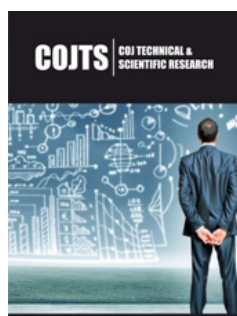


Novel Biodegradable Skin Tissue-Carrier for Advanced Cosmeceuticals and Nutraceuticals

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Abstract

The vehicles to be applied on skin, hair or mucous membranes have the scope to load and carry selected active ingredients releasing them at the designed areas, dose and time. Regarding their use in the field of cosmetics (i.e., cosmeceuticals) and diet supplements (i.e., nutraceuticals), these carriers are generally made by emulsions and solutions containing not only active ingredients, but also preservatives emulsifiers and other chemicals necessary to maintain the product integrity and stability during their storage and use. Moreover, they are packed by containers 60% of which are made by non-biodegradable plastic material. The aim of the paper is to suggest the possibility to manufacture and use alternative novel smart and biodegradable carrier-tissues that made by natural polymers embedded by active micro nano block polymeric complexes are free of any kind of chemicals and globally biodegradable. The suggested carrier-tissues have been made by biopolymeric fibers bound to micro-nanocomplexes of chitin nanofibrils-nano chitin (CN-LG) encapsulating different active ingredients. The ingredients have been selected in advance to characterize effectiveness and safeness of the realized advanced medical devices, innovate cosmeceuticals and nutraceuticals, packed by biodegradable containers. This proposal could be useful to make innovative products that able to improve human health and beauty, may contribute to safeguard the environmental conditions reducing the food and agro-forestry waste and slowing down the global Green House Gas (GHG) emissions invading our planet. At this purpose and to better understand the great problems caused by this increasing pollution. The paper will start giving a look on the land and ocean waste that continuously invaded by plastic and microplastic became dangerous for the health of humans animals and the environment. Moreover, some considerations will be reported regarding production and use of biopolymers which including chitin nanofibrils and Nanolignin have been utilized from our research group for making the proposed innovative biodegradable cosmeceuticals and nutraceuticals. In conclusion its considered necessary to change the today way of producing and consuming not only reducing the actual food loss and waste, but also becoming to utilize innovative cosmeceuticals, nutraceuticals and clothes made by biodegradable polymers finally adopting the circular economy.

Keywords: Waste; Microplastics; Cosmeceuticals; Nutraceuticals; Polymers; Chitin nanofibril; Lignin Tissues; Carriers

Introduction

Pollution and waste, increasing and invading lands and oceans by macro and microplastic debris are entering into the food of marine animals and humans resulting dangerous for the global health [1]. Moreover, waste ranging 1.6 billion tones yearly represents the 8/10% cause of the global Green House Gas (GHG) emissions [2]. Plastic materials. In fact, are widely

employed not only to make disposable medical equipments and textiles, but also to produce food and cosmetic packaging which, releasing 22 million tons of waste in 2019 is estimated to double the quantity by 2060, representing 3.4% of the today global GHG emissions [3]. Consequently, the worldwide people are starting to change the way of consuming and living, becoming more sensitive to their own health and the environment protection. Therefore, consumers are starting to purchase products made prevalently by biodegradable and sustainable technologies. They, wish to reduce or eliminate the actual great quantity of food and agro-forestry waste supposing that their rich content in active ingredients and natural polymers including polysaccharides could be utilized to make biodegradable and safe products. At this purpose and as discussed later biopolymers such as chitin and lignin might be useful to obtain biofibers utilized to make specialized carriers for manufacturing innovative tissues and films, necessary to produce. For example, textiles ,skin and environmentally disposable medical devices and equipments, smart cosmeceuticals, nutraceuticals and packaging [4]. Chitin and lignin are not only bio-eco-compatible, non-toxic and biodegradable polymers, but are also made by microstructure and morphology, easily modifiable in their mechanical properties [5]. At this purpose, the nitrogenous polysaccharide chitin and/or the polyphenol-rich lignin found in crustacean /microbes and plant/algae respectively result particularly useful to make carriers for biomedical food and cosmetic applications, because easily obtainable at low cost from waste material also [6]. However, as for chitin and lignin the complex linear or branched structure of the majority of natural polymers are composed by monosaccharide units connected by glycosidic bonds, giving rise to neutral (i.e.dextran), cationic(i.e.chitin/chitosan) or anionic(i.e.Hyaluronic acid) compounds useful to make biodegradable fibers (Figure 1). Moreover, Chitin/chitosan and lignin/yaluronic acid obtainable in great quantity from natural and waste material may be easily bonded each other by the ionotropic gelation method to produce micro-nano particles, being polymers covered by electrical positive and negative charges respectively (Figure 2); [6-8]. For all these reasons, our research group are proposing to make micro-nano technological block polymeric complexes by the use of these biopolymers at their micro/nano size to make micro-nano nanoparticles (NPs) capable to encapsulate different active ingredients into their structure. Successively and according to our experience the “activated” NPs may be embedded into films or bound to fibers of non-woven tissues to make innovative advanced medications and/or novel cosmeceuticals and nutraceuticals. These innovative cosme-nutraceuticals might be used to obtain the so-called Beauty from within (i.e. the use of the same ingredients applied on the skin by cosmeceuticals and taken by oral route by nutraceuticals) [9,10]. This new beauty approach seems to represent

the new way that consumers are becoming to consider at the basis of their appearance and health [9,10]. Therefore by this new patented carrier-tissues made by selected bioactive ingredients obtained from food and agro-forestry waste. It seems possible to make not only novel cosmeceutical/nutraceutical carriers and products but also biodegradable packaging. Therefore, it might be realized drugs, food, cosmetics and packaging of a new generation, by the use of the reported innovative film sand tissues [11-13]. Additionally, these novel products of great interest to reduce the actual no more sustainable organic waste could be useful for starting to create innovative goods for a better green economy also [14-16]. Finally, as soon after focused, it is to underline the possibility this technology could have for manufacturing water-soluble and water-insoluble tissues which being really free of water, preservatives, emulsifiers, fragrances and other chemicals might be useful to make innovative cosme-nutraceuticals in agreement with the EU rules for products “free from” [17-19].Therefore the aim of the paper is to range a better beauty with an healthy life and a more safe environment, according with the scientists and consumers request [6-8]. At this purpose it seems also useful to report some introductive news on the global pollution problems to better understand the consumers’ worry for the environmental disasters and the need to change the way of producing and consuming by the use of biodegradable goods as the proposed tissue-carriers.

NATURAL POLYMERS AND SOURCE

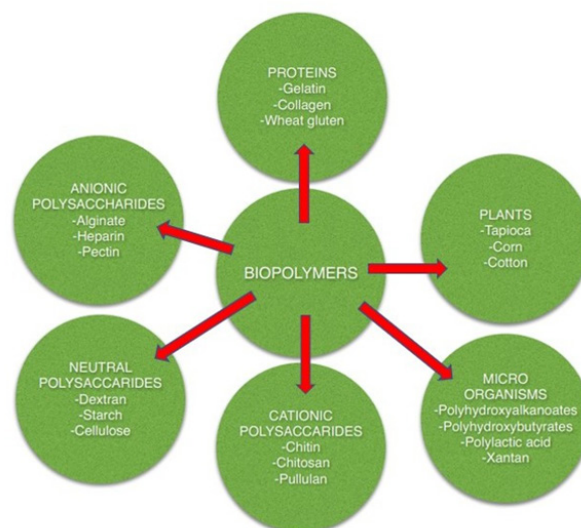


Figure 1: Source and characteristics of natural polymers.

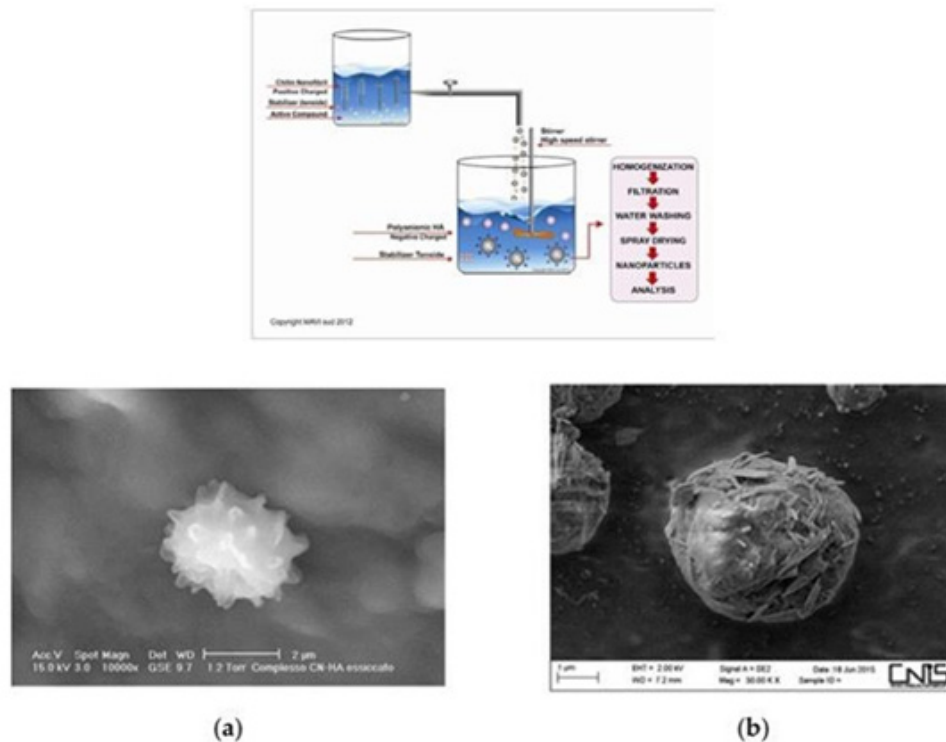


Figure 2: Micro nano particles of hyaluronan/chitin obtained by the gelation method up) between hyaluronic. acid Chitin
 a)
 b)
 Courtesy of Morganti et al. [6]

Plastic Pollution and Waste

As previously reported the main plastic waste is due mainly to food and cosmetics packaging, cloth-textile-fibers)and disposable medical equipments released in landfill and oceans as microplastics [20]. At this purpose, it is to know that “each year in Europe about 3800 tons of Microplastics(i.e.over seven kilos every minute!)are released into the environment through the use of everyday cosmetics and personal care products while the beauty industry produces annually 120 billion units of packaging 60% of which made by non-biodegradable plastic material [21]. This waste is increasing day by day for the microfibers continually released into the oceans by the dressings treated by the washing-machines as well as the city dust road markings, fertilizers and other chemicals which end up every day in the environment’ land and air (Figure 3); [3,22,23]. In conclusion regarding the 2.01 billion tons of municipal waste generated worldwide each year and expected to grow to 3.40 billion tons by 2050 around 14 million tons are due to microplastics accumulated on the ocean floor while 1.5 million tons enter into the oceans annually also [22-25]. Unfortunately, in 2019 only 9% of all plastic waste ever produced has been recycled while about 12% has been incinerated and at least 79% was mismanaged ending up in landfills, oceans or the natural environment [26]. However, waste composition differs across income levels, reflecting different production and consumption [27]. Thus, high-income countries with an high income level of 96%, are generating 51% of dry waste (i.e. plastic, paper, cardboard, metal and glass) while middle- and low-income countries with an income level between 82 and 39%

are generating 57% of food and green waste ,so that the recycled materials result different (Figure 4 & 5); [27]. At this purpose, its to remember that the daily per capita waste generation in high-income countries is projected to increase by 19% by 2050 compared to low- and middle-income countries where it is expected to increase more than three times [27]. Moreover, it has been estimated that in 2016 the 2.1 billion tons of carbon dioxide (CO₂) equivalent GHG emissions generated as wastes is supposed to increase to 2.38 billion tons by 2050. Additionally, it has to be underlined that 26% of these emissions (i.e.52.3 billion tons of CO₂ equivalent) are due to food production and consumption including supply chain (18%) livestock & fisheries (31%) crop production (27%) and land use (24%) [28]. At this purpose, it is to remember that the global food and textile systems, contributing to the use and consumption of plastics are among the main cause of the environmental economic and health problems affecting the society because of the release of gas emissions during their production and distribution processes [20-22]. It is not to be forgotten that clothes footwear and household textiles are responsible for many of the GHG emissions and landfills waste as well as for about 20% of water pollution [29]. It has been estimated that the European fashion industry in 2015 was responsible for 10% of global carbon emissions (i.e.654kg of CO₂ per person) causing the release of 79 billion cubic meters of water. Moreover, due to the washing and laundering of clothes made by natural and synthetic polymers 0.5 million tons of micro fibres are released in the oceans every year accounting for 35% of primary microplastics released into the environment (Figure 6);

[29]. Thus, the need to change the way of producing and consuming finding solutions that enable consumers to change their behaviours in terms of buying clothes cosmetics and food in the sufficient and right quantity for preventing incorrect storage and compact waste. Therefore, it should be necessary to drop across the linear economy based on the taking, making and producing waste adopting the circular economy based on redesigning, reducing, reusing and recycling goods [30]. In conclusion our proposed to use natural polymers for producing biodegradable t issue- carriers, innovative cosmeceuticals, nutraceuticals and packaging might be the best way to beat waste and save environment and human health from the plastics waste with its content of toxic chemicals [31]. Plastics on the one hand has transformed the everyday life bringing many

social benefits while on the other hand their usage, disposal and durability caused accumulation of waste in the natural landfills and marine environment with the consequent ingestion by the human food chain of the content they have in toxic chemicals including phthalates polyvinyl chloride, bisphenol-A and other ingredients as previously reported. Therefore, the necessity to go on by more research studies deeply verifying the effects these compounds could have on animal and human population [31]. Consequently, the necessity to use more bioplastics and natural polymers such as the suggested chitin and lignin to make biodegradable, plastic-free nanocomposite complexes and non-woven tissues for realizing advanced medications cosmeceuticals and nutraceuticals as reported below [7-16,32-34].

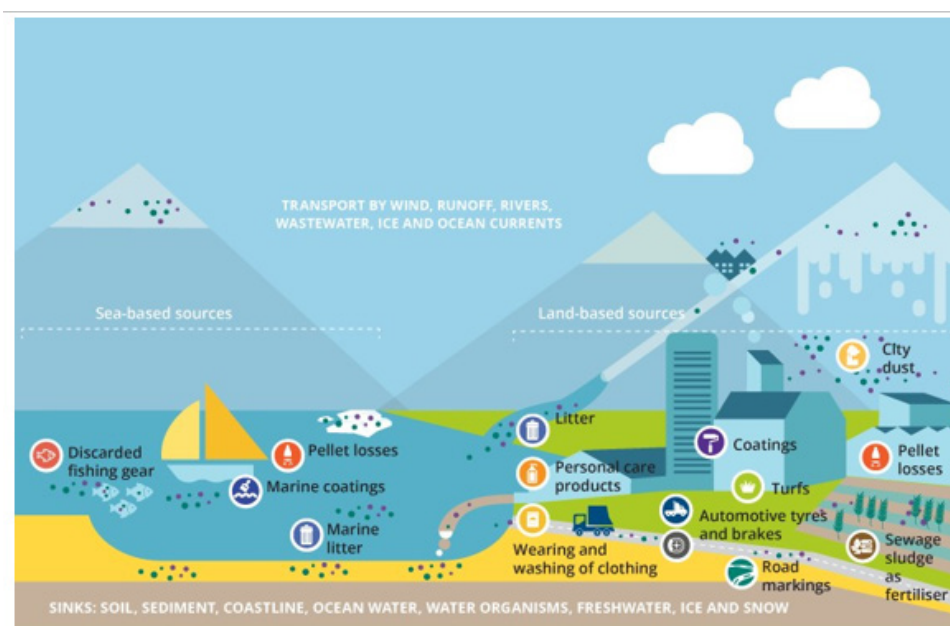


Figure 3: Microplastic debris recovered into land, air and ocean (By courtesy of OECD [3]).

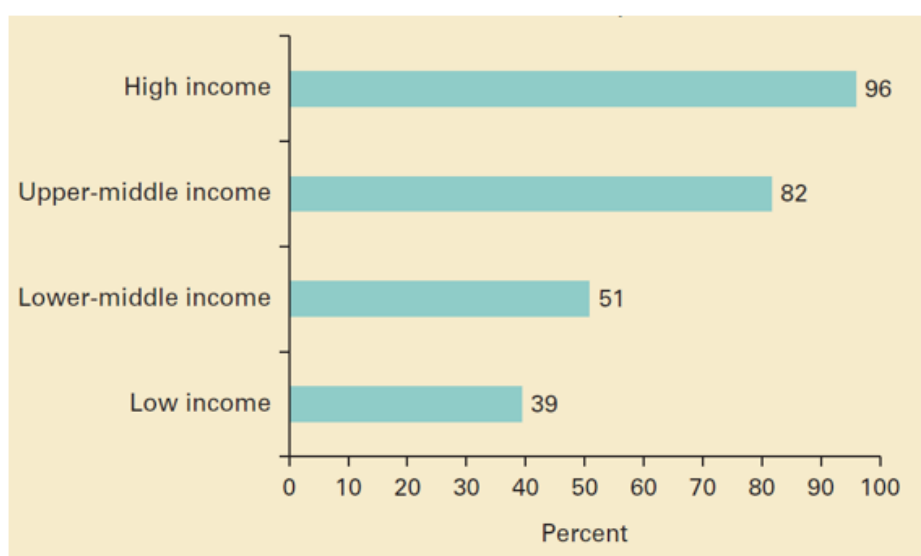


Figure 4: Per cent of global waste collection level of high, middle and low income Countries (by courtesy of the World Bank Group [27]).

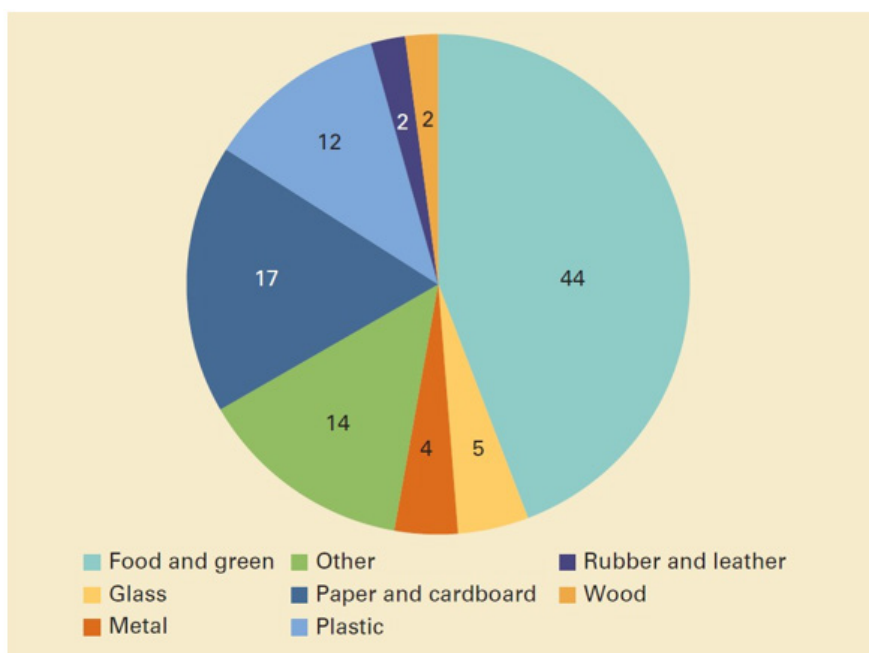


Figure 5: Per cent of different waste material by courtesy of the World Bank Group [27]).

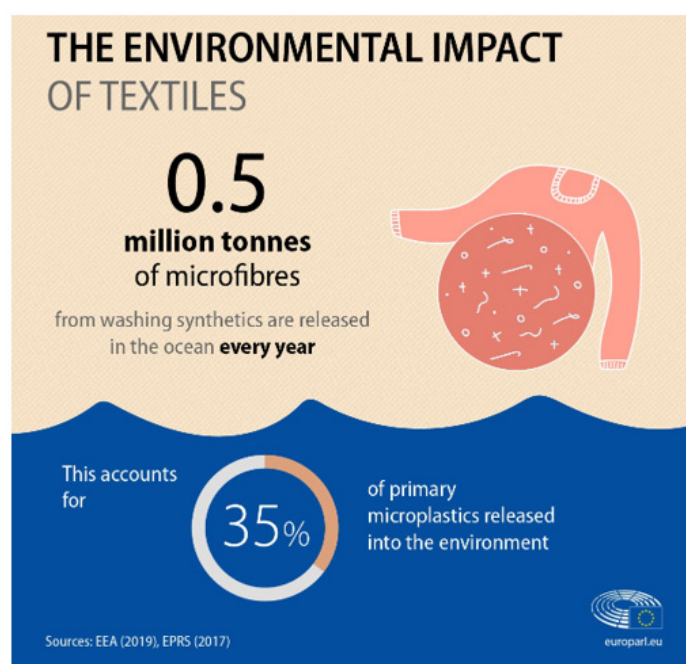


Figure 6: Synthetic microfibers from textiles released every year in oceans (Courtesy of EEA. [22,29]).

Biopolymers, Block Polymeric Complexes and Tissue-Carriers

A major use of natural polymers and natural-based polymers is the request of the majority of scientists and consumers looking for a greater production and use of bio-plastics and biodegradable products which obtainable by these ingredients extracted from food and agro-forestry waste result necessary for safeguarding the natural raw materials. Therefore, the so called green products are considered skin- and environmentally-friendly because safe for humans may reduce the negative environmental impact of the

traditional plastics also [35]. By the biopolymers, its possible to obtain monomers and composite polymeric compounds necessary to make biodegradable micro/nanoparticles and tissue-carriers useful for marking sustainable clothes, agricultural films and innovative medical devices, smart cosmeceuticals, nutraceuticals and different biodegradable packaging materials. In conclusion, the biopolymers obtainable by the valorization of agro-food waste result useful to reduce the use of petrochemical resources with the many derived-made compounds [35]. But how Biopolymers may be classified and produced? They are polymeric macromolecules that produced by living organisms may be directly extracted from

organic biomass-waste and renewable resources or synthesized from chemical polymerization of monomers produced by genetically modified microorganisms (Figure 7); [35]. Biopolymers obtainable in different shapes (linear, branched and cross-linked) and sizes (micro, nano) and from different sources, may have different functions (Figure 1 & 7). However, naturally occurring or man-made all the polymers are based on differing monomeric units covalently bonded to form larger structures made by repeating building blocks. Generally, they are classified in three main classes as reported in Figure 7; [35]. Thus, while synthetic polymers contain a random molecular mass (polydispersity), the natural ones are all alike with the mass containing similar sequence and number of monomers (mono dispersity). Regarding their production, these

macromolecules may be obtained from different sources (i.e. agricultural, forest and marine) and by different routes such as:

1. Direct extraction of biopolymers from plants and fishery waste (i.e. polysaccharides and macromolecules (starch, lignin, chitin etc))
2. Chemical processes: hydrolysis of biomass to obtain monomers (sugars) transformed in turn in building blocks such as polylactate and polyesters.
3. Direct polymerization of monomers into a finished product by microorganisms, including bacterial cellulose and polyhydroxyalkanoates.

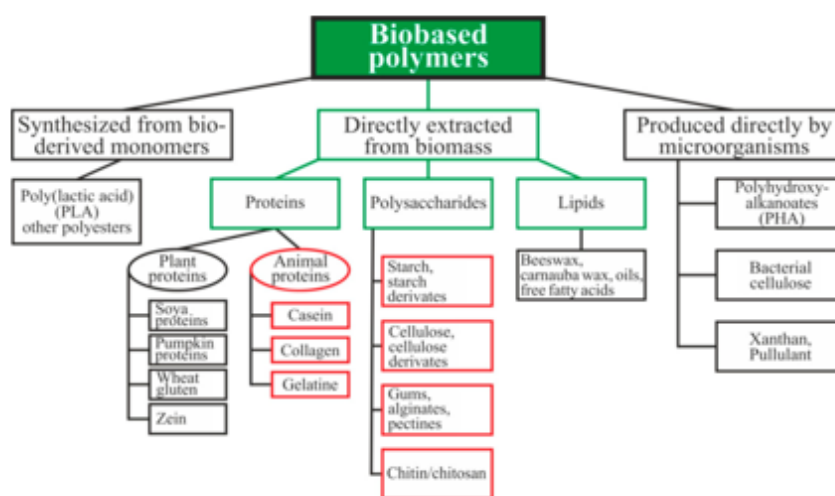


Figure 7: Biobased polymers (courtesy of Balart et al. [35]).

However, natural polymers such as chitin and lignin have attracted much attention because for their easy availability from waste, low cost, unique properties such as bio-degradability, bio-activity, bio-compatibility, film-coating ability, non-toxicity are considered skin- and environmentally friendly [35,36]. Moreover they result of great utility because, avoiding the use of non-biodegradable petrol-derived products and reducing the consumption of natural resources may increase the sustainable production of biobased polymers and goods [37]. For all these reasons biopolymers have a wide range of use by many industries including, clothing, food, cosmetics and medical products also because useful to make specialized scaffolds which easily interfacing with biological systems may act as support of human cell to build up different biomatrices [36]. Unfortunately, biopolymeric materials for textiles are not competitive for industrial applications because of their limited production-volumes with high cost. However, it seems possible to create innovative fashion combining biopolymers with other synthesized hydrocarbon-polymers by the use of the so-called green chemistry [37].

Tissue-Carriers

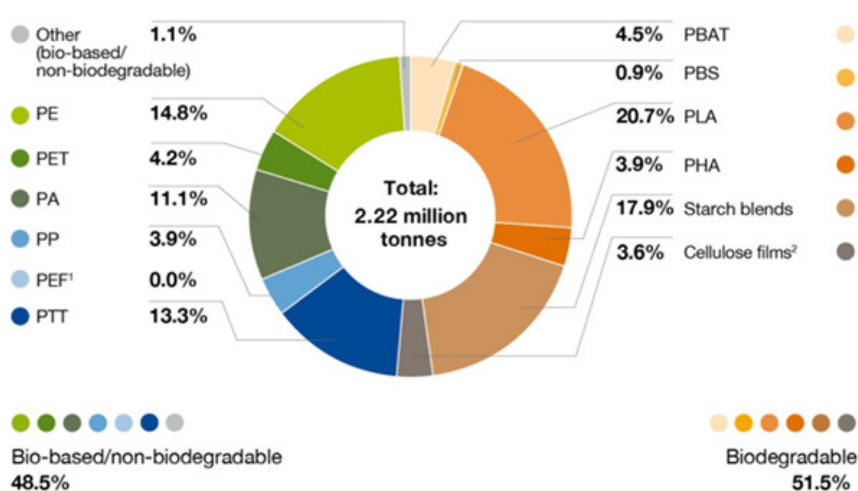
Regarding films and non-woven tissues to be used as carriers and packaging for medical devices, food, nutraceuticals and

cosmeceuticals. Our research group realized micro nanoparticles (NPs) and innovative matrices utilizing chitin nanofibrils and nano lignin [7-16]. Thus as previously reported the biopolymers chitin nanofibrils (CN) and nano lignin (LG) have been mixed by the ionotropic gelation method in water solution to obtain the CN-LG block polymeric micro/nanoparticle complexes which bound successively to the biodegradable tissues fibers have been utilized as novel active carriers (Figure 2); [6-8]. These smart innovative carriers applied on the skin and easily hydrolyzed by the human enzymes to their natural active polymers (chitin and lignin) have shown to possess interesting antioxidants, anti-inflammatory, immune modulates and skin-repairing activities [6-16,32-34]. Moreover, it's to underline that both the polymers have been used at their micro/nano dimension because for their higher surface-area-to-volume ratios, compared to the bulk materials. This size has shown to improve the effectiveness of the final product [6-26,32-34]. It has been shown that the topical administration of micro/ nano scaled delivery systems has the possibility to combine the advantage of a deeper penetration with a better local targeting being also cause of an increased effectiveness, differentiating the relative activities [37]. Thus on the one hand it has been used chitin nanofibrils, because the polymer at less than 40 milli microns size has shown to have an anti-inflammatory response by the

production of the anti-inflammatory Interleukine-10 (IL-10), while its intermediate at 40-70 millimicron size induced production of proinflammatory cytokines including IL-17 and IL-23 [38]. On the other hand, it has been used the active ingredient glycyrrhizin acid that encapsulated into the innovative carrier CN-LG complex has shown to increase its solubility, greatly ameliorating bioavailability and the pharmacological effectiveness, according to the activity shown when carried by micro emulsions [39]. At this purpose the different active ingredients, encapsulated into the CN-LG complexes have been selected and used to characterize activity, effectiveness and safeness of the realized tissues [9-14]. Therefore in our opinion, it might be possible to partially solve the many problems created by the excessive production and use of non-biodegradable plastics fossil-made products also, becoming to utilize the proposed tissue-carriers and increasing the utilization of the so-called biodegradable bioplastics to produce biodegradable packaging. Fortunately, it has been reported from different research companies that the new natural- derived polymers ranged in 2022 a global production of 2.2 million tonnes, should increase significantly to 6.3 million tonnes by 2027 by an estimated compound annual growth rate (CAGR) between 10 and 23%. However, just to better understand this market it is necessary to know that bioplastics are represented by two groups of different composite polymers: biodegradable plastics including polylactic acid (PLA), polyhydroxyalkanoates (PHA) starch blends and others that account for more than 51% and non-biodegradable plastics such as bio-based PE (polyethylene) PET (polyethyleneterephthalate) and others accounting for 48.5% of the global bioplastics as reported in Figure 8; [40]. With a view of the regional capacity development 25% of the bioplastics global production is actually located in Europe while Asia-Pacific, representing today more than 41%, has been predicted to increase its productive capacity to almost 63% by 2027, becoming the

major worldwide production hub [40,41]. Regarding the market production by segment, bioplastics are used prevalently to make, catering, consumer goods, electronics, agriculture/horticulture products and coatings, as well as textiles, automotive & transport or building & construction all materials and packaging that represent the more increasing segments (Figure 9); [40]. Fortunately, for maintaining an environmental equilibrium with a healthy life, it is useful to know that land, used to produce bioplastics will remain at very low levels (0.8 million ha in 2022 and 2.9 million ha in prevision by 2027) showing no competition with the necessity for the global agricultural area including feedstock for pasture (3.3 billion ha) food & feed (1.4 billion ha) biofuels (200 million ha) and relative produced materials (106 million ha). Unfortunately on other hand, the global plastics production valued in 2021 to be more than 390 million tonnes has been projected to grow by a CAGR of 3.7% during the forecast period 2022-2030 while on the other hand bioplastics represent today less than 1% only. For all these reasons and in our opinion the proposed final cosmeceutical-tissues could represent an interesting approach for utilizing biodegradable products being made by natural biopolymers and packed by the same biodegradable specialized paper used for the medical gauze. Therefore, these smart tissue-carriers are not only 100% made by biodegradable polymers but also embedded by natural and different active ingredients selected to realize the innovative skin- and environmentally-friendly cosmeceuticals and nutraceuticals. They have been conceived to make the same products requested from consumers who are dreaming to range beauty and health by the so called Beauty from within [10,14,42]. They are made of biopolymeric carriers and natural active ingredients free of water preservatives, emulsifiers, fragrances and chemicals, having shown high effectiveness and safeness also [10,32-34,42].

Global production capacities of bioplastics 2022
(by material type)



¹PEF is currently in development and predicted to be available at commercial scale in 2023. ²Regenerated cellulose films

Source: European Bioplastics, nova-institute (2022). More information: www.european-bioplastics.org/market and www.bio-based.eu/markets

Figure 8: Global production capacity of bioplastics 2022 by material type (Courtesy of European Bioplastics [40]).

Global production capacities of bioplastics in 2022 (by market segment)

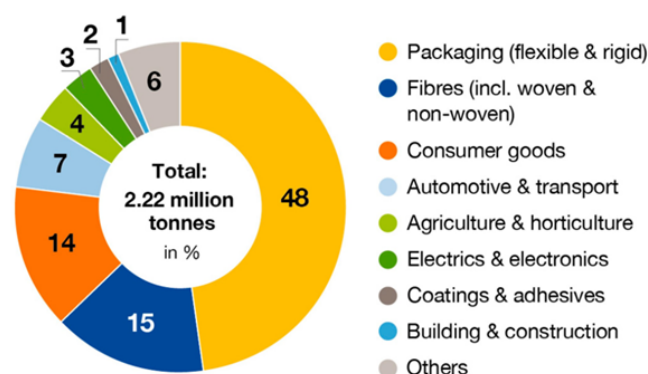


Figure 9: Global bioplastic production by market segment by 2022 (Courtesy of Europea Bioplastics [40]).

Conclusive Remarks

Consumers, especially after the COVID-19 pandemic and the more intensive use of skin care want to know how they're spending their money in the right way. Consequently, they are increasingly prioritizing and purchasing products characterized for effectiveness and safeness and first of all recommended from either dermatologist plastic surgeons and all the doctors working in the aesthetic-healthy field [42,43]. People are looking and willing to pay more for natural skin- and eco-friendly products rich not only of active ingredients and known for effectiveness and safeness, but because promoted and tested by scientific studies [42,43]. According to these requests, we are proposing the use of innovative cosmeceutical-tissues which may be applied on the skin and/or taken by oral route as functional food or diet supplements (nutraceuticals) being able to enhance the skin wellbeing and the global-healthy appearance [33,34]. Consumers desire that these innovative products should have the possibility to protect themselves against the global ageing effects respecting and maintaining the skin integrity by a combined immune boosting-activity balanced by the micro bioma activity also [10-16]. Additionally, people are demanding for an integrated wellness where they may work and play in peace. Thus, living in a healthy environment. Therefore, the necessity to realize the circular green economy slowing down or possibly eliminating the actual air and water pollution for going versus production and consumption at a zero waste, characterized by a clean and healthy environment. Air pollution is associated worldwide with about 3.3 million premature deaths per year with the indoor ones causing nearly 4 million deaths while the water pollution is the cause of 80% of diseases and 50% of children's deaths [44-46]. At this purpose it isn't to be forgotten that indoor air pollutants representing around 50-80% of human exposure to nano particles (NP) (10,000 to 240,000 NP/mL air) compared to outdoor air pollutants (estimated 10,000 to 50,000 NP/mL air) (Figure 10); [44-48] are provoking every year around 7 million of global deaths (Figure 11); [49]. Thus the necessity to produce goods according to the circular economy rules utilizing skin- and eco-friendly products as the proposed cosmeceuticals- and

nutraceuticals-tissues at zero waste and plastic free [8,32,33,50,51]. These cosme-nutraceutical products, made by smart biodegradable polymers used to manufacture innovative carrier-tissues seem to provide nutritional and protective benefits to skin and mucous membranes, because of the similarity of their scaffolds with the natural ECM components. Consequently, they may be used to revitalize and rejuvenate the skin and body's structures, combining the topic effectiveness to the and systemic ones. Therefore these specialized and well-studied tissue-carriers, made by human and eco-friendly biopolymers alternative to petrol-derived ones, result useful for manufacturing the reported active cosmeceuticals and nutraceuticals [9-11,32-34]. At this purpose the selected natural polymers [51,52] including chitin and lignin complexed each other at their nano size to form micro/nano particles bound to the fiber-tissues seem to represent the best option to produce biodegradable and eco-compatible carriers alternative to the actual emulsions for producing cosme-nutraceuticals [11-14]. Therefore these innovative skin- and environmentally-friendly products result in line with the request of consumers, who are looking for nature-derived innovative and advanced products considered necessary to obtain the so-called "Beauty from Within" [11-14,53-59]. As previously reported the realized tissue-carriers embedded by the right "actives" may be useful to "encapsulate" antioxidant and immunomodulant ingredients useful to help the skin to restore and balance some of its structures altered by the aggression of the environmental xenobiotic chemicals and UV radiations. On the one hand, the many -OH groups present on chitin-lignin block polymeric micro/nano particles establishing hydrogen bonds with molecules of water could contribute to maintain the skin hydration and the re-balancement of the immune system stimulating the pro-collagen synthesis. On the other hand, the nano-sized active ingredients, such as glycyrrhizic acid or lutein, embedded into the polymeric tissues may improve bioavailability, stability and protection of the target tissues, acting as antioxidant and anti-inflammatory ingredients [6-16,60-63]. They, easily released from the carrier because of their nano dimension may penetrate throughout the skin layers having the possibility to modulate and regenerate the cells functions [13,14,60-63]. In conclusion, the use of the reported

innovative carrier-tissues, embedded by selected biopolymers and made by a novel safe and patented technology open the possibility to manufacture biodegradable clothes, cosmetics, food and other goods. They avoiding the production of waste and toxic compounds can help to maintain the earth' natural raw materials and biodiversity for the future generations. Moreover, the realized nutri-cosmeceuticals result skin- and environmentally-friendly

having shown to possess sustainability, effectiveness, safeness and transparency having also the capacity to connect appearance and beauty with a body global health. In conclusion, the aim of our future research group is and will be focused on the use of selected biopolymers and active ingredients obtained by the food and agro-forestry waste to make innovative smart carriers.

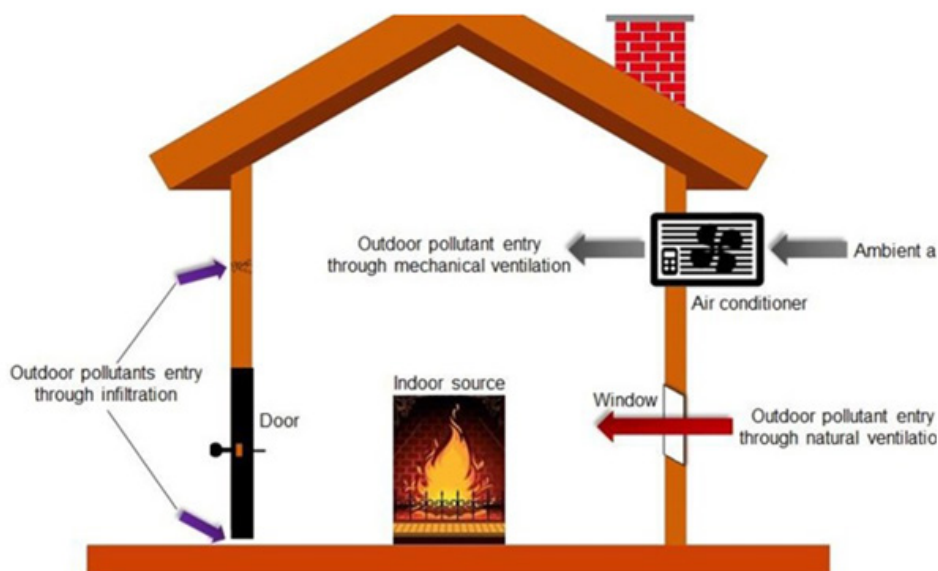


Figure 10: Outdoor and indoor pollutants (Courtesy Leung [46]).



Figure 11: Air pollution' Deaths (Courtesy of WHO [49]).

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