

Chemical Leachates from Food Contact Packaging Materials and Health Risks

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Abstract

Food contact packaging material should be stable and non-reactive with the food that it is in contact with, and must promote the safety of food. However, for a number of years, there has been evidence that indefinitely small amounts of chemicals could migrate or leach from food packaging materials into the foods they hold. The chemistry and toxicity of such leachates are well-known and documented; and there is a growing concern over the matter among the general public. In addition, food regulatory agencies are imposing more stringent regulations on the maximum allowable amounts of known leachates into foods in recent years. The majority of toxicity studies have revealed that trace amounts of leachates pose no immediate health problems in humans. However, studies on the effects of long-term exposure or synergistic/cumulative toxicity are still lacking. The present review provides an overview of the chemical nature and toxicity of certain leachates from various types of food contact packaging materials that are in wide use.

Key words: chemical leachates, food packaging, health risks, bisphenol A, phthalates, mineral oils

Introduction

More than 3000 chemicals have been approved by the US Food and Drug Administration (FDA) for use in food-contact applications. All these chemicals are considered as 'indirect food additives'. It's nearly impossible to detect what chemicals are leaching out from packaging material into foods as there's limited information disclosed by regulators, as well as the scientific challenges of this research. Food and packaging industries usually maintain the secrecy of components information since they view it as proprietary.

Leachates from plastic packaging materials

Bisphenol A (BPA)

BPA is used as the monomer to manufacture polycarbonate plastics and epoxy resins to line food cans or lids for glass containers. Epoxies are better suited for food can coating applications due to their desirable characteristics such as flavor-retaining property, chemical resistance, and have excellent mechanical strength. Condensation of BPA and epichlorohydrin yields BPA diglycidyl ethers (BADGEs), a

thermosetting resin. High levels of BADGE were discovered in canned fish in oil in 1996, and were found accumulated at concentrations in excess of 80 mg/kg, whereas the legal limit was 0.02 mg/kg. It has been reported that incomplete polymerization may result in a resin with free or residual BPA (in the ppm range). BPA can leach out of the protective internal resin coating of tinplate cans. Recycled paper containers are also contributing to BPA migration into food (Ozaki et al., 2004) as they usually contain ink additives. In a study, relatively higher amounts of BPA was found in canned foods (0-842 ng g⁻¹) compared to foods packaged in either plastic containers (0-14 ng g⁻¹) or paper containers (0-1 ng g⁻¹), and these results indicated that the liners in food cans are the major source of BPA exposure to humans (Sajiki et al., 2007).

Widespread human exposure to BPA has been reported, especially among the U.S. population. In a study, BPA was detected in more than 90% of the urine samples from participants over 6 years of age with total concentrations in the range of 0.4 to 149 µg/L (Calafat et al., 2008). BPA has estrogenic properties and studies have shown that it can cause multiple health problems, such as heart disease (Lang et al., 2008; Melzer et al., 2010), an increase in prostate and breast cancer, uro-genital abnormalities in male babies, male sexual dysfunction and reduced sperm quality (Li et al., 2010, 2011), early onset of puberty in girls and polycystic ovarian syndrome (Kandaraki et al., 2011), metabolic disorders including insulin-resistant (Type 2) diabetes (Melzer et al., 2010) and obesity,

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and neurobehavioral problems such as Attention Deficit Hyperactivity Disorder.

BPA is now classified as a category 2 substance, which indicates that its reprotoxicity is only suspected. However, in light of ample evidence, the French health agency ANSES has recently called on the European Chemicals Agency to propose a more stringent classification level, category 1, for BPA. According to the European Union classification of carcinogens, category 1 includes substances known to be carcinogenic to humans.

Epoxidized soybean oil (ESBO)

ESBO is used as a plasticizer and stabilizer for plastics such as polyvinyl chloride (PVC). It is particularly used in sealing gaskets of metal lids for glass jars, where it can be present at up to 40% of the weight of the gasket. Potential for migration into food both during processing and storage has been estimated (EFSA, 2006). Based on average concentration levels of ESBO in foods packaged in glass jars with PVC lined lids, the potential high dietary exposure of adults was estimated to be 0.25 mg/kg bw/day and was below the total dietary intake (TDI) of 1 mg/kg bw. ESBO is used up to 10% in PVC cling film. There was no acute toxicity observed. Long-term dietary administration to rats produced increased mortality, reduced body weight gains, kidney and liver changes (enlarged, fatty infiltration of the liver), degeneration of the testes, and slight changes in the uterus. However, these changes were noted at elevated concentrations.

Semicarbazide (SEM)

It has been detected in food contact materials made using azodicarbonamide, a substance which has been used to make plastic seals for lids on glass jars. A wide range of foods including baby foods have been reported to contain SEM at levels ranging from non detectable up to 25 ppb (EFSA, 2005a). Leaching of SEM from the breakdown of azodicarbonamide in sealing gaskets has been determined. SEM has been shown to be carcinogenic in mice, but not rats. Therefore, EFSA scientific panel concluded that the issue of carcinogenicity is not of concern for human health at the concentrations of SEM encountered in food.

Polystyrene plastic

One of the most widely used polymers for food package is polystyrene. Polystyrene widely used for packaging dairy products including yogurt, cream, cottage cheese, ice cream. Apart from that, meat trays, biscuit trays, fast food containers,

egg cartons, drink cups and instant cup noodle containers are usually made of polystyrene (Khaksar & Ghazi-Khansari, 2009). The degradation and migration of polymers into food products could be due to increased temperatures used in polymer processing (Till et al., 1982). There are studies on the migration of residual styrene from solid polystyrene into food and food stimulants (Reid et al., 1980; Till et al., 1982, 1987).

Plasticizers from plastic food wrap

Phthalates

Phthalates are a family of chemicals used in lubricants and solvents and predominantly used as plasticizers in plastics to increase their flexibility, transparency, durability, and longevity. Typically encounter in processed foods, and these chemicals passed into food from food processing equipment. Gaskets, flexible tubing, conveyor belts, food-prep gloves, seals on non-plastic containers, and inks used on labels, which can permeate packaging are the potential sources of phthalates (Freinkel, 2012). Di-2-ethylhexyl phthalate (DEHP) or dioctyl phthalate together with diethyl phthalate is widely used in the manufacture of food packaging materials, especially for food with high water content. Recently, substitution of phthalate plasticizers for expensive vegetable oils in emulsifiers/clouding agents and flavorings has been detected in certain food and beverages exported from Taiwan (USFDA, 2011). Especially DEHP contamination was confirmed in several products, primarily fruit juices, syrups, drinks, and jams.

Many thin plastic films (plasticized PVC), also known as 'cling films' that are widely used for packaging food at home contain di-(2-ethylhexyl) adipate (DEHA) as plasticizer. The levels of DEHA in retail foods packaged in plasticized PVC films found in the ranges, 1.0-72.8 ppm in uncooked meat and poultry; 27.8-135.0 ppm in cheese; 11-212 ppm in baked goods and sandwiches, and < 2.0 ppm in fruit and vegetables (Robertson, 2007). And, it seems that level of migration is directly related to exposed fatty portions of the food. However, nowadays, PVC is being replaced with linear low density polyethylene in the manufacture of cling films, which doesn't require plasticizers.

Relationship between phthalates exposure and preterm birth has been determined in a Mexico City-based study (Meeker et al., 2009). In that study, phthalate metabolite concentrations above median level were found in urinary samples of women who delivered preterm (<37 weeks of gestation). In another study, significantly higher phthalate levels, especially di-*n*-butyl phthalate were detected in low birth weight infants

compared with the controls (Zhang et al., 2009).

In the wake of deleterious effects of phthalates, food industry is being tried to eliminate or substitute them with alternatives. Recently, phthalate-free flexible tubing has been launched by Saint-Gobain, which uses a bio-based plasticizer Tygon S3, for a number of applications including specialist beverage, food and pressure processes.

Acetyl tributyl citrate (ATBC)

Plastic kitchen wrap (polyvinylidene chloride films) is commonly used for packaging of a large variety of foodstuffs. It contains about 5% ATBC. Nowadays, these wrapping films are widely used in microwave ovens to cover foods while cooking and heating. Leaching of ATBC from kitchen wrap into aqueous liquids has been established (Nara et al., 2009). In that study, among the tested food-stuffs, highest leaching rate was observed in protein-containing aqueous skim milk solution. The citrates are reported to have a higher toxicity to HeLa cells than phthalates though their leaching rate is low (Nara et al., 2009). The ATBC toxicity is well documented, and also found to inhibit the proliferation of lymph-node T cells (Ametani et al., 1998).

PET bottle leachates

The neutrality of odor and taste are essential requirements for any packaging material intended for beverage packaging including mineral water. Poly(ethylene terephthalate) (PET) is the most suitable polymer to serve this purpose and therefore PET bottles are widely used for beverages especially for mineral water.

Antimony from PET bottles

Antimony trioxide (Sb_2O_3) is used both as an additive and a catalyst in the PET manufacturing. As a result most commercial PET material typically contains 190-300 mg/kg Sb. Antimony trioxide is a suspected carcinogen (IARC, 1989), and has been listed as a priority pollutant. A number of studies have confirmed the leaching of Sb from PET(E) bottles used for mineral water, flavored soft drinks, citrus fruit juices (Shotyk & Krachler, 2007; Welle & Franz, 2011); and found that it existed at 100 times elevated level in commercially available bottled waters compared with uncontaminated ground water (2.2-3.8 ng Sb/L) (Shotyk et al., 2006).

Migration of acetaldehyde from PET bottles

Acetaldehyde is a degradation product of the PET polymer generated during preform production. After cooling down, acetaldehyde is trapped in the PET bottle wall and migrates

into the mineral water after filling and storage. The migration of acetaldehyde is of special interest because acetaldehyde is detectable in low concentrations in mineral water as a fruity off flavor. The taste threshold limit of acetaldehyde in mineral water is reported to be in the range of 10 to 20 ppb (Haack & Ewender, 2000).

Mineral oil migration from recycled food packaging

Recycled cardboard contains different types of mineral oils, and these oils consist of hydrocarbons in the form of mineral oil saturated hydrocarbons (MOSH) and mineral oil aromatic hydrocarbons (MOAH). Paperboard produced by recycled newspaper is usually thought to contain increased levels of mineral oils due to their origin from newspaper print inks. They may contaminate the packed food at levels far above the legal limits framed by Joint FAO/WHO Expert Committee on Food Additives (Biedermann & Grob, 2010).

MOSH is linear and cyclic (non-aromatic) in nature with predominantly 16-25 carbon atoms. MOSH having molecular mass even up to C_{28} can transfer to dry food through the gas phase because of their volatility, which ranges from 200 to 810 mg/kg (Biedermann et al., 2011). On the other hand, MOAH constitutes 21% of the mineral oil, and consist of 3-ring and 4-ring compounds. Dry food stored in recycled paperboard boxes was found to contain 10 to 100 times higher levels of mineral oil than the prescribed safe limit.

Toxicity of the MOAH is largely unknown, however they are largely considered unacceptable in foods. Currently, insufficient data is available on human exposure levels to mineral oils, their storage in body and elimination. Therefore, further research is needed to determine level of mineral oil that could pose a potential health risk.

Health risks of mineral oils

Mineral oils may cause damage to liver, the heart valves and the lymph nodes, according to the German Federal Institute for Risk Assessment (BfR). As certain mineral oil compounds resemble that of the polycyclic aromatic hydrocarbons, they are assumed to exert mutagenic/carcinogenic effect. Mineral oils are stored in body fat because of hydrophobic nature (ifp Institut für Produktqualität GmbH, 2012)

Photo-initiators from recycled paperboard

Isopropylthioxanthone

It's a well-known photo-initiator in ultraviolet light-cured inks. Traces of isopropylthioxanthone (ITX) have been found in packaged milk and fruit juices as it is often used in printing

inks for offset printing on the outside of milk packaging materials and beverage cartons. According to the European Food Safety Authority report, levels of ITX in dairy products varied from 27 to 445 µg/L, and from 5 to 249 µg/L in fruit juices, nectars, and drinks (EFSA, 2005b).

Limited genotoxicity has been observed with ITX under *in vitro* conditions (Kirkland et al., 2005). Change in lipid structure in multilamellar liposomes of saturated phospholipids of different length with and without cholesterol was observed in the presence of ITX (Momo et al., 2007). However, *in vivo* genotoxicity study using the mouse bone marrow micronucleus didn't show any signs of systemic toxicity, no increase in the incidence of micronucleated polychromatic erythrocytes (Notox, 2004).

Benzophenone (BP) and 4-methyl benzophenone (4-MBP) in cereals

Trace amounts of the photo-initiators BP, 4-MBP were found in certain cereals when 4-MBP was used in the printing inks for cardboard printing. These photo-initiators can migrate from packaging to the food through the vapour phase, even from the secondary packaging (Pastorelli et al., 2008). The European Food Safety Authority (EFSA) published recommendations and the Standing Committee for the Food Chain and Animal Health endorsed a limit of 0.6 mg kg⁻¹ for the sum of BP and 4-MBP. Multilayer material consisting of PET/silic oxide/PP has been reported to prevent BP migration. In a survey conducted on recycled paperboard, BP was found in 61% of samples, and 4-MBP in 30% of the samples (Koivikko et al., 2010).

Hormone-disrupting leachates

Nonylphenol (NP), an estrogen-mimicking chemical produced by the breakdown of antioxidants (alkylphenol ethoxylates or tris(nonylphenyl)phosphate) used in plastics, from certain food contact materials has been reported. NP was detected at parts per million levels in examples of polystyrene and PVC, but also in ethylene vinyl acetate, polycarbonate and rubber at much lower levels. There is no EU specific migration limit for this chemical since it is an impurity or a degradation product. Leaching of this chemical from PVC cling film may pose health risk (FSA, 2006).

Fire retardants in butter: Food contamination with polybrominated diphenyl ether (PBDE) flame retardants was reported recently (Schechter et al., 2011). Surprising levels of deca-BDE, a PBDE compound widely used in electronics as well as in textiles, wire and cable insulation, was found in some

samples.

Teflon components in microwave popcorn: Scientists established probable links with high cholesterol, kidney and testicular cancer, thyroid disease, pregnancy-induced hypertension/preeclampsia and ulcerative colitis and perfluorooctanoic acid (PFOA) exposure. PFOA has been used for many years by DuPont in the synthesis of Teflon for oil and grease-repellant coatings for food packaging.

Dibutyltin compounds are used as stabilizers in containers and packages made of polyvinyl chloride. Dibutyltins have been found in beer, margarine, processed cheese and wine.

Cumulative or synergistic toxicity

Residual solvents, stabilizers, antioxidants, and plasticizers (trapped temporarily in materials) in multilayer packaging materials can migrate into packaged foods. Generally, multilayer food packaging materials are composed of several layers of unrelated materials such as aluminum, polyethylene, cellulose, PET, nylon, carton, etc. The residual solvents including toluene, methanol, ethanol, ethyl acetate, methyl ethyl ketone, isopropyl alcohol, etc., can change flavor of food and can lead to unexpected health problems. Possibility of synergistic effects between the known leachate chemicals cannot be overlooked. As there appears to be a mixture of chemicals migrates simultaneously from a recycled or multilayered packaging material, there's always the possibility for synergism in exerting toxicity despite their occurrence in minute amounts. Further research in that direction is warranted.

Conclusions and future perspectives

The scale of possible damage by a chemical leachate and probability of such occurrence can depend on the harmfulness of the substance and the degree to which the consumer comes into contact with the substance. As the majority of corrugated board is manufactured from recycled fiber, there's always high potential for exposure of food packed in corrugated board to contaminating chemicals. More and more studies are needed to ascertain clearly the effects of long term exposure of these chemical leachates. And, there is a real need to develop more sensitive techniques for determining leached chemicals in food matrices.

Recently, the trend towards development of biodegradable, environmentally friendly food packaging materials is gaining momentum. Polylactic acid, a bio-degradable polymer derived renewable resources, is one of the promising polymers that can replace petroleum-based plastics. Biofoams made of starch

derived from potatoes, wheat or corn may replace polystyrene foams (USDA, 2009). All these trends will eventually replace the conventional petro-derived toxic hydrocarbons in food packaging materials. Meanwhile, wise selection appropriate fiber for recycling, inks and recycling process technologies can help reduce the risk of chemical migration.

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