Migration of contaminants from food packages to its content – Brazilian scenario and regulation

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Abstract — *Consumers demand pleasant taste and odor in the final food product. Changes in these characteristics may be due to food deterioration or migration of substances from the packaging into food. The production of plastics for food containers involves catalysts which may contain toxic elements. In Brazil, there are limits of migratory substances and procedures for determining its rate. We aim to emphasize the food risks of migration of toxic substances from plastic packaging into its contents, and the Brazilian regulations regarding this issue. PET (polyethylene terephthalate) as raw matter for food packaging, is a concern in Brazil. The research methods currently adopted in Brazil are efficient for As, Cd, Co, Cr, and Sb determination. During PET synthesis and recycling, secondary reactions may form acetaldehyde, diethylene glycol and toxic oligomers. Monitoring by government mechanisms is essential to enforce safe food packaging. However, migration over the product shelf life remains an issue.*

Keywords — *polyethylene terephthalate, PET, migration, recycling, legislation.*

I. INTRODUCTION

The materials found in nature were useful to man as packaging for transportation and food storage. For this purpose, bladders and stomachs of animals, leather bags, leaves of plants, pieces of bamboo and hollow tree trunks, horns, gourds, pots of cooked clay, basket of vines, sewn straw and other materials were used as food containers (Risch, 2009). Foods are now marketed in packages made of various types of materials, such as paper, cardboard, glass, metal and plastic, among others. Due to its versatility, low weight, flexibility and low cost, plastic has been the most widely used material for manufacturing of packages (Vasco, 2012). Plastic food packaging was introduced after the World War II and one of the first products manufactured in 1946 were cups of polyethylene produced by Tupperware Co. founded by Earl S. Tupper.

Plastic arrived in Brazil in the 1950s (Wiebek and Harada, 2005).

As a consequence of its use, the polymeric material used in the preparation of the food packaging may contain residues of chemical elements from the materials used in its manufacturing, and these elements may migrate to the food (Shinamoto et al., 2011), in amounts compatible to its migrating rate and the time extent of contact between the food and its packaging (Risch, 2009).

Several studies have been carried out for the quantification of substances present in plastic packaging of soft drinks, water, milk, dairy products, juices and fatty foods, such as arsenic (As), cadmium (Cd), chromium (Cr), cobalt (Co) and antimony (Sb); being these metals great conductors of electricity and heat, of solid nature, flexible and malleable, with the exception of As that is not a metal and the Sb that is a semi-metal (metalloid), toxic in some chemical configurations; therefore, considered a great risk to human health (Soares et al., 2005).

The growth of the plastic packaging industry caused great concern, especially regarding its impact on the environment, due to the increase in the number of discarded packaging after the consumption of its contents. In Brazil, the National Sanitary Surveillance Agency (ANVISA), authorizes the reprocessing of PET (polyethylene terephthalate), which is a form of polyester and the most used plastic resin. However, its subsequent use in food packages is conditioned to strict rules.

Scientific studies report possibilities to use recycled PET in food packages, in three ways: (1) "half-PET" packages (Moura, 2011), where small proportions of PET-PCR is mixed to other substances in food packages manufacturing; (2) the external coating of virgin PET with recycled PET, for food packages; and (3) the use of recognized recycling technologies which have scientifically proved to supply uncontaminated

food-grade PET (ANVISA, 2016).

The migration of substances from plastic packaging into the packaged food is considered a public health issue. For this reason, many legislation systems have pursued harmonization policies, where regulations occur through lists of authorized substances and their use restrictions (ANVISA, 1999).

II. OBJECTIVE

In this context, we aim with this survey work, to emphasize the food risks that the general population runs due to the migration of toxic substances from plastic packaging into its contents, and the Brazilian regulations regarding this issue.

III. THE PET RESIN

The PET (polyethylene terephthalate) resin, a form of polyester, belongs to the group of three-dimensional polymers in which it has a structure that expands in all directions, that is, between the adjacent chains there are bonds through atoms that bind to others (Tavares, 2010). Polyester belongs to the group of condensation polymers whose polymerization takes place in two stages. The esterification of either the diacid or the diester with ethylene glycol, produces the monomeric diester, the bis-(2-hydroxyethyl)-terephthalate (BHET). When the starting substance used is terephthalic acid (PTA), such a reaction is recognized as a direct esterification, and when dimethylterephthalate (DMT) is used, the reaction is known as a transesterification reaction.

The byproducts of the direct esterification and transesterification reactions are water and methanol, respectively. Obtained by the oxidation of p-xylene, terephthalic acid is obtained, while ethylene glycol is synthesized from ethene, both of which are obtained from the Brazilian petrochemical industry. The PET obtained with the PTA route results in a polymer with a high content of impurity due to the difficulty in the purification of terephthalic acid (Nasser, 2005).

As example, in Brazil, Rhodia® operates with the PTA route while Proppet® uses the DMT route. They add catalysts such as calcium, magnesium, cobalt, manganese and zinc salts in order to accelerate the reaction and render it commercially viable. As a by-product at this stage, diethylene glycol (DEG) can be produced by the dimerization of ethylene glycol (MEG) (Nasser, 2005).

Another method was proposed for the synthesis of PET, based on the cyclic dimer polymerization without the use of catalysts and under atmospheric pressure, producing PET free of secondary products during the process (Al-Sabagh et al., 2016).

IV. RECYCLING OF PLASTIC MATERIALS

Defined as chemical recycling, or resin recovery, it comprises the depolymerization of plastic packaging materials, and the recovery and purification of the original monomers (Al-Sabagh et al., 2016). It consists in transforming or subjecting plastic materials to mechanical processes, shaping them physically into a form even different from the original. As in the case of thermoplastics, the primary plastic food packaging precisely fit the process, preserving to a large extent its physical, chemical and mechanical property of authentic polymers (Wiebek and Harada, 2005).

In the recycling units, the selected materials are subjected to an extrusion process, followed by abrupt quenching which, after being ground and dried, originates the recycled material, which is sold to the plastics industry for manufacture of new products or other materials. The main consumer market for recycled plastic in the form of beads are the plastic artifacts industries, which use the material in the manufacture of buckets, hangers, bottles for sanitation products, conduits and accessories for automobiles, among others (Vasco, 2012).

V. MAIN ADDITIVES AND CATALYSTS USED IN PET PROCESSING

Some of the most frequent additives for polymers are classified as plasticizers, thermal stabilizers, anti-UV and antioxidant substances. Even some studies mentioning PET as an additive-free material, some researchers have identified and, in some cases, quantified the presence of these additives in food-grade PET (Romão et al., 2009).

Despite being found a large and variable quantity of inorganic compounds that have catalytic activity for the production of PET in both stages, transesterification and polycondensation, the antimony dioxide (Sb_2O_3) is the most used catalyst because it shows a good balance of its catalytic activities, such as chemical stability in the presence of phosphorus-based stabilizers, final product color and low cost. Germanium oxide is also used; but even though it presents a higher catalytic activity compared to antimony, its high cost for PET production proves to be commercially unfeasible (Romão et al., 2009).

The production of PET for food and beverage packaging has been analyzed in the European and Asian continents; in some end products of food packaging or carbonated beverages, antimony-based residues were found, posing a risk to public health (Romão et al., 2009).

In the standardized test procedure ("challenge test" or equivalent) the compliance concentration limit of contaminants, compared to a standardized model for food-grade PET-PCR (Post-Consumer Recycled PET), is 220 ppb ($\mu g\ kg^{-1}$) for each contaminant into the PET used for manufacturing the food packages, or the limit of

$10\,\mu g\,kg^{-1}$ in the packages, for each contaminant. These two limits for the case of food grade PET-PCR derive from the maximum concentration of contaminants admitted in the human diet of $0.5\ \mu g\ kg^{-1}$ of food (ANVISA, 1999).

VI.   PET RECYCLING AND THE ENVIRONMENT

Recycling in one of the best alternatives to reduce the environmental impact of the discarded plastic material; the possibility of reprocessing the polymer can help reducing the environmental impact of the uncontrolled disposal of plastic wastes (Vasco, 2012). Since the PET bottle takes about 100 years to decompose, recycling proves to be a good proposal (Formigoni and Campos, 2012). The recovery of resins, also known as chemical recycling, comprises the depolymerization of the materials that can then be polymerized again for the production of new primary plastic packaging or other materials (Vasco, 2012).

PET packaging is 100 % recyclable and the process can be mechanical, energetic or chemical. Among these the most used is the mechanic (Figure 1) because it is cheaper (Wiebeck and Harada, 2005).
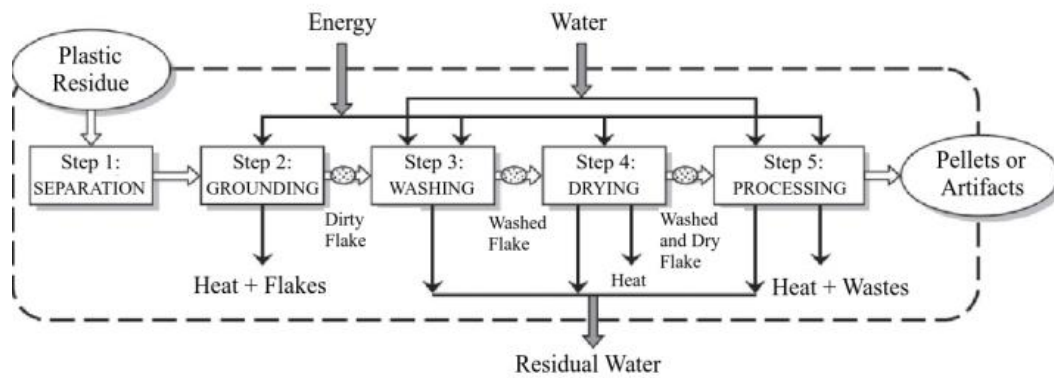

Fig.1: Schematic steps for plastic resin mechanical recycling. Source: (adapted from Formigoni and Campos 2012).

The processes of mechanical recycling of PET from beverage bottles produce flakes or grains and generally consist of the following steps: selection; crushing; milling; washing; separation by density difference; drying and extrusion. After drying the flakes, the material can be extruded and processed into grains for various applications (Wiebeck and Harada, 2005). This process is critical; during the melting of the material for grain production, the presence of small amounts of contaminants are evident and cause degradation, reducing the quality of the recycled PET and limiting its future applications (Risch, 2009).

In addition to the original chemical contaminants that may occur in the manufacture of PET, there are also the contaminants that can occur in recycled PET, either accidentally introduced or by chronic failures along the recycling process. The most evident contaminants of recycled PET are PVC (Polyvinyl chloride) in particular; but also, metals, sand and earth, glue, other plastics, and rust. Notwithstanding, multicolor packaging among several PET-packaged products shows a factor much more focused on the expansion of consumption than on environmental responsibility (Vasco, 2012); they may be an issue on recycling.

The development of new technologies applied to recycling of plastic, aims to produce a material to replace the virgin plastic, helping reducing the exploitation of mineral resources and the environmental impacts caused by the mentioned exploitation itself and the inappropriate waste disposal (Formigoni and Campos, 2012).

The Brazilian Association of Technical Normalization (ABNT), through the Brazilian Normative (NBR) 13.230, published in 1994, establishes symbols for the identification of thermoplastics used in containers and packages; a task of paramount importance for economic and industrial viability of recycling. These standardized symbols are usually embossed on the bottom of the package (Formigoni and Campos, 2012), as shown in Figure 2.
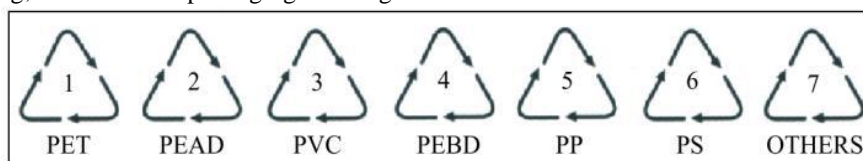

Fig. 2: Symbology used for recycling-able plastics. Brazilian Normative (NBR) 13230. Source: adapted from Formigoni and Campos 2012.

Such symbols only indicate that the materials are potentially recyclable. However, the coding system adopted warns that the presence of the symbol is not a stated or implied guarantee that any container is fit to be transformed into another product. Even if it is technically recyclable, no material should be considered precisely recyclable if there is no market for it (Formigoni and Campos, 2012).

VII.   QUANTIFICATION OF CONTAMINANTS

In Germany, a method of mass spectroscopy of isotopic dilution with inductively coupled plasma source (ICP-IDMS) was used for the determination of Pb, Cd, Cr and Hg, and its results were compared with those obtained by isotopic dilution mass spectroscopy with thermal ionization (TI-IDMS), with promising results (Soares et al., 2005; Shimatomo et al., 2011; Aghaee et al., 2014).

X-ray fluorescence spectroscopy (XRF) analysis was also tested in commercial polyethylene produced by Ziegler-Natta®, Philips® and metallocene® technologies. The XRF allows the direct determination of metals in polymeric matrices present at low concentration (1:1000) for elements presenting atomic number greater than, or equal to, that of Ti ($Z \geq 22$), that is, the technique proved to be effective for the determination of Ti, V, Cr, Al and Zr in polyethylene (Soares et al., 2002, Risch, 2009; Tavares, 2010).

VIII.   MIGRATION OF TOXIC COMPOUNDS AND SUBSTANCES

The global migration limit established by the Brazilian legislation is 50 mg kg^{-1} of simulant and 8 mg dm^{-2} of surface area, for packages with capacity greater than, or equal to 250 mL (≥ 250 mL). There is a specification of migration limits (ANVISA Resolution 105) for some PET starting substances (process-generated contaminants) (Table 1).

Table.1: Starting substances for PET manufacturing and its respective Specific Migration Limits established by the European Community (EC) and by the Brazilian Policies (Anvisa).

Starting Substance	SML (mg kg^{-1}) EC, 2016	SML (mg kg^{-1}) Anvisa, 1999
Terephthalic acid	7.5	7.5
Dimethyl terephthalate	Not established	7.5
Isophthalic acid	5	Not established
Dimethyl-isophthalate	0.05	Not established

| Ethylene glycol | 30 | 30 |
| Diethylene glycol | 30 | 30 |

The ANVISA resolution no. 105, of May 1999, is the current one. This resolution establishes the technical regulations on the use of plastic packaging and equipment that contact the food, as shown on Table 2.

Table.2: Maximum contaminant concentration (MCC) and specific migration limit (SML) of contaminants for some polymers (ANVISA, 1999).

Polymer	MCC (µg kg^{-1})	SML (µg kg^{-1})
PET	220	10
PS	180	6
PVC	90	5
PEAD	123	4
PP	78	25
PEBD	92	3

The health risk associated with the use of chemical substances depends, among other factors, on the dose of exposure and how they are metabolized by the human body. To assess the extent of this risk, it is necessary to know the physicochemical properties of the substances and their toxic effects in the short-, medium- and long-term. From these data, it is possible to characterize the potential adverse effects of human exposure to these substances (Nasser, 2005). The maximum amount of metals and / or simulant established by the Brazilian legislation is presented in Table 3.

Table 3: Maximum metal migration rate food content (specific migration limits – SML) established by the Brazilian legislation (ANVISA, 1999).

Metal	SML
Antimony (Sb)	0.05% (m/m)
Arsenic (As)	0.005% (m/m)
Cadmium (Cd)	0.01% (m/m)
Chromium (Cr)	0.10% (m/m)
Cobalt (Co)	0.005 (m/m)
Antimony (Sb)	0.05% (m/m)

IX.   POSSIBLE HEALTH RISKS ASSOCIATED TO CONTAMINANT MIGRATION

Risks to health with respect to substances that migrate from food packages to their content are varied (Tavares, 2010). The present survey highlights only some of the main migratory elements, such as antimony (Sb), arsenic (As), cadmium (Cd), chromium (Cr) and cobalt (Co).

9.1 Antimony (Sb)

Since antiquity, antimony compounds are well known. In cultures of the ancient East and Egypt, compounds with antimony were used for medicinal and cosmetic purposes.

Nowadays, drugs with Sb are still applied in treatments of leishmaniasis and schistosomiasis; it can, however, result in various side-effects and potentially develop heart-, kidney- and liver-related problems. In addition, Sb_2O_3 is classified as possibly carcinogenic to humans (Sundar and Chakravarty, 2010).

Aghaee et al. (2014) studied the contamination of water packaged in PET bottles, regarding the presence of antimony. This contamination occurs from the leaching of this metal from the polymeric material into the water. The results indicated that, over a six-month period, the concentration of antimony in stored water may increase by up to 90 %. Such information highlights the need for the development of studies that quantify antimony in PET and that sanitary authorities establish maximum tolerable limits. They also provide an alert regarding the need for this parameter to be monitored in the quality control of the material.

9.2 Arsenic (As)

According to the World Health Organization (WHO), the most common route of human exposure to As is the consumption of contaminated water. Other natural sources of contamination include minerals and rocks that contain the element, which includes soils and sediments formed from these rocks and geothermal and volcanic phenomena; the anthropogenic sources come mainly from mining activities, with tailings piles being the main sources of release of this metal (Smedley, 2003).

It is an important element of attention. Due to its harmful effects on the body, exposure to As, mainly through the ingestion of inorganic As present in water, is an important public health problem. Its consequences include the occurrence of various forms of cancer, diabetes mellitus, peripheral neuropathies and numerous pathological effects on the skin (hyperpigmentation and hyperkeratosis), gastrointestinal tract, and vascular system (Rodrigues and Malafaia, 2008).

9.3 Cadmium (Cd)

As pure metal, it is used in various industrial processes as a component of anti-corrosive coatings, metallic alloys, pigments and stabilizers, electrical batteries, and also in the manufacture of PVC. This metal can be found in phosphate-based agricultural fertilizers, cement manufacturing residues and in industrial sewage. The main forms of exposure to cadmium are contaminated air, water and food (Angerer et al., 1989). Due to its slow excretion and long half-life (decades, in human organism), it has become one of the most researched metals. The ingestion of foods contaminated by this metal can cause renal malformations and disturbances in the calcium metabolism (Jean et al., 2018).

9.4 Chromium (Cr)

Chromium, Cadmium and Arsenic are metals that do not exist naturally in any organism, with no nutritional or biochemical functions. Furthermore, the presence of these metals in any living organism is harmful, at any concentration. With the discovery of metallurgy, the use of these metals has grown and its toxic effects have generated permanent health problems for mankind and permanent damage to the ecosystem (Vincent and Lukaski, 2018).

Chromium is also used in leather tanning processes (Chakraborty et al., 2009), being associated with hypersensitivity reactions, being highlighted as the second most frequent cause of contact dermatitis. Among the general population, about 8 % is sensitive to this metal (Vincent and Lukaski, 2018).

9.5 Cobalt (Co)

Like all essential micronutrients, cobalt has two sides of incompatible exposure; both deficiency and excess can lead to death as well as trigger some diseases (Alves and Rosa, 2003). In the industry, this metal is mainly used in the production of metallic alloys; the exposure occurs during ore milling, mixing the powder with the other components, synthesizing and later machining of the steel in the manufacture of tools and parts for machinery, such as drills and polishing discs (Wehner et al., 1977). Cobalt has its role in therapy to replace radio in the treatment of some types of cancer. In the therapeutic field, it has its purpose for the treatment of cyanide intoxication as CoEDTA (Nagler 1978). The respiratory system is the major route of exposure, with effects on the cardiac system and thyroid gland, as well as the potential for cancer development (Alves and Rosa, 2003).

The migration of the mentioned above contaminants, from PET packaging to packaged products, is particularly important for the Brazilian population. The industry was able to establish the public concept that in Brazil, the so-called *mineral water* (water obtained from a natural water spring, licensed by the Government, without any treatment) is the only source of healthy water for consumption; families have acquired the habit of buying bottled water in 1 L, 2 L or 20 L PET bottles, storing it at home and consuming it continuously. The increased contact time between the PET and the bottled water, mainly due to the storage time, can worsen the scenario of the migration of contaminants to the PET bottled water.

The majority of the Brazilian population seems not to have the view that the water provided by the public system is properly treated, and that the use of ordinary household filters would be enough to obtain drinking-quality water with less contaminants. In Brazil, there is a specific transportation market and all logistics for supplying the population with PET bottled water, and

there is no industry interest in replacing PET packaging for water storage with other materials.

X. BRAZILIAN LEGISLATION

The Brazilian legislation (Resolution 105 of the Sanitary Vigilance Service - SVS, 19 May 1999) prohibits the use of plastic materials from packaging, fragments of objects, or recycled materials, except for PET. The Ordinance No. 978, 08 Dec. 1998, allows the reuse of recycled PET resins only for the manufacture of bottles as a constituent of the functional barrier layer with thickness greater than, or equal to 25 µm ($\geq 25\,\mu m$) and the recovered PET layer less than 200 µm ($< 200\,\mu m$), for products with shelf-life up to one year, under temperature limited to room temperature (ANVISA, 1999).

In Brazil, it became possible to use post-consumer resin (PET-PCR) in multi-layer packaging for non-alcoholic, carbonated beverages. However, one aspect often discussed is the risk involved in using recycled post-consumer polymers to contain food, beverages and pharmaceuticals, because of possible – and probable – contamination. For this purpose, co-injected PET packages could be used with three layers, by making a virgin PET sandwich with recycled PET filling (Romão et al., 2008).

The use of plastic materials in Brazil for manufacturing of food / beverage containers was regulated after the creation of MERCOSUL (Padula and Cuervo, 2004). The MERCOSUL regulations were implemented in Brazil through Ordinance No. 26 / SVS. The need for constant improvement of sanitary control in food production to protect the population's health, led ANVISA to review the Administrative Rule No. 26. This ordinance was revoked and currently, the Resolution No. 105, 19 May 1999, is the one in force. Therefore, this Resolution establishes the technical regulations on the use of plastic packaging and equipment in contact with food (Nasser, 2005). However, limits have not yet been set for the monomer bis (2-hydroxyethyl) terephthalate, as well as for PET oligomers.

The use of recognized recycling technologies which have scientifically proved to supply uncontaminated food-grade PET, is authorized in Brazil since 2008 (ANVISA, 2016). In this case, the food content can not be in contact with the recycled portions of the package, being the recycled resin used in external coating of virgin PET resin. The United States of America had recently approved the reuse of PET compounds by two technical processes (Supercycle® and Ecoclear®), and more recently, the technology Joncryl® claims to confer to recycled PET the same characteristics of a brand new PET resin. To date, Supercycle® is allowed in Brazil.

XI. CONCLUSIONS

The vast use of PET for food packages manufacturing, especially as container for beverages such as mineral water, fruit juice, and soft (carbonated or not) drinks, is a concern in Brazil. PET contains compounds with potential to migrate to food in contact; among these are the oligomers, for which there is not yet legislation established in Brazil. In this respect it is extremely important to know the hazardous components present in PET packaging and to have sensitive analytical methods that can be used to control and monitor its content in PET and migration rates.

The research methods currently adopted in Brazil are capable of evaluating the migration of elements from PET to its food or beverage content, mainly for As, Cd, Co, Cr, and Sb, which may constitute food contamination, causing human health harm. There are quantities allowed in the legislation, with the defined maximum tolerance limits (LMT); there should be analyzed, however, food and beverages to check if these limits are respected at the end of the storage period of the food packaged in PET.

During synthesis and also during PET recycling process, secondary reactions may occur forming acetaldehyde, oligomers and diethylene glycol. Such by-products, e.g. the oligomers, have the potential to migrate and contaminate packaged foods and beverages. There are few reports in the literature to explain the formation and possible migration of these compounds.

Therefore, in the initial synthesis or recycling of PET packaging, it is of great importance the monitoring by governmental Agencies, about the tolerable limits of contaminants, but the greatest issue seems to be a government monitoring program to evaluate residue levels in PET-packaged foods and beverages, at the end of its shelf life, or expiring date.

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