	21-23	13.5	*3*	13.04±0.53
Hg	<0.017		0.04*	0.0802	*8*	0.0938±0.0037
Mo	1.13	*5*		0.693	*9*	0.8*
Nd	7.5	*5*		3.1	*5*	
Ni	<10			<6.8		9.32±0.51
K (%)	0.12	*4*	*0.140-0.150*	0.081	*3*	0.11±0.0041
Rb	8.3	*6*	*6.7-9.5*	5.4	*6*	7.52±0.33
Sm	1.7	*5*	*1.9-2.2*	0.72	*5*	1.078±0.028
Sc	5.1	*3*	*4.0-4.7*	2.0	*3*	2.905±0.036
Se	0.2	*31*		1.1	*5*	1.326±0.071
Ag	<0.2			<0.1		
Na (%)	0.013	*3*		0.052	*3*	0.03±0.005
Sr	31	*15*	*42-45*	109	*4*	63.8±1.4
Ta	0.36	*3*	*0.3**	0.14	*3*	
Te	<0.9			<0.3		0.05*
Tb	0.29	*6*	*0.3**	0.10	*9*	
Th	3.8	*3*	*3.0-4.3*	1.2	*3*	
Sn	<11		*1**	<7		1*
Ti	0.067	*3*	*0.111-0.116*	0.047	*4*	0.052±0.003
W	1.5	*3*	*2**	0.4	*6*	
U	1.9	*3*	*1.5-2.0*	0.40	*4*	0.513±0.012
V	22	*3*	*21-25*	15	*4*	23.72±0.51
Yb	1.26	*3*		0.34	*4*	
Zn	6	*10*		10	*6*	12.1±1.3
Zr	20	*19*	*62-71*	<15		16*

@ Range refers to 95% confidence limits.
* Uncertified value.
RSD: Relative Standard Deviation.

Table 1. Comparison of elemental concentration of the standard reference material (SARM18 and NIST 1632c) in this work and their certified values.

2.4 Study of elemental migration

Four types of food stuffs were examined, water, 30% ethanol, olive oil and 5% acetic acid. The aim was to study the immigration of Al, Sb, Cu, Mg, Ti and Zn.

To assess the maximum possible value of elements migration, the samples were prepared under the influence of severe contact conditions between the bag materials and the food stuff. This influence have been created by mixing the bag material, in the form of powder, with the food stuffs and exposing the mixture to high temperature (80-100 °C). Such a contact case cannot be exist in normal human use, but it can show the maximum potential migration.

2.5 Experimental arrangement and ICP-MS analysis

Four samples were prepared in four flasks. Each flask contained a mix of 2.5 g of the sample powder and 25 ml of one of the four food stuffs. The flasks were placed in a shaker at 160 rpm stirring rate at temperature of 100 ^{o}C (80 ^{o}C for ethanol case).

The samples were elementally analyzed using a Perkin-Elmer Sciex Instruments multi-element ICP-MS spectrometer, type ELAN6100, equipped with a standard torch, cross flow nebulizer and Ni sampler and skimmer cones.

2.6 Quality assurance for ICP-MS analysis

To assess of the analytical process and make a comparative analysis, Standard Reference Material (SRM), Nist-1640 Natural Water purchased from the National Institute of Standards and Technology (NIST), USA was analyzed in the same manner as all other samples. Table 2 compare the certified values with those obtained in this work. The results are generally in good agreement with the certified values.

3. Results and discussion

The elemental concentrations in the samples and the corresponding concentrations migrated to food stuffs are shown in table 3. The results show that the Antimony (Sb) has the highest migrated concentration ratio (37 %), in the acetic acid, followed by Zink (Zn) with 22 % , Magnesium with 17 %, in ethanol and Titanium (Ti) with 12%, in olive oil. The lowest migration ratio was that of Aluminum (Al) which was only 0.012 %, in ethanol. Table 3: Concentration of elements in bags materials and migrated to food stuffs.

4. Conclusion

The NAA and the ICP-MS analytical methods used in this work gives good results. These results were confirmed by the analysis of the standard reference materials as shown in tables 1 and 2.

Migration of substances from plastic packaging materials into food stuffs is clearly measured in the conditions which have been created to represent the worst case of contact between packaging material and food stuff. This suggests that a further studies on migration of substances from plastic packaging into food stuffs in the normal conditions should tack place. The results also suggest expected a seriousness harmful effect of using wrong plastic packaging material for heating foods in microwave ovens.

Elements	This Work		Certified values	
	Concentration in ppb	RSD	Concentration in ppb	RSD
Li	49.8	1.50	50.7	2.76
Be	36.4	4.95	34.94	1.17
B	264	8.60	301.1	2.03
Na	27800	0.34	29530	1.05
Mg	5800	0.50	5819	0.96
Al	47.4	0.32	52	2.88
K	899	0.93	994	2.72
Ca	6990	0.97	7045	1.26
V	11.9	1.16	12.99	2.85
Cr	37.1	0.45	38.6	4.15
Mn	117	0.46	121.5	0.91
Fe	35.2	15.65	34.3	4.66
Co	19	0.50	20.28	1.53
Ni	27	1.19	27.4	2.92
Cu	89.2	0.19	85.2	1.41
Zn	64.7	1.36	53.2	2.08
Ga	0.198	30.20		
As	27.3	1.59	26.67	1.54
Se	23.7	3.95	21.96	2.32
Rb	2.22	1.65	2	1
Sr	108	0.40	124.2	0.56
Mo	43.5	2.04	46.75	0.56
Ag	6.48	0.39	7.62	3.28
Cd	21.9	2.14	22.79	4.21
Sb	12.6	2.85	13.79	1.46
Te	0.328	26.89		
Ba	139	0.96	148	1.48
Tl	0.146	42.60	<0.1*	
Pb	26.8	0.79	27.89	0.50
Bi	0.143	40.49		
U	0.834	7.41		

RSD: Relative Standard Deviation.

Table 2. Comparison of elemental concentration of the reference material (NIST 1640) in this work with the certified values.

Element	Concentration of elements in bags materials	concentation of element migrated to food material and the migration percentage							
	ppm	ppm	%	ppm	%	Ppm	%	ppm	%
Thermal bag		Water	Migration	Ethanol	Migration	Olive Oil	Migration	Acetic Acid	Migration
Al	100	0.0147	0.15	0.0127	0.13	0.073	0.7	0.181	1.8
Sb	0.362	0.00307	8.48	0.00372	10.28	0.000631	1.7	0.00824	22.8
Cu	2.7	0.00668	2.47	0.0076	2.81	0.00224	0.8	Not Detect	
Mg	246	0.0461	0.19	0.553	2.25	0.014	0.1	Not Detect	
Ti	6.6	0.0101	1.53	Below Detection Limit		0.00665	1.0	Below Detection Limit	
Zn	98	0.0175	0.18	0.0652	0.67	0.0206	0.2	0.982	10.0
Food bag									
Al	768	0.0968	0.13	0.0247	0.03	0.244	0.3	0.413	0.5
Sb	0.283	0.00372	13.14	0.00391	13.82	0.00066	2.3	0.0105	37.1
Cu	4.75	0.00638	1.34	0.0154	3.24	0.00661	1.4	Not Detect	
Mg	32	0.0274	0.86	0.555	17.34	0.0374	1.2	Not Detect	
Ti	4.4	0.0115	2.61			0.055	12.5	0.000447	0.1
Zn	52	0.026	0.50	0.124	2.38	0.163	3.1	1.15	22.1

Table 3. Concentration of elements in bags materials and migrated to food stuffs.

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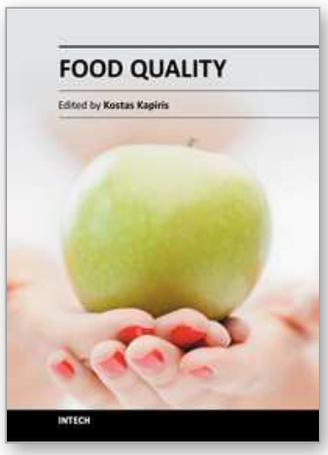

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The book discusses the novel scientific approaches for the improvement of the food quality and offers food scientists valuable assistance for the future. The detailed methodologies and their practical applications could serve as a fundamental reference work for the industry and a requisite guide for the research worker, food scientist and food analyst. It will serve as a valuable tool for the analysts improving their knowledge with new scientific data for quality evaluation. Two case study chapters provide data on the improvement of food quality in marine and land organisms in the natural environment.

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