

INVESTIGATION OF PLASTICIZERS AND NON-INTENTIONALLY ADDED SUBSTANCES (NIAS) FROM THE YOGHURT BOTTLES AND DISPOSABLE PLASTIC CUPS

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Abstract

The aim of this research work is to investigate the presence and comparative amount of plasticizers and non-intentionally added substances (NIAS) from the yoghurt bottles and disposable plastic cups. The types of polymer were identified by Fourier Transform Infrared (FT IR) spectrophotometer and plasticizers and NIAS were identified by Gas Chromatography-Mass Spectrometry (GC-MS). The type of polymer of the yoghurt bottle samples was high density polyethylene (HDPE) and that of disposable plastic cups was polypropylene (PP). The plasticizers and NIAS mostly found in both types of polymers were Fatty Acid Methyl Ester (FAME) compounds and long chain hydrocarbon compounds. The other compounds found from these types of food containers were aromatic hydrocarbon compounds, ether compound, ketone, bicyclic compounds, epoxy compound and sulphur containing compounds. Overall migration percents were evaluated by using three solvents, chloroform, water and olive oil. The migration percents of HDPE polymer were found to be 0.11 %, 0.81 % and 0.06 % and those of PP polymer were found to be 0.34%, 1.35 % and 0.07% in water, chloroform and oil, respectively. Elemental analysis of the samples by Wavelength Dispersive X-ray Fluorescence (WDXRF) showed that the common elements found in both types of polymer samples were Ti, Zn, Ca, Fe, Cl, Si, K and Al.

Keywords: plasticizers, NIAS, high density polyethylene, polypropylene, migration percents

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Introduction

Food packaging and disposable plastic wares are an important part of the food industry for preservation of food as well as mechanical support and protection in transit. A range of different polymers are used in food packaging with their compatible plasticizer and other additives such as fungicides, flame retardants, stabilizers and antioxidants that applied on the specific characteristics of the food products, the storage conditions, and the shelf life. Among various types of food packaging polymers, the polymer used in yoghurt bottles and disposable plastic cups are most interesting areas by consumers due to their wide range of applications. Even the plastic additives are inert and non-toxic; they can interact with food and become harmful in large doses (Kerry *et al.*, 2006). A number of factors can influence the migration of plasticizers, other additives and degradation products of polymer to food, including the chemical properties of the compound, the contact surface and the type of plastic material, the character of food, the packaging temperatures, the heat or sterilization treatment, and the storage time of the product. The presence of plastic additive or plasticizer at high concentration may trigger health problems.

Besides the plastic additives some non-intentionally added substances (NIAS) may be present in polymer products and they can also migrate into food and drinks. The presence of NIAS has been suggested as the source of toxicological effects (Leivadara *et al.*, 2008). Due to their leaching potential and widespread use, independent research, such as recording current materials used in common supermarket products, was conducted to obtain a better understanding of what polymers are currently used in food packaging. And then, it was then important to understand the impact on both society and the consumer by researching possible hazards in the use of plastics food containers to assess whether there are any public health and safety risks by determining a range of chemicals having potential to migrate from food packaging into foods.

The aim of this research work is to investigate the presence of plasticizers and NIAS and relative amount of those plasticizers and NIAS from the yoghurt bottles and disposable plastic cups and relative abundance of elements.

Materials and Methods

Sample Collection

The Yoghurt bottle (150 mL, imported brand) and disposable plastic cup (150 mL, 7 cm (diameter) x 8.5 cm (height) new container) were collected from the market of Pyin Oo Lwin Township, Mandalay Region. The samples were cleaned by distilled water and cut into small pieces and stored for further analysis.

Identification of Types of Polymer

The types of polymer of the selected samples were identified by FT IR by using modified KBr method. In this method, the samples were cut into disc and inserted in the KBr holder and pressed by hydraulic press. The prepared discs were analyzed in the mid-IR region. The spectra obtained were compared with the library spectra of specialized data-base software. These measurements were done at the Pharmacology Research Division, Department of Medical Research (Pyin Oo Lwin Branch).

Extraction of Polymer Components from the Polymer Samples

In this experiment, three selected solvents were used to extract the polymer components from both samples. The three solvents were chloroform, distilled water and olive oil. The exact weight (1.0 g) of each sample was refluxed with 30 mL of solvents, chloroform and distilled water at refluxing temperature for 3 h and the olive oil at 95- 110 °C for half hour. Then, each extract was filtered through the filter paper and the weights of residual extracts were also recorded. The filtrates were analyzed by GC-MS (GCMS-QP 2010).

Identification of Polymer Components from the Extracts

Each extract was subjected to measure the GC-MS in order to identify all the components present in the extract. The chloroform extract and distilled water extract were measured directly without measuring the solvents by setting as blank. But in the case of olive oil extract, olive oil was first measured for GC-MS to set it as blank. Then, all extracts were measured under specified conditions. GC was equipped with the normal septum. The GC conditions were as follows: GCMS-QP 2020 (Shimadzu, Kyoto, Japan)

equipped with a methyl silicone coated fused-silica capillary DB-5MS (0.25 mm i.d. x 30 m, 0.25 μm film thickness) column were used. The oven temperature was maintained at 50 $^{\circ}\text{C}$ for the first 1 min and then ramped up at 10 $^{\circ}\text{C min}^{-1}$ to 270 $^{\circ}\text{C}$ where it was held for 22 min. The injection port and interface were kept at 260 $^{\circ}\text{C}$. Helium (100.0 kPa) served as the carrier gas. The injection mode was splitless, the sampling time was 3 min, and the injection volume was 1 μL . An auto-injector and auto-sampler AOC-20 i+s(Shimadzu,Japan) were used. The MS conditions were as follows: ionization mode, electron impact and detection voltage at 1.3 kV. The analysis was carried out at the Pharmacology Research Division, Department of Medical Research (Pyin Oo Lwin Branch).

Elemental Analysis of Polymer Samples by WDXRF

Relative abundance of elements of the polymer samples were determined by WDXRF (Wavelength Dispersive X-ray Fluorescence) at Department of Research and Innovation on Science, Yangon. In this analysis the sample powders were used.

Results and Discussion

Identification of Types of Polymer Samples by FT IR

In this FT IR analysis, Raman of Basic Monomers and Polymers database library software was applied for polymer identification.

The FT IR spectrum of Yoghurt bottle sample is shown in Figure 1 and the compared FT IR spectrum of Yoghurt bottle sample with library software are shown in Figure 2. According to the FT IR spectra, the Yoghurt sample was confirmed as a type of polyethylene polymer. All of the absorption maxima of the individual peaks in the sample are consistent with those of library spectrum of polyethylene polymer. It was found that asymmetric and symmetric stretching vibrations of C- H appeared at 2920 cm^{-1} and 2850 cm^{-1} respectively. Moreover, CH_2 bending at 1470 cm^{-1} and CH_2 rocking at 730 cm^{-1} (Jung *et al.*, 2018) were also observed in FT IR spectrum of Yoghurt bottle sample.

The FT IR spectrum of the disposable cup sample is shown in Figure 3 and the compared FT IR spectrum of the disposable cup sample with library software is shown in Figure 4. Based on the observed absorption maxima of the sample and those of library spectrum, the major peaks are found relatively in the same position and the library software also showed that the disposable cup sample was polypropylene sample. Asymmetric and symmetric stretching vibrations of C-H appeared at 2950 cm^{-1} and 2830 cm^{-1} , respectively. The peaks at 1450 cm^{-1} and 1370 cm^{-1} are due to CH_2 bending and CH_3 bending respectively. The peaks due to CH_3 rocking appeared at 1150 cm^{-1} , 1000 cm^{-1} and 970 cm^{-1} and CH_2 rocking appeared at 830 cm^{-1} and 811 cm^{-1} (Jung *et al.*, 2018).

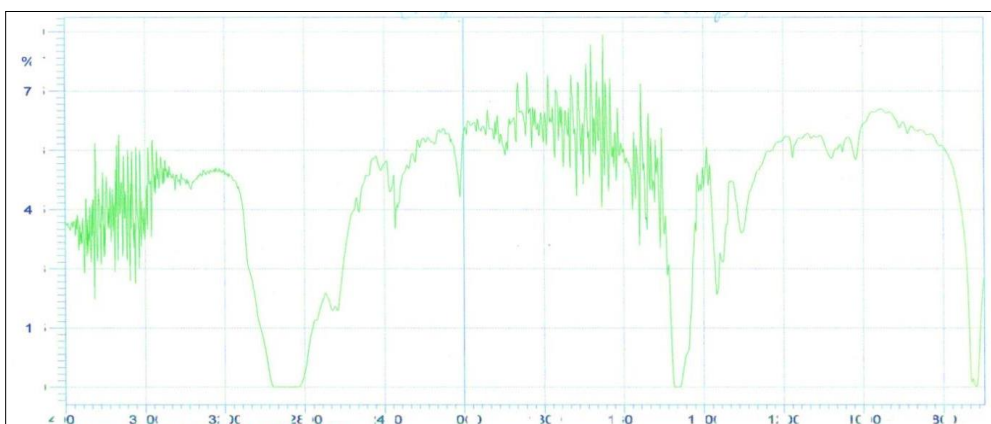


Figure 1: FT IR spectrum of Yoghurt bottle sample

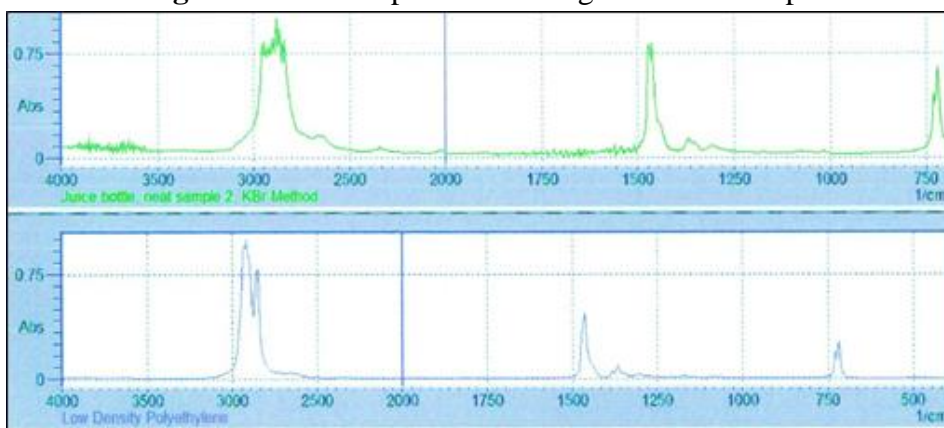


Figure 2: Comparison of FT IR spectra of Yoghurt bottle sample and library spectral data

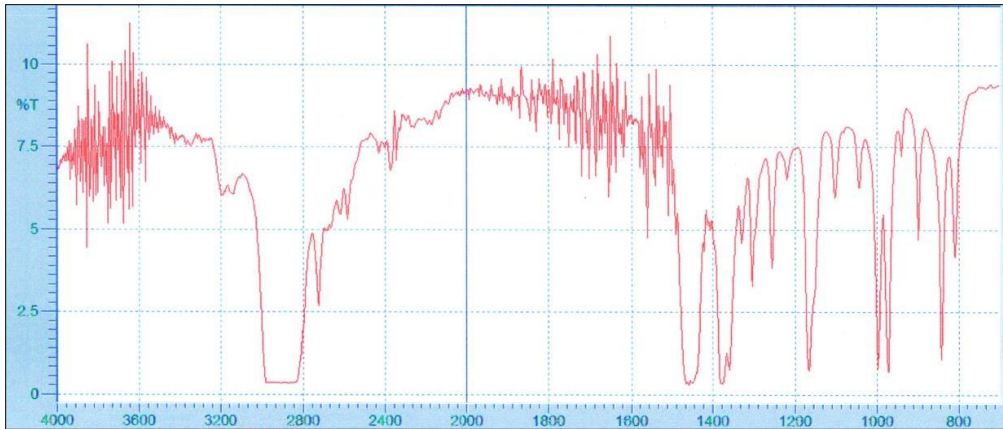


Figure 3: FT IR spectrum of disposable plastic cup sample

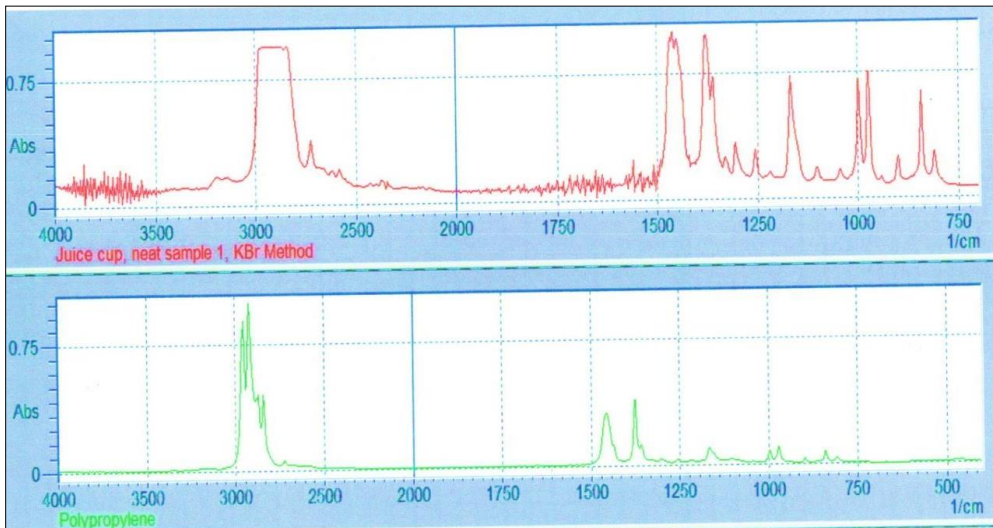


Figure 4: Comparison of FT IR spectra of disposable plastic cup sample and library spectral data

Migration Percents of High Density Polyethylene (HDPE) and Polypropylene (PP) Polymers

In the extraction of polymer components from the confirmed type of polymer samples, the migration percent of polymer constituents were directly calculated based upon the water extract and chloroform extract. But the migration percent into the oil was calculated by using 95 % ethanol

(Crompton, 2007) due to the fact that both of the polymer samples were partially dissolved into the oil during extraction. The migration percents of HDPE polymer into water, chloroform and oil were found as 0.11 %, 0.81 % and 0.06 % while those of PP polymer into water, chloroform and oil were 0.34% ,1.35% and 0.07 %, respectively (Table 1).

Table 1: Migration Percents of HDPE and PP Polymers

No.	Type of Polymer	Migration percent (%)		
		Water	Chloroform	Olive Oil
1.	HDPE	0.11	0.81	0.06
2.	PP	0.34	1.35	0.07

Identification of Polymer Components by GC-MS

GC/MS-QP 2020 was applied for identification of plasticizers, NIAS and other extractable matters from the polymer samples. The compounds found in the three different extracts were compared with mass spectra of compounds in National Institute of Standard and Technology (NIST) libraries which can be applied as data-base software.

Identification of polymer components from the CHCl₃ extract of HDPE polymer

A total of twelve compounds were found in the gas chromatogram of chloroform extract of polyethylene polymer that can be divided into three main groups (Figures 5 and 6 and Table 2), namely: (1) aromatic hydrocarbon compound (2) long chain hydrocarbon compound and (3) fatty alcohol and methyl ester of long chain fatty acid(both saturated and unsaturated).

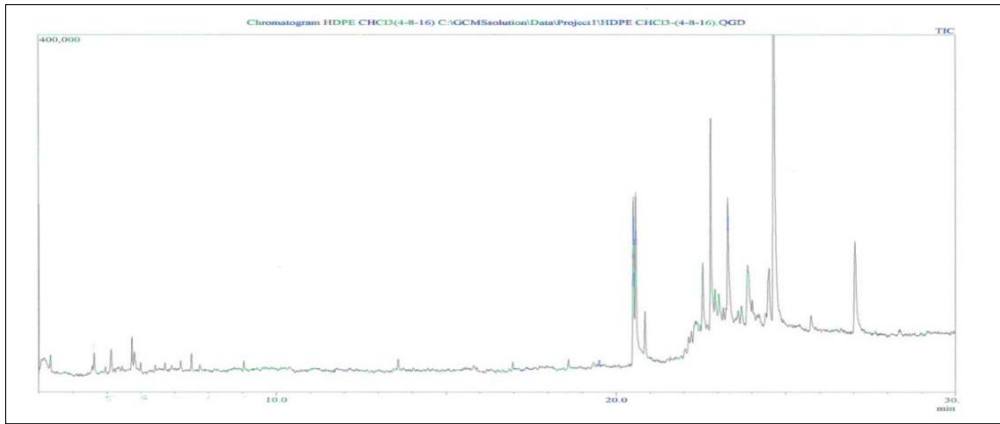


Figure 5: Gas chromatogram of CHCl_3 extract of HDPE sample

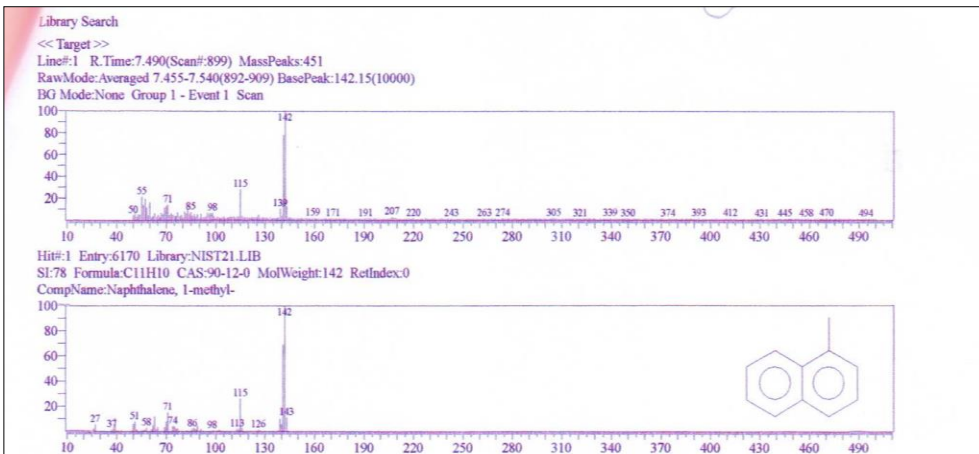


Figure 6: Mass spectrum of compound (retention time 7.49 min) from CHCl_3 extract

Long chain hydrocarbon found in the chloroform extract of polyethylene polymer was dotriacontane, $\text{C}_{32}\text{H}_{66}$. This compound is a normal alkane present in a typical middle range wax. This compound is used in polymer additives for diffusivity purpose. This type of long chain hydrocarbons compounds are also used for flexibility of polymer as the plasticizer or external lubricant in polymerization process (Fulmer,2000). Some of the migrant compounds in the chloroform extract are likely to be the oligomer components which come from the degradation product of polymer backbone. Therefore, this type of migrant compounds may be intentionally

added plasticizer or oligomer migrant compound. But, its relative percent area is very little (0.72%) among all of additives. Sometime this type of compound also occurs in some food such as butter, vinegar, and coffee for aroma carrier when taken into body serve to nourish and supply body heat (Dorland, 2007). Therefore, the health problem due to this compound is not considerable.

The fatty alcohol in chloroform extract was 2-hexadecanol. The fatty alcohols are obtained from the natural fats and oil or petrochemical sources. They are also used in the polymer production as the internal lubricant (semi-soluble plasticizer). Typical lubricants are fatty alcohols (C₁₂–C₂₂) or fatty acids (C₁₄–C₁₈). Therefore, these compounds might be leached out as product of polymeric lubricant, plasticizer.

Table 2: Identification of Polymer Components from the Chloroform Extract of HDPE Polymer

No.	Name of Compound	Retention time (min)	Base m/z	SI	A%
1.	1,2,4,5-tetramethyl Benzene	5.12	119.10	78	1.49
2.	Naphthalene	5.73	128.10	94	1.63
3.	1-ethyl-4-(1-methyl ethyl)- Benzene	5.80	133.15	82	0.43
4.	2-methyl Naphthalene	7.49	142.15	78	0.33
5.	Dotriacontane	13.58	57.10	84	0.72
6.	2-Hexadecanol	16.97	57.10	80	0.34
7.	9,12-Octadecadienoic acid methyl ester	20.50	67.05	95	8.80
8.	Methyl hexadecadienoate	20.57	55.10	89	7.87
9.	Eicosanoic acid methyl ester	22.79	74.05	84	10.75
10.	6,9,12,15-Docosatetra-enoic acid, methyl ester	23.30	93.10	82	6.87
11.	20-methyl-heneicosanoic acid methyl ester	24.64	74.05	92	45.62
12.	Heneicosanoic acid methyl ester	27.04	74.05	78	7.14

m/z- mass to charge ratio, SI- similarity index, A %-relative area percent

Six methyl esters of long chain fatty acid found in chloroform extract of PE polymer were 9,12-octadecadienoic acid methyl ester, methyl hexadecadienoate, eicosanoic acid methyl ester, etc.. All of these compounds are biodiesel components. According to chromatogram, these fatty acid methyl ester (FAME) compounds are most abundance compared to all of additives found. Among all FAME compounds, heneicosanoic acid methyl

ester, a type of saturated fatty acid ester is the highest in relative content. The migration of these compounds may have a worse effect on cholesterol level because “low in saturated fat” and “low in cholesterol”.

Then, fatty acids are frequently used in food packaging products as antistatic agents and ester of fatty acid, fatty acid methyl ester (FAME) can be formed trans-esterification of fatty acid with methanol. Sometime, these FAME compounds are usually come from vegetables oil and used in polyethylene production as lubricants to ensure proper mold release, processing aid (Fulmer, 2000). Therefore, these substances are assumed to be the intentionally added substances.

The other leachable compounds found from the chloroform solvent are aromatic hydrocarbon compound, 1,2,4,5-tetramethyl benzene, naphthalene, 1-ethyl-4-(1-methyl ethyl)- benzene, 1-methyl naphthalene and their relative percents of total chromatogram are 1.49%, 1.63%, 0.43% and 0.33% respectively. 1,2,4,5-tetramethyl Benzene is used for curing agents, adhesives, coating materials or cross-linking agent in polymer production. The LD₅₀ value of this compound is 6989 mg/kg (oral, rat) and thus it is not listed as a carcinogen by any association such as IARC (2002) .

Naphthalene is used as a secondary plasticizer in plastic and resin and also in fuels and dyes. It is usually made from crude oil or coal tar. This compound had been defined a possibly human carcinogenic compound based on the results of animal studies (IARC,2002). A large amount of naphthalene may damage or destroy some of red blood cells and can cause hemolytic anemia (PubChem, 2005). 1- Methyl naphthalenes are also used to make dyes, and resins. Naphthalene and naphthalene related compounds are same toxic manner according to ATSDR, (2005). Therefore, it can be concluded that these aromatic hydrocarbon compounds are assumed to be intentionally added substances that are not appropriate for human health. But their percent migrations found in this study were very low.

Identification of polymer components from the water extract of polyethylene polymer

In the gas chromatogram of water extract of polyethylene polymer, only five small speaks were observed (Figure 7). According to library mass

spectral analysis, most of the components in water extract are fatty acid methyl ester (FAME) compounds and only one component is one kind of medium chain aldehyde, nonanal (Figure 8 and Table 3). Nonanal is used as processing aid in industrial and lubricants and greases for commercial usage. Most aldehyde compounds are relatively reactive compound and has significant genotoxic effect at higher concentrations and mutagenicity for mammalian cells was observed with concentrations of 0.1-0.3 mM (Pub Chem, 2005). But relative abundance of this compound is very small and the content is at about 5.44% (of total migration percent into water, 0.11 %) compared to other components.

These FAME compounds in water extract were fatty acid derivatives of natural sources, such as vegetable oil, wax or animals' fat. But, the molecular ion peaks of every compound found from these extract are very small intensities when compared to others two extracts, therefore, the relative concentrations of migrant compound will be very low levels. So, the adverse health effect due to the presence of these compounds will be very low. From this finding, the food containers of PE polymer were appropriate for watery food types.

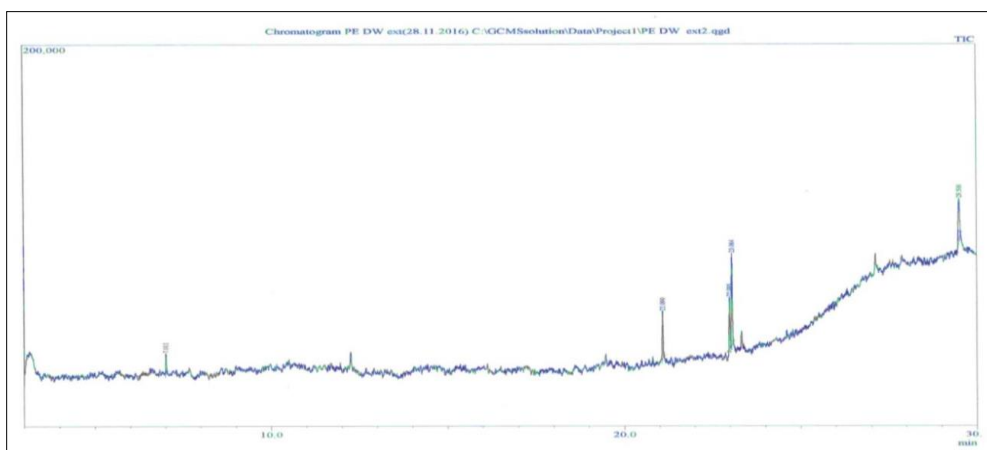


Figure 7: Gas chromatogram of water extract of HDPE sample

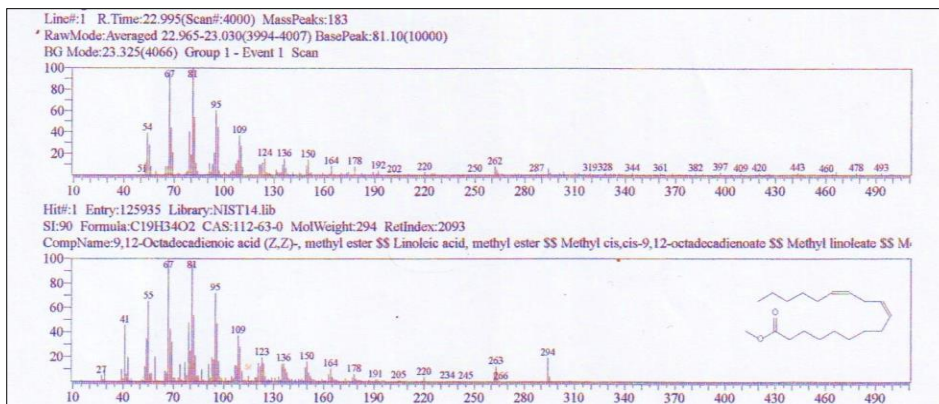


Figure 8: Mass spectrum of compound (retention time 22.99 min) from water extract of HDPE sample

Table 3: Identification of Polymer Components from the Water Extract of HDPE Polymer

No.	Name of Compound	Retention time (min)	Base m/z	SI	A%
1.	Nonanal	7.01	57.10	86	5.44
2.	Palmitic acid, methyl ester	21.09	74.05	92	16.45
3.	Linoleic acid, methyl ester	22.99	67.05	90	17.49
4.	9-octadecenoic acid(oleic acid), methyl ester	23.06	55.05	92	34.33
5.	Lignoceric acid, methyl ester	29.50	74.05	83	26.30

m/z - mass to charge ratio, SI - similarity index, A %-relative area percent

Identification of polymer components from the oil extract of HDPE polymer

The polymer components found in the oil extract of polyethylene polymer can be categorized into five main groups (Figures 9 and 10 and Table 4) namely: (1) long chain fatty alcohol, aldehydes and acids, (2) bicyclic compound (2-vinylbicyclo[2,1,1] hexan-2-ol), (3) epoxy compound, (4) thio compounds and (5) ether compound.

The usage of long chain fatty compounds in polymer production and their health effects were discussed in the previous section. Very long-chain fatty alcohols, fatty aldehyde, and fatty acid are reversibly inter-converted in a fatty alcohol cycle (Lunn and Theobald, 2006). Therefore, these compounds might be either intentionally added substances or inter-converted compounds of fatty alcohols.

The bicyclic compound found in the oil extract of polyethylene polymer is 2-Vinylbicyclo [2,1,1] hexan-2-ol. The various compounds that contain the vinyl (C₂H₃) group are used to make plastics, fabrics, phonograph recorder and paints for get tough, flexible, shiny plastics. The exact safety information of this compound is not available at the present although 4-Vinylcyclohexene is classified as a group2B (possible carcinogen) (IARC, 2002).

Another potential migrant compound found from the oil extract of PE polymer is epoxy compound, 2,3-epoxy-1-dodecyloxy propane. In the production of food packaging polymer, modified vegetable oil, alkyl aromatic sulfonate, polyol ester and cyclohexane diacid ester are also used as alternative plasticizers (Singh *et al.*, 2012). Epoxidised vegetable oil such as epoxidised soybean oil (ESBO) is used in a range of plastic, as a green plasticizer instead of phthalate plasticizer in food packaging materials (Hammarling *et al.*, 1998). Generally, ESBO is also used in various food-packaging plastics as heat stabilizers, and also as lubricants and plasticizers (Boussoum *et al.*, 2006). Therefore, the epoxy compound found in the oil extracts may be intentionally added substances.

The epoxidized oil and their derivatives are similarity in physicochemical properties, and absorbed and metabolized manner also similar to vegetable oils. There is no evidence concern with these compound groups to be carcinogenic, genotoxic or to have reproductive or developmental toxicity (OECD, 2006). It has been defined tolerable daily intake (TDI) for 1 mg/kg body weight and does not exceed 0.2 mg/kg from food package by the European Food Safety Authority (EFSA, 2006).

The thio compounds found in the oil extract of polyethylene polymer are N-(2-methyl-3-oxobutyl)-S-methyl dithiocarbamate and 3-methyl-4-(phenylthio)-2-prop-2-enyl-2,5-dihydro-thiophene 1, 1-dioxide. Actually,

dithiocarbamate compounds are one kind of pesticide which is used in farming of edible fruits that can produce thyroid cancer(USEPA, 2001). It has been shown to cause reproductive and birth defects in laboratory animals in long-term exposure. But, sulfur-containing antioxidants (such as dialkyl thiocarbamate, distearyl thiodipropionate) are added to various polymers (including PE) with phenolic antioxidants to get synergistic interaction between additives. This blend of phenolic antioxidants and divalent sulfur compound provides excellent combination for improving a polymer's long-term thermal stability.

Ether compound found in these extract is di-n-decyl ether, which may come from impurities of engineering machine or recycling process because polyether ether ketone polymers are used in engineering bearing (ball and roller) applications. Therefore, this type of migrant compounds may be non-intentionally added substances.

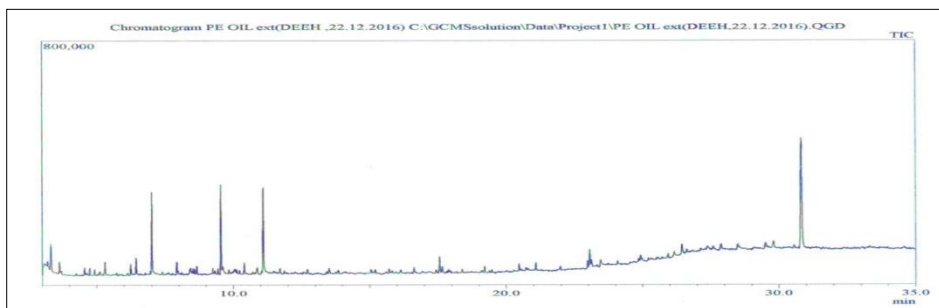


Figure 9: Gas chromatogram of oil extract of PE sample

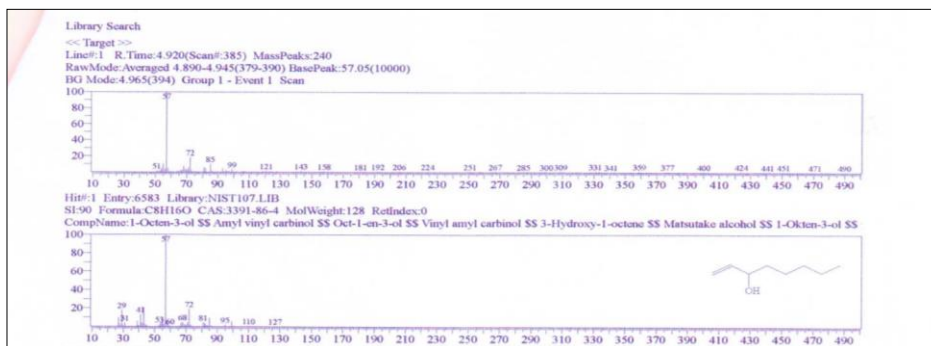


Figure 10: Mass spectrum of compound (retention time 4.92min) from oil extract of PE sample

Table 4: Identification of Polymer Components from Oil Extract of Polyethylene Polymer

No.	Name of Compound	Retention time(min)	Base m/z	SI	A%
1.	2-Vinylbicyclo[2,1,1] hexan-2-ol	3.64	91.05	92	1.19
2.	(E)-2-Heptenal	4.55	83.05	88	0.80
3.	1-Octan-3-ol	4.92	57.05	80	0.65
4.	Octanal	5.31	56.05	83	1.48
5.	(E)-2-Decenal	6.24	91.10	79	1.61
6.	E-11,13-Tetradecadiene-1-ol	6.43	56.05	85	1.81
7.	Nonenal	7.02	57.05	95	8.77
8.	2-hexyl Cyclopropanacetic Acid	8.43	58.05	80	1.20
9.	2,3-epoxy-1-dodecyloxy propane	8.58	57.05	81	0.48
10.	n-Decenal	8.66	57.05	83	0.91
11.	3-hydroxy Dodecanoic acid	9.25	71.10	79	0.54
12.	N-(2-methyl-3-oxobutyl)-S-methyl Dithiocarbamate	9.44	85.05	80	0.72
13.	(E)-2-Decenal	9.54	70.05	93	9.78
14.	9-Oxa bicyclo[6,1,0] nonan-4-ol	9.84	57.05	80	0.51
15.	di-n-Decyl ether	10.13	57.10	81	0.29
16.	(E,E)-2,4-Dodecadial	10.40	81.05	82	1.33
17.	(E)-2-Tridecenal	11.10	70.05	90	10.43
18.	HeptadecanoicacidHeptadecyl ester	13.85	57.10	79	0.67
19.	3-methyl-4-(phenylthio)-2-prop-2- enyl-2,5-dihydrothiophene1,1-dioxide	15.05	57.05	79	0.55
20.	12-Heptadecyn-1-ol	16.62	96.10	79	0.75
21.	Z-5-Nonadecene	17.56	69.05	86	2.48
22.	3-hydroxy-Dodecanoic acid	18.39	57.05	80	0.39
23.	Z,Z-8,10-Hexadeca- diene-1-ol	20.42	71.10	83	0.87
24.	(Z,Z)-9,12-Octadecadienoic Acid, methyl ester	22.99	67.10	91	0.84
25.	Triolein	23.06	55.05	83	2.18

m/z - mass to charge ratio, SI - similarity index, A %- relative area percent

Identification of Polymer Components from the CHCl₃ Extract of PP polymer

GC spectrum and MS spectrum of the compounds from the chloroform extract of polypropylene polymer are shown in Figures 11 and 12 respectively. All of the compounds found from the chloroform extract were fatty acid methyl ester (Table 5). So, most of the potential migrant compounds were polymer lubricants. The highest migration percent (83.85%) of total migrants (1.35%) is FAME compound, intentionally added substances and the rest percent will be trace amount of NIAS or impurities.

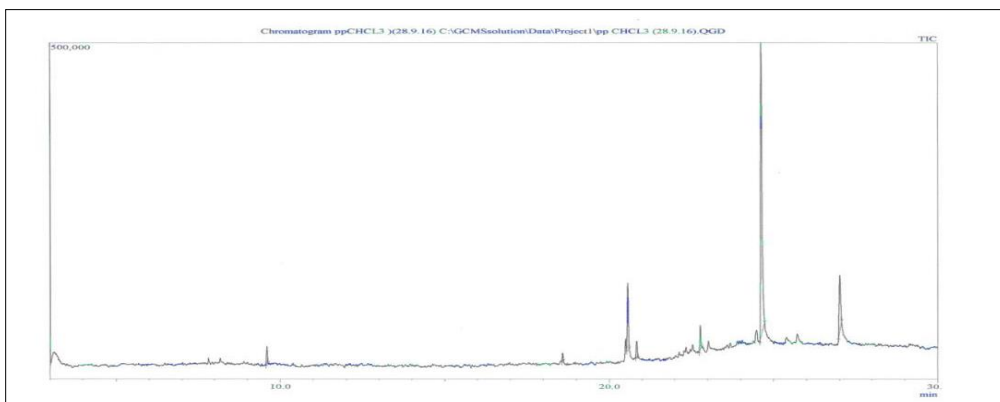


Figure 11: Gas chromatogram of CHCl₃ extract of PP sample

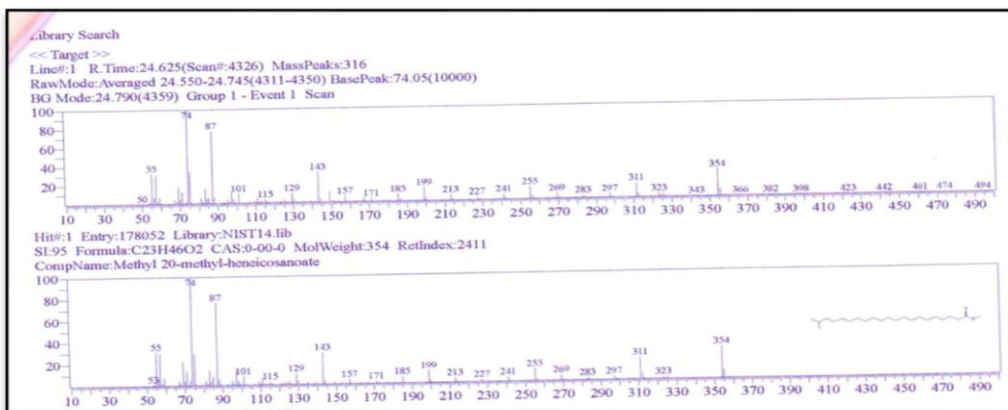


Figure 12: Mass spectrum of compound (retention time 24.63min) from CHCl₃ extract of PP sample

Table 5: Identified Polymer Components from the CHCl₃ extract of PP Polymer

No.	Name of Compound	Retention time (min)	Base m/z	SI	A%
1.	trans-Cyclopropanepentanoic acid, 2-undecyn-methyl ester	9.95	74.05	81	2.43
2.	12-methyl tetradecanoic acid methyl ester	18.58	74.05	82	1.02
3.	Hexadecanoic acid, methyl ester	20.49	67.05	89	11.25
4.	7-hexadecenoic acid, methyl ester	20.83	74.05	77	2.65
5.	Methyl 20-methyl-heneicosanoate	24.63	74.05	85	66.50
6.	Tetracosanoic acid, methyl ester	27.02	74.05	93	12.57

m/z - mass to charge ratio, SI - similarity index, A % - relative area percent

Identification of polymer components from the water extract of PP polymer

The gas chromatogram of polymer components from the water extract of PP polymer (Figure13). All identified compounds found from the water extract of polypropylene polymer are also FAME compounds (cyclic and straight chain), polymeric lubricants (plasticizers) (Table 6). But, the common used polymeric lubricants are not in the positive additive list. The safety assessment of saturated free fatty acids, the precursor of FAME compounds such as oleic acid, lauric acid, palmitic acid, myristic acid, and stearic acid shows that high intake has been associated with the incidence of atherosclerosis and thrombosis (Pub Chem, 2005). Chronic Exposure to palmitic acid is related to breast carcinomas and "leukemia-lymphoma" in animal study. These fatty acids are also related to insulin secreting function of islet beta-cells and CNS function (Warnotte *et al.*, 1999).

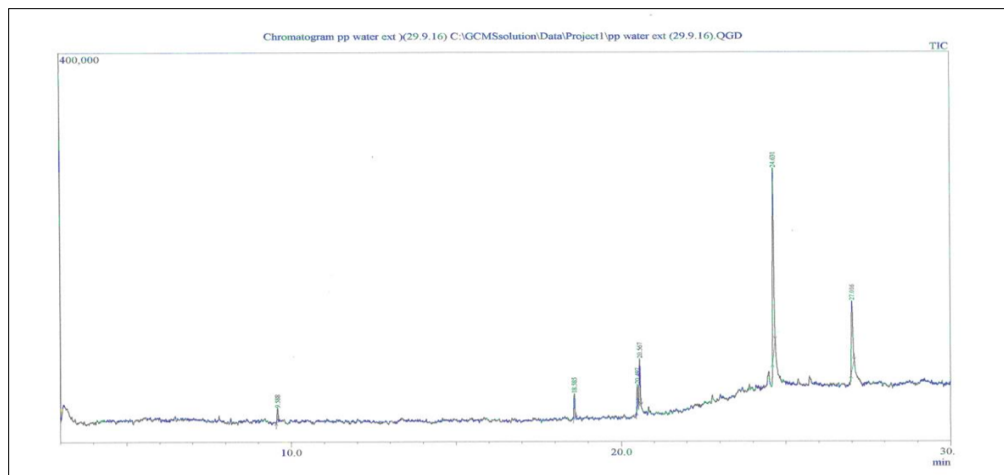


Figure 13: Gas chromatogram of water extract of PP sample

Table 6: Identification of Compounds from the Water Extract of PP Polymer

No.	Name of Compound	Retention time (min)	Base m/z	SI	A%
1.	Tras-2-Undecyl-cyclopropanepenoic acid, methyl ester	9.59	74.05	78	3.64
2.	Hexadecanoic acid, methyl ester	18.58	74.05	93	5.20
3.	9,12- Octadecenoic acid(Z,Z)-methyl ester	20.49	67.05	91	4.80
4.	7-octadecenoic acid, methyl ester	20.56	55.10	89	0.18
5.	Methyl 20-methyl-heneicosanoate	24.63	74.05	95	52.17
6.	Tetracosanoic acid methyl ester	27.01	74.05	90	24.02

m/z - mass to charge ratio, SI - similarity index, A % - relative area percent

Identification of polymer components from the oil extract of polypropylene polymer

The GC spectrum of the oil extract of PP polymer is shown in Figure 14 and MS spectrum of compound (retention time 18.59 min) is shown in Figure 15.

The oil extract of PP polymer contain the following groups of compound: long chain hydrocarbon compound (-ane, -ene, -ald, both cyclic and aliphatic) and fatty acid methyl ester and ketone (Table 7).

The polypropylene polymer is very closely similar to polyethylene polymer. The methyl branch group on polypropylene polymer makes it stronger and more flexible than polyethylene polymer. Their usage of additive may slightly different from each other in the amount and type of additives. From the results, most of the migrant compound groups from the PP polymer are similar to that of PE polymer. The constituents of olive oil (blank) were subtracted from the results.

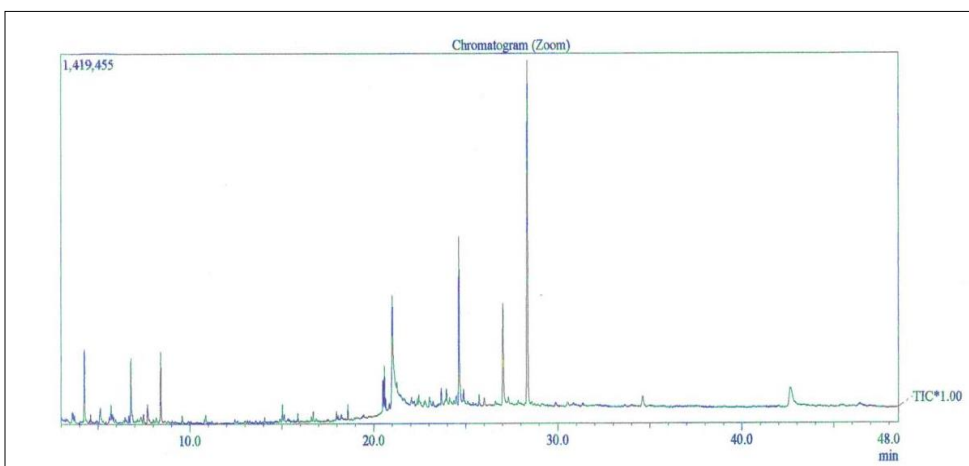


Figure 14: Gas chromatogram of oil extract of PP sample

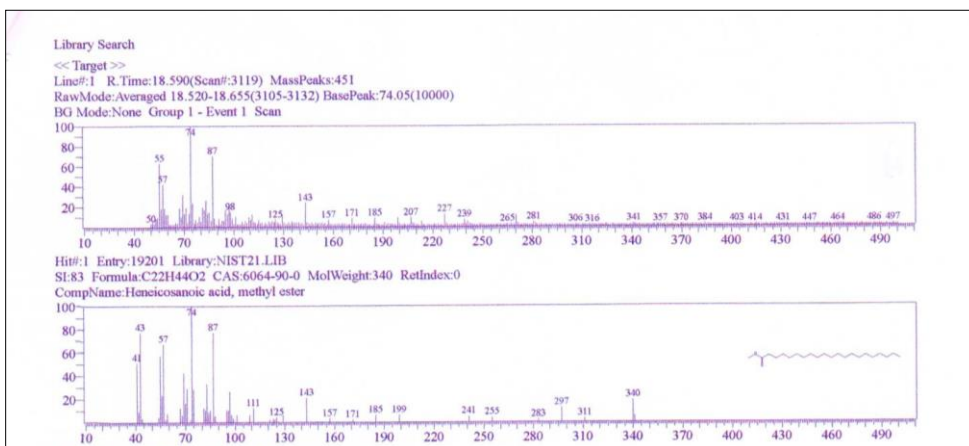


Figure 15: Mass spectrum of compound (retention time 18.59 min from oil extract of PP sample)

The long chain hydrocarbon compound (-ane, -ene, -ald, both cyclic and aliphatic), methyl ester of fatty acid and ketone are also used in PP polymer. Their usages and possible health effect were discussed in the migrant compounds groups from PE polymer.

According to the findings, the migration percent of IAS and NIAS from PP polymer is more than that of PE polymer. The migrant compounds released into the organic solvent (chloroform) were more than those into water, nearly seven times in PE polymer (0.81% in chloroform and 0.11 % in water) and four times in PP polymer(1.35% in chloroform and 0.34% in water). This means that watery food have less chance to contain these migrant compounds than other food types, except oily food, because most of migrant compounds are fat soluble. Comparison of two types of polymer, migration amount of polymer additives from PP polymer is more than those from PE polymer. Therefore, it can be concluded that PE polymer is more appropriate for all types of food rather than PP polymer.

Table 7: Identification of Polymer Components from the Oil Extract of PP Polymer

No.	Name of Compound	Retention time (min)	Base m/z	SI	A%
1.	Nonanal	4.26	57.10	94	4.29
2.	(E)-2-Dacenal	6.79	70.05	90	3.30
3.	(E,E)-2,4-Dodecadienal	7.70	81.05	87	1.37
4.	(E)-2-Tridecenal	8.41	70.05	90	4.15
5.	(Z)-Hexadecenoic acid, methyl ester	9.59	74.00	80	0.36
6.	1-Pentadecene	10.86	69.05	87	0.46
7.	Z-5-Nonadecene	15.03	55.05	88	0.88
8.	Cyclooctane (methoxymethoxy)	15.87	56.10	83	0.42
9.	1-Pentadecene	16.70	55.10	87	0.46
10.	9-Octadecenal, (Z)	17.97	55.05	87	0.34
11.	Hexadecenoic acid, methyl ester	18.59	74.05	83	0.66
12.	7-hexadecenoic acid, methylester	20.57	55.05	86	1.87
13.	2-Hydroxy-Cyclo pentadecanone	21.00	55.05	87	14.38
14.	(E,E,E)9-Octadecenoic acid,1,2,3-propanetriyl ester	23.66	55.05	83	0.96
15.	Heneicosanoic acid, methyl ester	27.01	74.05	80	8.87

m/z - mass to charge ratio, SI - similarity index, A % - relative area percent

Elemental Analysis of Polymer Samples by WDXRF

Elemental analysis by WDXRF showed that the common elements found in both types of polymer samples were Ti, Zn, Ca, Fe, Cl, Si, K and Al. Pb and Cu were found only in PE polymer and Ru and S were only in PP polymer (Table 8).

Table8:Relative Abundance of Elements in HDPE and PP Polymer by WDXRF

Elements	Relative Abundance (%)	
	HDPE Polymer	PP Polymer
Al	0.0169	0.0126
Si	0.0291	0.0391
S	-	0.0133
Cl	0.0410	0.0516
K	0.0104	0.0156
Ca	0.1080	0.1230
Ti	1.5200	0.0133
Fe	0.0846	0.0656
Cu	0.0122	-
Zn	0.1380	0.2040
Ru	-	0.0929
Pb	0.0404	-

From the observation of WDXRF spectra, high concentration of Ti element and medium concentration of Pb were observed in PE polymer. Ti is used as catalyzing agent in initial polymerization process. The contamination of Pb can influence the effect of human metabolism. Therefore, Ti may be IAS for the polymerization reaction and Pb may be contaminated from the raw materials sources or engineering process. Therefore, Pb may be NIAS. The other elements found from both types of polymers are not health concern and relatively small in abundance.

Conclusion

According to the results obtained from the FT IR spectra, the type of polymer of the selected Yoghurt bottle was confirmed as polyethylene (PE) polymer and that of disposable plastic cup was polypropylene (PP) polymer. According to GC-MS analyses, most of the additives found in two types of polymer were same chemical nature. The plasticizers mostly found from the two types of polymers are fatty acid methyl esters (FAME) such as tetracosanoic acid methyl ester, hexadecanoic methyl ester and methyl 20-methyl-heneicosanoate. Non-intentionally added substances (NIAS) such as di-n-decyl ether and 2-hydroxy-cyclo pentadecanone which may come from impurities of engineering machine or recycling process were also detected in these types of polymers. The migration percents of HDPE polymer were found to be 0.11 %, 0.81% and 0.06% and those of PP polymer were found to be 0.34%, 1.35% and 0.07% in water, chloroform and oil respectively. This study provided that polypropylene (PP) polymer has more leachable probability of additives than high density polyethylene (HDPE) polymer. The polymer additives compounds of both polymers are more soluble in organic solvent, chloroform, than in water. The migrant compounds in polyethylene and polypropylene food packages were mostly found to be oil soluble compounds. So, the migration would enhance into the oil at the elevated temperature up to 110°C. According to migration manner from this study, it can be concluded that HDPE polymer is more appropriate for the food packaging than PP polymer.

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