



Commentary

On the origin of primary aromatic amines in food packaging materials

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ABSTRACT

Primary aromatic amines (PAAs) are substances that can be transferred from food packaging materials into foodstuffs and are “possibly carcinogenic to humans”. The formation of PAAs from multilayer packaging materials consisting of aromatic polyurethane (PU) adhesives occurs from the reaction between residual isocyanic monomers that have migrated to the surface of the inner layer of the package and water molecules making contact with the same plastic surface. However, for foods subjected to thermal treatments, an alternative formation of PAAs should also be taken into consideration. Due to the detrimental effect of the temperature, some secondary bonds (namely allophanate and biuret bonds) displaced on the main PU backbone may be disrupted, originating neo-formed isocyanic monomers. The migration of these monomers from the adhesive layer across the inner sealing film can lead to PAAs as soon as they come into contact with the water molecules of the liquid or high a_w packaged food. Although the existence and the mechanisms of the formation of allophanate and biuret linkages during the polymerization process with poly-isocyanates has been known for a long time, the negative impact on public health possibly arising from the migration of the neo-formed isocyanic monomers into the foods during thermal treatments seems to have not been fully perceived, with special regard to preservative heat treatments such as pasteurization and sterilization. In this viewpoint article, after covering both chemical and physical aspects involved in the formation of PAAs, especially in thermally-treated PU-based multilayer packaging materials, we have stressed the necessity for a more careful consideration of the risks associated with the potential formation of PAAs, in a first instance by strict compliance with the provisions included in the current European legislation. The necessity for alternative analytical tools for the PAAs quantification is also highlighted.

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1. Introduction

Multilayer packages consist of several layers of different materials (generally from two layers up to 15 layers) on top of one another to yield an aggregate thickness defined, by convention, to be less than 250 μm (thicker structures are typically identified as “sheet” materials) (Dunn, 2009). The combination of different plastic materials, sometimes including metal or cellulosic substrates, underlies the rapid popularity of laminated structures, as they allow for fine-tuning the final performance of the ultimate package to precisely match the food’s requirement for an extended shelf life.

Several converting operations concern multilayered packaging materials, namely printing, coating, laminating, and finishing.

Lamination, in particular, is the operation that allows for the holding together of different layers for the entire life cycle of the package. Although extrusion lamination and coextrusion are widely adopted processes, the lamination mediated by an adhesive (often called “tie” layer) finds application in most converting lines due to the ease of manufacturing and the low cost involved (Rosato, 1998) (Fig. 1a). Among the wide assortment of adhesive systems commercially available, polyurethane (PU) adhesives are well-known for superior flexibility, mechanical and adhesion properties, and weathering resistance (Malucelli et al., 2005), which play an important role when severe processing/environmental conditions (e.g., high temperature of retort processing, aggressive chemicals, or high-moisture environments) may affect the package performance and the quality of the food inside it.

PU adhesives come from reactive systems usually including two main components, one carrying isocyanic moieties (NCO-terminated) and the other containing hydroxyl groups (OH-terminated). These components, when mixed together, undergo chemical

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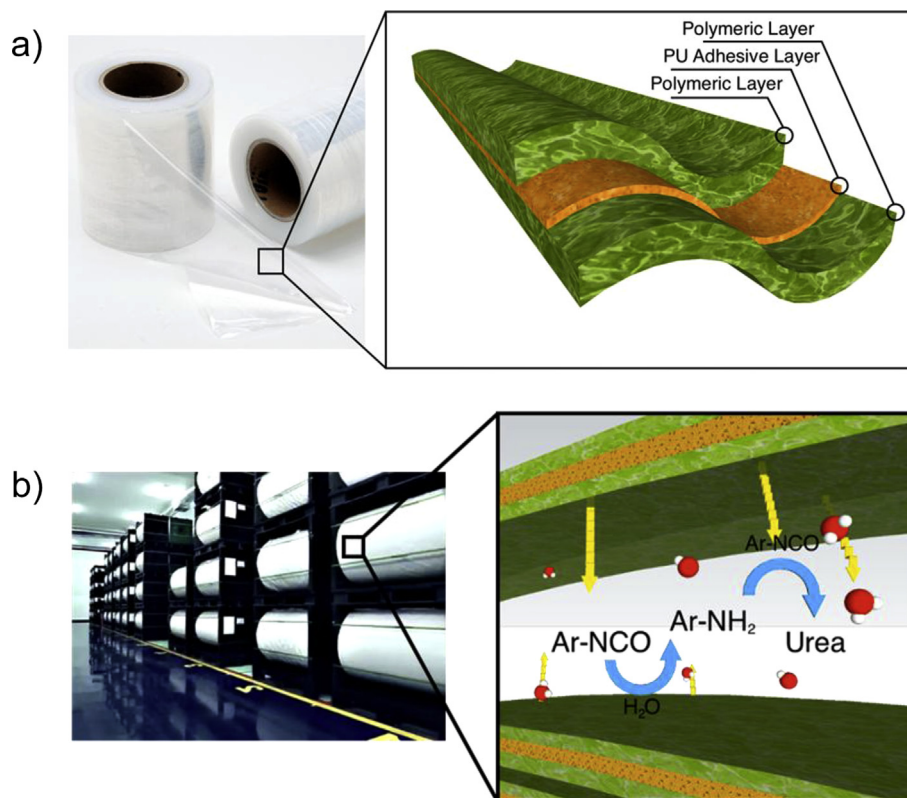


Fig. 1. (a) Schematic representation of a PU adhesive-based laminate packaging material (3-layer structure). (b) Sketch on the formation of poly(urea) in PU adhesive-based multilayer packaging materials wound in reels. Red-white spheres: water molecules; Ar-NCO: aromatic isocyanate monomers; Ar-NH₂: aromatic amines. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

reactions (i.e., chain extension) that bind the applied materials into a solid layer of high molecular weight PU adhesive (Fig. 2, scheme 1). The adhesive components react after lamination, further increasing the molecular weight of the PU adhesive to achieve the required performance (Meier-Westhues, 2007). Because the adhesives are made of reactive chemicals that are expected to polymerize by linear extension and/or cross-link, government food-safety-related agencies (e.g., the Food and Drug Administration [FDA] in the United States and the European Food Safety Authority [EFSA]) have established a strict legislative body to control the risk associated with the potential migration of toxic substances from the adhesives to the food (e.g., unreacted residuals), which in turn might also have harmful effects on the consumers' health (De Fátima Poças & Hogg, 2007). In the case of multilayer packaging materials (e.g., pouches, trays, bags, wrappers, etc.), in particular, one of the main issues related to PU adhesive systems is the potential presence of primary aromatic amines (PAAs) in the food matrix (Ellendt, Gutsche, & Steiner, 2003). The potential risk associated with PAAs arises from the suspected carcinogenic activity of some of them that can affect humans (e.g., 2,4 and 2,6-diaminotoluene and 4,4-methylenedianiline), as stated by the International Agency for Research on Cancer (Vineis & Pirastu, 1997).

Unlike the U.S. regulation, which has banned the aromatic-based PU adhesives to the advantage of fully aliphatic systems (U.S. Food and Drugs Administration, 2000), the European legislation establishes that "... plastic materials and articles shall not release primary aromatic amines in a detectable quantity into food or food simulant. The detection limit is 0.01 mg of substance per kg of food or food stimulant (namely, 10 ng g⁻¹ of food). The detection limit applies to the sum of primary aromatic amines released (expressed as aniline)" (European Commission, 2011). Excluded

from this provision are the species reported in Table 1 of Annex I of the same regulation (e.g., 1,3-phenylenediamine and 1,3-benzenedimethanamine).

The most popular method for the quantification of PAAs is the spectrophotometric method, developed by the German Federal Institute for Health Protection of Consumers and Veterinary Medicine (BgVV) (Brauer & Funke, 1991). This method is based on the derivatization with N-(1-naphthyl)ethylene-1,2-diamine dihydrochloride (NEDA) of the amines present in the aqueous food simulant (acetic acid 3%, w/v) after the migration test. The final colored compound is then measured spectrophotometrically at 550 nm (i.e., the maximum absorbance of the aniline derivative). Quantification of PAAs is expressed as equivalent to aniline. The main drawback associated with this method is its non-selectivity, which may lead to an overestimation of the total amount of PAAs found in the aqueous simulant (Aznar, Canellas, & Nerín, 2009). To overcome this issue, several methods have been developed during the last decade (Akyüz & Ata, 2008; Andrisano, Gotti, Di Pietra, & Cavrini, 1994; Aznar et al., 2009; Nogueroles-Cal, López-Vilarino, Fernández-Martínez, Barral-Losada, & González-Rodríguez, 2008; Shelke, Sanghi, Asthana, Lamba, & Sharma, 2005). Most modern analytical techniques based on mass spectrometry and advances in ionization processes for non-volatile compounds have made use of UHPLC-MS and their hyphenated techniques such as UHPLC-MS/TQ and, most recently, UHPLC-Q-TOF/MS (Matarozzi, Lambertini, Suman, & Careri, 2013; Pezo, Fedeli, Bosetti, & Nerín, 2012). All of these methods, besides targeting increasing sensitivity, demonstrate that the unequivocal identification of all compounds present in food packaging materials, including the non-intentionally added substances (NIAS), can be achieved.

The origin of PAAs is primarily linked to residual (unreacted)

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