Chapter

Fillers for Packaging Applications

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Abstract

Packaging in general is frequently overlooked and demonized. The lack of educational programs and efficient waste treatment lead packaging to be treated as an environmental problem. However, packaging is an enabler of our society because it makes feasible the availability of any and every good, regardless of its production location. Furthermore, the packaging business plays a significant role in the global economy, following a continuous trend of growth. The use of fillers in various packaging types can be a valuable resource not only for reducing its cost but also improving its mechanical strength (therefore reducing the number of raw materials required for making that specific package), improving its visual properties to ensure customer attractiveness, creating new possibilities of use, and extending the shelf life of perishable foods. However, the use of fillers in packaging should be made in a way that permits proper recovery and recycling after use.

Keywords: packaging, packaging market, paper packaging, plastic packaging, nanofillers, circular economy

1. Introduction

Before proceeding, it is worthwhile to make a little observation about the environment around you. Take a quick break and behold what surrounds you and the place where you are.

Unless you are reading this in the wild of nature, everything you see was made available through some sort of packaging, even the materials used in the construction of the place where you are were bundled, contained, or packed.

Also, everything you eat—regardless of being an ordinary meal or a gourmet delight—came to you via packaging entrusted to keep high food safety and hygiene standards. Even if one has a backyard or farm that provides a wide variety of fresh produce, at some point in time some packaged foods will be bought and brought home.

Now think about all medicines that are consumed by millions of people every day to keep their well-being and health. Without being noticed, packaging allows for one of the most noble uses: to provide a longer and healthier life. You may now have realized for the first time that life as we know it exists at current standards due to a powerful enabler: packaging. Some studies suggest an association between the quality of life in a certain location and the level of packaging development in the same place [1].

The reason for this is simple: our society has developed in a way that knowledge and experimentation are part of everyday life. We are informed about the latest trends and we want to try new things. At the same time, just a small fraction of the goods we buy is produced in our vicinity. Most goods are produced thousands of kilometers away from where we live. Some make journeys to our home longer than the longest travel we have ever done.

The definition of the term "packaging" is very broad [2, 3], but it is possible to define it as "a system that consists of different materials, machinery and symbols which are set up in a way to contain, protect, communicate and allow access of its contents to make goods available in a cost-efficient way."

It is not the intention of this chapter to go into detail on marketing aspects of packaging, but references will be made when necessary since most products on shelves rely on their packaging as the sole communication tool with their consumers.

2. Packaging market

The global market of packaging was estimated to be as of US$ 851 billion in 2018 as per recent estimates (**Figure 1**) [4, 5].

Considering the market share by material types, cardboard (including corrugated boxes) and plastic packaging in their flexible and rigid forms correspond to almost 80% of the whole market. Fillers are used mainly in these three types of packaging.

Cardboard production has increased due to the rapid growth of e-commerce in recent years. Flexible packaging has proven to be a lightweight and cheaper alternative to glass and metal. The annual growth of the packaging segment is estimated to be between 3.5 and 4.3% and is influenced by the urban shift of populations, the trend of pre-packed products in the food segment, and diversification of retail landscape [6].

It is important to highlight that these estimates were made before the COVID-19 pandemic of 2020, economy was still not precise by the time this chapter was being written. Although exact figures cannot be given, packaging market growth is expected to follow global trade trends, according to previous studies [7].

It is estimated that in 2018 the global plastic production almost reached 359 million tonnes. Even in Europe, where environmental concerns are usually high and public opinion plays a big role, packaging is of key importance since it represents almost 40% of the plastics demand. This share is very expressive since it is more

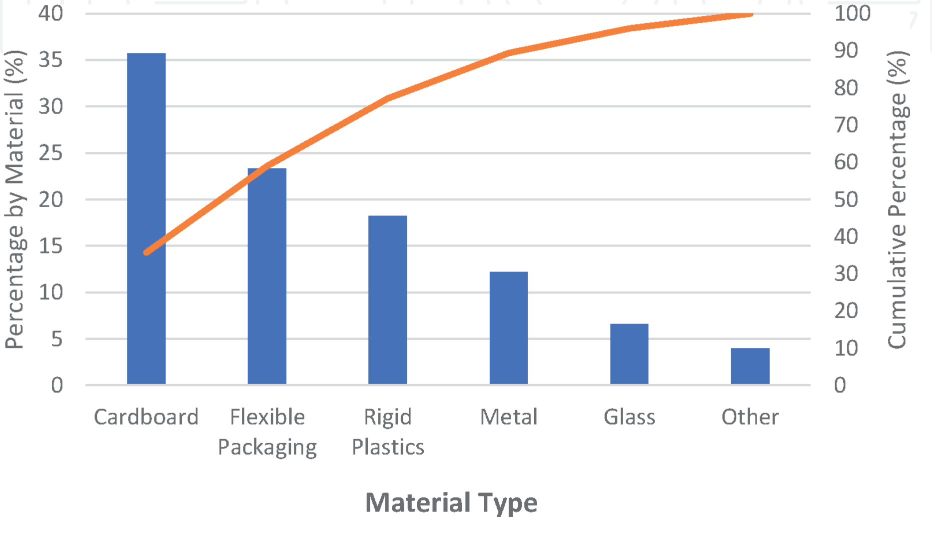

Figure 1.
World packaging market (share by material) [4, 5].

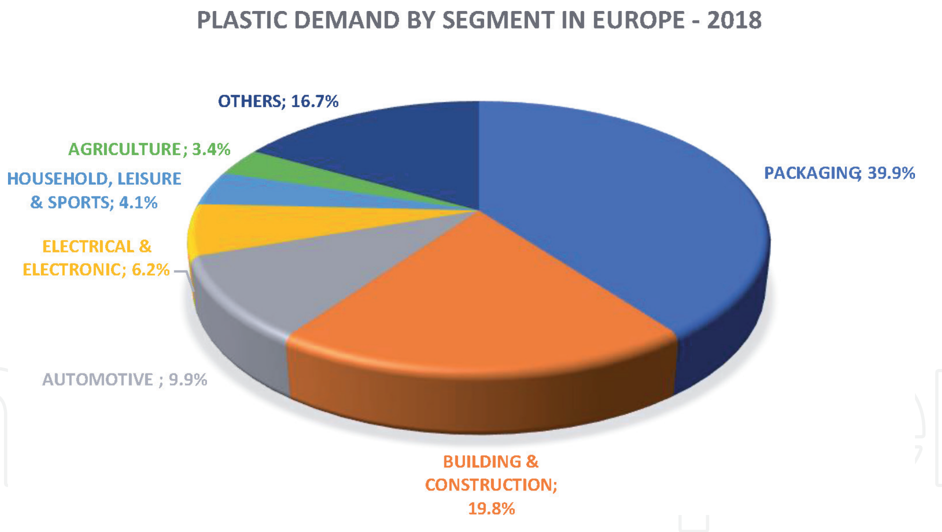

Figure 2.
Plastic uses in Europe [8].

than double the second main use of plastics (building and construction, which corresponds to almost 20% of share) (**Figure 2**) [8].

3. Fillers used in paper packaging

There are two major categories of cellulose-base packaging: corrugated cases—the standard brown box using for transporting—and paperboard carton as you may see on supermarket shelves containing cereals, toothpaste, frozen food, etc. There is a wide array of drawing possibilities in both categories and the choice about what should be used depends on product requirements, machinability, and design strategy. These materials are used for "secondary packaging" (because it does not get into contact with the product) in the case of paperboard carton or "transport packaging" in the case of corrugated cases for obvious reasons.

The enhancement of visual properties in paper packaging given by fillers makes its use imperative for assuring customer attractiveness. This is not the case for corrugated cases, which have the function of protecting and grouping various units of products during transport. However, for paperboard carton packaging, this feature is crucial for standing out on shelves, grabbing consumer's attention, and providing improved sales.

Fillers are not added to the process of "converting" cellulosic packaging (printing, cutting, creasing, gluing, etc.) but in the production of the various paper grades that will be further transformed.

Fillers used in papermaking are defined as insoluble particles (from 0.1 to 10 μm) added to the slurries of cellulosic fibers before the formation of paper. The uses of fillers vary from zero to 30% [9]. On average, it is estimated that minerals used for wet-end addition and coatings make up 8% (by mass) of the materials used in the paper industry [10].

What defines the amount of fillers used in papermaking is the grade and purpose of that specific paper or paperboard. For instance: an aseptic multilayer packaging like the milk box may have little or no fillers at all, while printing papers may have 30% (by mass) or more of its content made by fillers.

There has been a noted increase in the number of fillers being used, driven by lower cost of fillers themselves compared to the fibers itself, and by meeting market demand for higher opacity and/or brightness. The most common fillers found in papermaking can be seen in **Table 1** [11].

Chemical composition	Natural source	Synthetic source
$CaCO_3$	Ground limestone (GCC) Chalk (ground)	Precipitated calcium carbonate (PCC)
$Al_2O_3.2SiO_2.2H_2O$	Clay (hydrous kaolinite)	Precipitated aluminum silicate
TiO_2	—	Titanium dioxide
$Mg_3Si_4O_{10}(OH)_2$	Talc	—
$CaSO_4.2H_2O$	—	Gypsum

Adapted from [11].

Table 1.
Common fillers used in papermaking.

Fillers from renewable sources can also be used in papermaking [12]. Most common types of natural fillers are:

- **Starch or starch derivates**—for increasing fiber bonding and consequently mechanical properties like tensile, tearing and folding strength.

- **Cellulose derivates (like microcrystalline cellulose)**—for encapsulating fillers and increasing its bonding strength to cellulose fibers, as well as improving heat capacity.

- **Chitin or chitosan**—also for improving bonding, since chemically speaking the structure of the molecules of chitin and chitosan are very similar to that of cellulose.

- **Xantham gum or anionic guar gum**—for improving paper strength.

Although the use of fillers can influence a wide array of attributes in papermaking (coefficient of friction, permeability, burn rate), the two main drivers for their use in cellulose-based packaging are:

- **Cost reduction**—this can be achieved in two ways: either by partially substituting cellulose content or by reducing the energy demand required for drying paper.

- **Visual properties**—the addition of fillers greatly enhances printing quality on paper packaging by improving opacity, brightness, and smoother surface from calendaring. This is particularly important for products that communicate their brand through their shelf packaging, with the need of attracting customers.

Other uses of fillers can lead to the development of nonconventional applications, although commercial use of several novel applications is still strict [13]. Examples are deodorant paper [14], antimicrobial paper [15], flame retardants [16], and magnetic paper [17].

However, fillers reduce the flexibility of paper, posing extra attention to converting operations. Side effects, like creasing, if not done properly the packaging may show small cracks when folded, resulting in reduced mechanical strength and visual defects that can compromise the packaging function of protection and its attractiveness at the point of sale. Furthermore, and due to its mineral nature, fillers increase abrasion and dusting.

To overcome these drawbacks, fillers have been modified in several ways to achieve desired properties in an optimized way. Examples of processes that allow filler modification are modification with inorganic substances, modification with natural polymers (or their derivatives), modification with water-soluble synthetic polymers, modification with surfactants, hydrophobic modification, cationic modification among others [13].

4. Fillers in plastic packaging

Plastic packaging is widely used due to its wide range of applications, low cost, relatively high level of safety in terms of migration, and the convenience it can add to packaged products. They have been extensively employed for substituting more expensive packaging materials like glass, metal, and even wood [18].

Plastics are commonly used as "primary packaging," meaning it directly contacts the product. Plastics can also be used as secondary packaging to bundle bottles for instance or to transport packaging in the form of a stretch film.

Regardless of its use, there are two major groups of plastic packaging, based on their physical characteristics:

- Flexibles—which include all wraps, bags, and other packaging that can be used manually or automated through vertical or horizontal machines.

- Rigid—which includes all sorts of tubes, bottles, containers, and drums. This type of packaging is suitable for liquids and products requiring mechanical protection or specific positioning. Due to the higher amount of mass required for achieving rigidity, it is also more expensive and substituted with flexible alternatives whenever possible.

Recent developments in the area of material engineering make use of a wide variety of fillers to modify the original properties of plastics. Advances in the production of nanofillers also have shown promising areas of research. These functional fillers can result in unique properties that enhance the performance of packaging materials even at small loads.

In theory, any material that can be found or transformed into small particles can be used as a filler for plastics in general. Potential combinations are endless but for commercial purposes, fillers for plastics should be readily available, insoluble, and chemically inert; have a low hardness to avoid wear; and be nontoxic, non-flammable, and finally free from metal impurities, which can degrade plastics [19].

In general, the most common fillers used in plastics are calcium carbonate, aluminum trihydrate, talc, kaolin, mica, wollastonite, glass fiber, aramid fiber, carbon fiber, and carbon black. However, it is important to highlight that fillers are used mostly in engineered plastics (**Table 2**).

The use of fillers affects plastics by modifying original material properties or adding features that did not exist in the original polymer. Specifically for this aim, properties can be changed due to the use of glass fibers, mica flakes, nano-clays, carbon nanotubes/nanofibers, natural fibers, wood flour, talc, and kaolin [21].

Plastic packaging containing nanofillers has been shown to have an enhanced performance especially for food packaging, where high barrier properties against water vapor, aromas, and oxygen are mandatory [22].

Filler source	Group	Examples
Inorganic	Oxides	Glass, SiO_2, ZnO, Al_2O_3, MgO
	Hydroxides	$Mg(OH)_2$, $Al(OH)_3$
	Salts	$CaCO_3$, $CaSO_4$, $BaSO_4$, phosphates
	Silicates	Talc, kaolin, mica, montmorillonite, wollastonite, feldspar
	Metal	Steel, boron, (silver for antimicrobial nanofillers)
Organics	Carbon, graphite	Carbon fillers and nanotubes, carbon black, graphite fibers and flakes
	Natural polymers	Cellulose, starch
	Synthetic polymers	Polyester, aramid, polyamide and PVOH fibers
Natural fibers	Straw	Wheat, corn, and rice
	Bast	Hemp, jute, kenaf, lax
	Leaf	Pineapple leaf, sisal
	Seed/fruit	Cotton, coir
	Grass fibers	China reed, bamboo, grass
	Wood fiber	Hardwood, softwood

Adapted from [20, 21].

Table 2.
Examples of fillers used in plastics.

Because the surface area of nano-clays can be more than 750 m^2/g, the use of nanofillers even in loads smaller than 2% (in volume) creates a tortuous path for the diffusion of gases through the polymer matrix, therefore improving its gas barrier. This is of special interest because nanofillers can be an alternative to multilayer coextruded packaging, which are commonly not recyclable. Another advantage of adding nanofillers (like montmorillonite-MMT, kaolinite, carbon nanotubes, and more recently graphene nanosheets) in plastic packaging is the improved mechanical properties that allow downgauging and consequent economic and environmental benefits.

The use of nanofillers can also enable broader use of biopolymers in packaging. This group of materials has been studied for several decades but, usually, their properties are much weaker when compared to petroleum-based polymers. They include natural sources like starch; cellulose; proteins (collagen, soybean protein, zein, etc.); and polylactic acid (PLA). The low-performance properties of these "green" materials and can be compensated by the addition of nanofillers, broadening the possibilities of use at commercial scale [23, 24].

On top of the mechanical reinforcement and improved barrier properties, nanofillers can extend the shelf life of products that may spoil due to the development of pathogenic or spoilage microorganisms. It is known that the use of nanofillers made of silica through the sol-gel method has improved the shelf life of fruits due to their hydrophobic characteristics [25].

Besides the features mentioned above, other beneficial functions of nanofillers in food packaging are the possibility of exploring and developing new technologies in the area of active packaging, where the role of packaging goes beyond traditional purposes. Main groups of research areas can be listed as follows [26, 27]:

a. Oxygen scavenging—oxygen causes food spoilage due to oxidation or due to enabling aerobic bacteria development. Even under proper vacuum, package materials may allow the permeation of O_2.

b. Nanobased sensors—for compensating transport and storage conditions (e.g., temperature variations) and informing about the potential shorter shelf life of products.

c. Detection of gases produced by food spoilage—microbial development is commonly followed by gas production, which can be detected by conducting polymer nanocomposites.

d. O_2 indicators—since the development of aerobic microorganisms happens during the presence of oxygen, the control of this gas is necessary to evaluate product decay.

e. Enzyme immobilization at the nanoscale—sometimes the direct uses of enzymes is restricted due to its potential degradation during processing. The absorption of enzymes by nano-clays embedded in a polymer matrix is a promising mechanism for efficient release control.

However, the fillers at nanoscale should be only used after a careful study of their toxicology and potential harm to humans and the environment. It is known that migration may happen from food-contact materials through various mechanisms that may not be fully extracted by standardized simulants or quantified by current analytical methods [28].

5. Environmental concerns

More than ever, the packaging is under public scrutiny and blamed for environmental problems. Although only approximately 10%, of all solid waste, is plastic, up to 80% of all waste accumulated in land, shorelines, seabed, and ocean surface is plastic due to its long decomposition time. This is a threat to aquatic animals since they can be entangled or ingest plastic fragments. Some researches suggest that over 260 species of animals (invertebrates, mammals, seabirds, fishes, turtles) have ingested or been entangled by plastic debris.

Due to its fragmentation properties, plastic packaging can decompose into microplastics and pose yet another type of hazard. The bodies of all marine species, ranging in size from plankton to the blue whale, contain plastics [29]. The food chain has detrimentally changed.

While attention has been focused on the number of microplastics in oceans, almost no effort has been made in fighting root causes. The pollution on land, in rivers and the ocean, is not caused by plastic packaging or any other packaging material, but by the lack of educating the public on the consequences of littering.

Furthermore, massive marketing is promoting miraculous solutions and materials for the sake of the environment without technical evidence about its effectiveness. Deceiving customers with the promise of mitigating their environmental impact is called greenwashing.

Another problem in evaluating the environmental benefit of packaging is to neglect the components of the product itself. It happens for the vast majority of products that the packaging impact is much lower than that of the product itself [30] and eventual packaging failure will lead to increased waste.

The most advanced tool to evaluate the eco-efficiency of packaging is the Life Cycle Assessment (LCA), which has already been used for a wide array of applications with great acceptance from the scientific community. It provides comparative data about the environmental impact of the product under analysis considering various aspects like Global Warmth Power (GWP), ozone depletion, toxicity (both cancerous and noncancerous), particulate respiratory effects, ionizing radiation, photochemical ozone creation potential, acidification potential, aquatic eutrophication, freshwater ecotoxicity potential, and nonrenewable resource depletion. Other parameters to be considered are land use, freshwater use, and cumulative energy demand (CED) [31].

It was already mentioned that fillers reduce the drying energy required in papermaking, which will be transformed into cellulosic-based packaging. Although the use of fillers may reduce the carbon footprint of papermaking in some aspects [32], a full LCA analysis should be performed to confirm assumptions. Nevertheless, the environmental impact of mineral fillers in the production of paper or paperboard is expected to be negative according to perspectives below:

a. **Substitution of cellulosic fibers**—during plant growth, CO_2 from the atmosphere is captured resulting in negative emissions. Although industrial processes have positive emissions for the production of paper, the net absorption of CO_2 is much larger. Numbers vary according to countries, paper grades, and technological development, but public data from companies that have an integrated paper facility show that every ton of finished product captures the equivalent of 2.6–3.8 tons of CO_2 equivalent ($CO_{2eq.}$) from the atmosphere [33, 34]. Adding fillers for the sake of sheer cost reduction with no technical benefits increases the environmental impact of the final packaging (in terms of $CO_{2eq.}$).

b. **Replacement of a renewable raw material by a nonrenewable one**—although synthetic fillers can be produced, mineral fillers of natural sources are extensively used. They are mined in several parts of the world, which means dependence on the natural availability of that specific resource. On the other hand, cellulose fibers come from trees, which can be planted and harvested repeatedly, if responsible soil and water handling are provided.

c. **Harder to recycle**—many cellulosic-based packaging may be produced from recycling another packaging due to availability and costs. If the paperboard can not be easily recycled, it is likely to be sent to energy recovery (incineration) or, more commonly, landfilling. During the degradation of cellulose, CH_4 is emitted, representing a GWP of about 25 times that of CO_2 [35]. Depending on the quality and grade of the recycled paper pursued, fillers have to be removed to provide a higher quality of recycled materials (which also have a higher market value). It is estimated that the removal of fillers along with other contaminants (stickers, inks, fines, etc.) require 30–100% more recovered paper for the same final product, resulting in a higher amount of nonrecyclable waste to deal with [36].

d. **Environmental impact for making fillers**—the process of making fillers comprehends an extensive production chain, which includes removal of impurities, engineering particle shape and size, and finally enhancing its properties through thermal and/or chemical treatments. Also, fillers should be transported to the paper mill facility, which may be located far away, incurring fuel consumption and extra environmental burden. The mining activity alone is very complex: it includes exploration of the deposits, resource development, feasibility and reserve development, mine planning (including permitting and construction), and production. When resources are exhausted and the mine is closed, further work is necessary for restoring the area [37].

Fillers of organic origin like wood waste-derived fillers have been studied for several decades and can contribute toward reducing solid waste, even in large-scale commercial activities, although compounders are somehow reluctant to consider this technology because they are not sure how to handle it or are not aware of a market to justify production [38, 39].

Unfortunately, for the time being, the amount of LCA studies evaluating the environmental impact of fillers added to polymers is likewise small but indicates potential lower GWP comparing to standard plastics. Better eco-performance is achieved using nanofillers, which reduces the amount of petroleum-based raw materials for achieving the same performance although some exceptions can be found (e.g., LDPE and talc).

The addition of natural fibers to partially substitute polymers may also be an opportunity for reducing the environmental impact especially because they come from renewable sources, but again an LCA analysis is necessary for confirmation due to the production burden of raw materials [40]. It is important to highlight that LCA studies should include not only the resources used to produce that specific type of packaging but also the after-use impact. This broad view limits the boundaries of the analyses "from cradle to grave" and is fundamental for having an overall picture of the system.

The process of recovering material by recycling is becoming increasingly important due to environmental and economical concerns. One recent driver toward reduced environmental impact is the concept of circular economy, where the traditional linear model of production is substituted by the concept of keeping goods in use for the longest possible time (**Figure 3**) [41].

One of the models of circular economy in use was proposed by the Ellen MacArthur Foundation, commonly known as the "butterfly diagram." The smaller the loop, the greater the value of the material in question. The right side of the diagram (in blue) refers to "technical cycles" where goods are designed to stay in use for the longest possible period through sharing, reusing, extended durability, repairing, and recycling, the last possible loop.

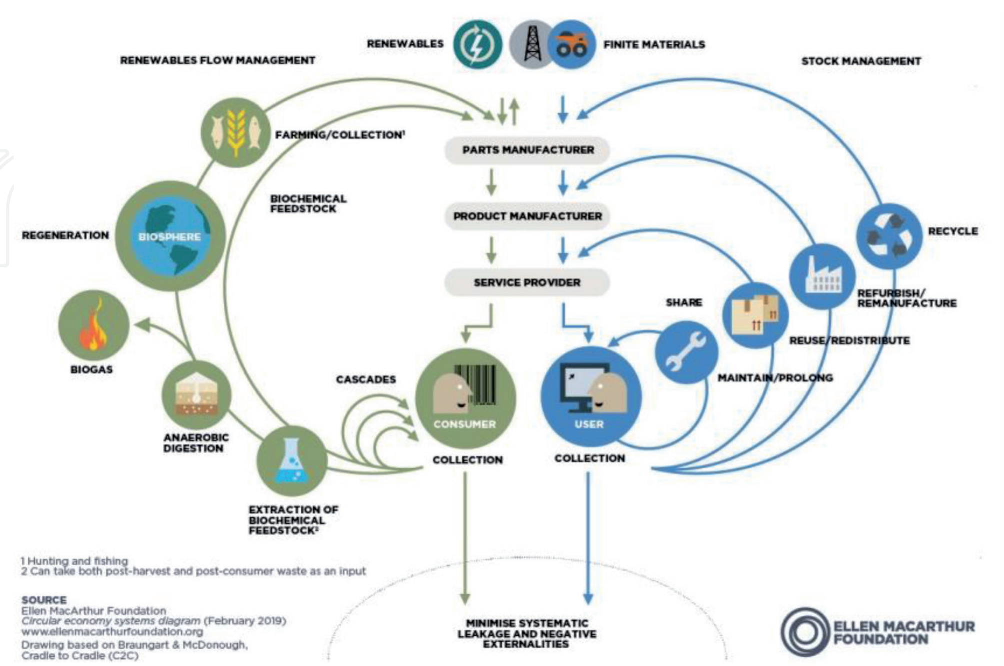

Figure 3.
The circular economy model with technical (right) and biological (left) cycles proposed by the Ellen MacArthur Foundation. Reproduced with permission.

The left part of the diagram (in green) refers to "biological cycles," where components of any goods or packaging may biodegrade. This is a feasible alternative for organic waste or sewage, but it is the least wanted solution for packaging because recycling has a greater value and lower environmental impact than biodegrading.

6. Conclusion

Packaging plays a fundamental role in our society by providing accessibility to food and any other type of physical goods. Broad multidisciplinary knowledge is necessary for making the definition of a packaging system accurately.

The choice of packaging which is more adequate for a specific product must pass through the definition of suitable materials and its properties for best fulfilling production, distribution, preservation and sales requirements. Most important packaging materials are made of cellulose (cases and folded carton) and polymers (flexible or rigid packaging), which are the types of materials where fillers are mostly used.

No matter what type of packaging system is chosen, the end of life of that specific packaging (reuse, recycling, incineration or composting) should always be considered in the project phase. The selection of the proper filler(s) combined with the optimum amount used to contribute to the success of that package either during its lifespan as well as during the recycling stage.

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Conflict of interest

The author declares no conflict of interest.

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