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Biobased Tissues for Innovative Cosmetic Products: Polybioskin as an EU Research Project

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Introduction

Biological material science is strictly connected with the regenerative medicine and tissue engineering, the latter needing to use biomaterials with the possibility of interacting with the body in a specific and predictable manner [1]. Thus, the necessity arises to study in greater detail structure and physicochemical properties of these materials to investigate, for example, how nonwoven tissues, made by natural polymers, may interact with the biological systems, stimulating the growth of cells they have contact with. The ability of cells for adhering to the nonwoven tissue, indeed, depends on the nanoscale spacing between possible binding sites of the dressing and/or the presence of additional factors, such as integrin receptors.

Integrins play a critical role in adhesion to the native extracellular matrix (ECM), by generating force, translating mechanical cues into biochemical signals and communicating with growth factor receptors [2]. Developing multifunctional, biodegradable and eco-compatible nonwoven tissues with biological materials and the suitable hierarchical structure is the goal of the PolyBioSkin European research project, started in June 2017 with its kick-off meeting hold in Castelldefels, Barcelona, Spain. The group working on this project (Figure 1) consists of material scientists, engineers, biologists and biomedical doctors, is represented from academia scientists: the Consorzio Inter Universitario di Scienza e Tecnologia dei Materiali (INSTM, Italy), the University of Westminster (UK), the Association pour la Research et le Development des Methods et Processes Industrials (ARMINES, France), the Tehnoloski Fakultet Novi Sad, Serbia) and the University of Gent (Belgium); six SMEs are also participating to the project: Innovacio I Ricerca Sostenibile (IRIS, Spain, project coordinator), Bioinicia (Spain), Fibroline (France), Texol (Italy), MAVI Sud (Italy) and Exergy (UK), as well as the European Bioplastics Association (Germany).

Figure 1: Picture of Polybioskin Consortium.

The aim of the project is to study and fabricate biodegradable and innovative bio-nano-fibers, for making skin- and environment-friendly nonwoven tissues, useful to industrially produce diapers, facial beauty masks and advanced skin medications. It is to remind that nanofibers have properties quite different from those of larger scale fibers, such as microfibers, because of their particular morphology, extremely high surface-to-volume-ratio, and nanometric size, which enable unique characteristics, including mechanical, optical and other physical properties [3]. To produce biodegradable fibers and tissues, the PolyBioSkin group will use two main classes of bio-based polymers:

a. polysaccharides among which cellulose, starch, chitin and chitosan, selected for their high absorbency power and peculiar functionality of antiinflammatory and antibacterial activity, and

b. biopolyesters ,such as polylactic acid (PLA) and polyhydroxyalkanoates (PHAs) obtained from bacterial fermentation.

All the selected ingredients are biodegradable, skin-friendly and obtained from renewable resources.

Natural Fibers and Nanotechnology

In the world of bioengineering tissues, the use of natural fibers as biomaterials has been deeply explored due to their various advantages including their robustness, degradation and the ability to mimic the ECM in three dimensions. For the same reasons the use of nanofibers became of great interest due to their particular and unique physicochemical and biological properties. For their nano-dimension, these natural fibers can provide high durability to the nonwoven tissues, possessing an extremely high surface-to-volume ratio, and high surface energy, unique optical and mechanical properties, with an improved biocompatibility and affinity for the skin [4].

A nanofiber is generally defined as a fiber having a diameter lower than 100nm of and an aspect ratio higher than 100. Moreover, starch-based and chitin-based nanofibers are becoming to be used as non-toxic bio-stimulating agents. They possess beneficial properties and are produced by natural raw materials by sustainable processes, compared to the traditional polymers obtained from fossil resources. At this purpose, it is to consider that carbohydrates, selected and used as polysaccharides to prepare PolyBioSkin fibers, have important roles in living organisms, including energy transportation and production. When combined together to form polymers, they can function as long-term food storage molecules as well as protective membranes and support cellular structures. Moreover certain complex carbohydrates, known as glycans, mediate the interactions between recognition molecules, thereby contributing to the formation of a complex molecular mesh at the cell surface and in the ECM (Figure 2).

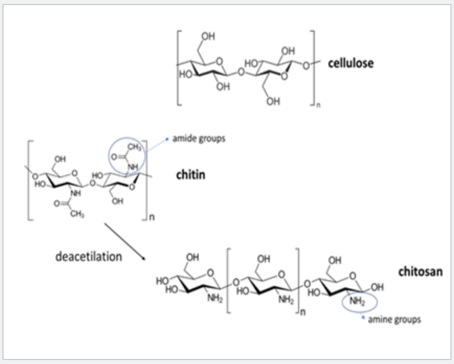

Figure 2: Chemical structure of cellulose, chitin and chitosan.

Reproducing activity and function of the skin ECM, represents an important goal for all cosmetic products, which, by acting as effective moisturizing and anti-aging agents, have to be capable of turning the cell-cell and cell-matrix interactions at biological level. In this view, nanotechnology offers many advantages compared to the conventional processes in term of economy, energy saving, eco-friendliness and controlled release of substances under defined conditions, although much efforts have to be made to increase their production scale up and to lower their industrial costs and risk assessment issues [5].

Global Cosmetic Market and Eu Polybioskin Project

The global cosmetic market, evaluated at US$ 445 billion, has shown an average growth approaching 6% in the years 2016 and 2017 [6]. Specifically, it had a growth of 10.6% in Indonesia, 9.2 % in India and Brazil, 8.7% in Africa (Saudi Arabia and Egypt) and 6.8% in Russia, 4.6% in Australia, 4% in USA and 1.8% in Europe. In conclusion, the highest cosmetic consumption has been registered in South-East Asia with 31.7%, USA and EU 20.6%, South Americas 13.5%, Russia, Africa and Australia 6.7%. The purchasing decisions of this market are increasingly motivated by the request for healthy green consumption, where the natural is wider-encompassing to include products with eco-credential, sustainable sourcing and clean labels. Additionally, a customized and personalized beauty has to be reinforced by the smart technology that is catapulting forward desired lifestyles.

To this purpose, beauty facial masks, made of biodegradable nonwoven tissues, are a target product for PolyBioSkin project. The goal is to entrap different active ingredients in the nanocomposite fibers to obtain cosmetic products with a different structure, namely, non-woven tissues as an alternative to traditional emulsions utilized to beautify the facial skin. These innovative cosmetics are designed to act by slowly releasing the active compounds at the different skin layers, in different time, depending on the designed activity of the final product will be based on smart technologies and formulations, as currently required by customers and consumers. Therefore, the relationship between the material hierarchical structure of the tissue and the skin properties and functions will be explored to create new families of cosmetics based on smart biological systems. Indeed, such facial beauty masks, free of emulsifiers, colors, preservatives and other petrol-derived chemicals, will be based on natural biological materials. The attention to both the renewable origin of materials and the end life of cosmetic products, often not kept into account by EU or local regulation, is therefore, an interesting part of the project activities with the aim of promoting a responsible behavior towards environmental concerns.

Facial beauty masks market

According to Transparency Market Research [7], the global facial sheet mask market is expected to reach US$ 336 million by 2024 with a year growth of 8.7% from 2016 to 2024 into a global textiles market that will growth at a CAGR of 3.3% and a value of US$ 160.38 billion. The consumers' growing demand for face masks, is connected to the request of innovative cosmetics easy to use, effective as anti-aging products, offered in the market at more accessible prices. Analysts have identified emerging economies such as Indonesia, Argentina, China, Brazil and India as lucrative markets for facial sheet masks for the next years. The strengthening economies of these countries, with growing

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disposable incomes and changing lifestyles, are expected to give a significant contribution to the rising revenue of the global cosmetic market. These markets may benefit from the increasingly strong presence of manufacturers in the region [6].

According to Mintel & Asia Personal Care & Cosmetic Market Guide [8,9], in China the facial mask market is growing at an impressive rate, having registered an annual growth of 29% in the period 2011- 2016. Both in China and Hong Kong, 88% of urban females aged in 30-39 used a sheet mask overnight twice a week while 69% of these consumers in the evening. In Singapore, skin care regimes including facial masks that incorporate healing and anti-aging agents, such as vitamin C, Argan oil serum, green tea, ginseng, lotus and pomegranate extracts, mandelic acid and water-melon, became very popular by capturing the consumers' imagination. According to Kwek [8], senior and Insight Analyst of Mintel, "the popularity of sheet masks remains high and will continue to do so as a result of the production's ability to offer consumers skin care benefits in a convenient manner and shirt turnaround time". At this purpose, it is to underline that the beauty facial masks developed within the PolyBioSkin project will be innovative and smart. They will be innovative because of the active ingredients which, bound directly to the nonwoven dry fibers, will be released only when in direct contact with a wet skin and according to the function they have been designed to perform. They will be smart, since the ingredients will be slowly released during the applied time of~20-30 minutes, according to the composite used to make the nonwoven tissues.

Additionally, as previously mentioned, PolyBioSkin beauty masks will be biodegradable (with a potential end life with a lower impact on environment than current products), auto-preserving and free of any added chemicals. On the other hand, it is to remember that the majority of the beauty masks in the market are made of tissues imbibed with solutions or emulsions containing emulsifiers, preservatives, different chemicals and sometime dyes. Moreover, up today the majority of the tissues used are made of petrol-derived fibers and therefore often non-biodegradable and usually non-recyclable because of the commingling of materials and compounds biodegradable and not.

Basic Raw Materials Used by the Project

PolyBioSkin project will focus on two main classes of bio-based polymers, polysaccharides and bio-polyesters. Among these compounds, cellulose, starch and chitin/chitosan together with PLA and PHA have been selected for their natural origin, biodegradability and biocompatibility.

Starch

Owing to its complete biodegradability, low cost and renewability, starch is a promising candidate for developing sustainable polymers and therefore nonwoven tissues [10]. Starch-based polymers are used, for example, to make medical scaffolds for bone-tissue engineering because of their biocompatibility and porous nature, which allows blood-vessel proliferation during bone growth. Due to the many hydroxyl groups on its chains, starch can easily interact with other polar polymers through the formation of hydrogen bonds and can be also oxidized or reduced to modify its structure and functionalities. As a consequence, it is possible to improve the final physicochemical properties of composites made with other natural polymers. At this purpose for example, to improve the compatibility between the hydrophilic starch and the hydrophobic PLA or PHAs suitable plasticizers are often used. On the other hand, the starch/chitin blend exhibits good film forming properties due to the intermolecular hydrogen bonding, formed between the chitin amino and amide groups on chitin and the hydroxyl groups on the backbone of starch.

Crystalline cellulose

Cellulose, as an important structural component of the primary cell walls of plants and many algae, is a polysaccharide consisting of a linear chain of hundred-to-thousand D-glucose units, present in crystalline micro-fibrils with an amorphous part at the surface [11](Figure 2). As in the case of chitin in crustaceans, cellulose in plants is combined with other compounds to produce strengthened tissues. Compared to chitin, more and many crystalline structures (Figure 3) are known in this sugar-like compound, corresponding to the location of hydrogen bonds between and within strands. Its properties depend on the chain length and the degree of polymerization. About the medical activity, different cellulose-derived polymers are used such as, cellulose acetate utilized as coating for pills or as microspheres to improve the gut retention of anti-diabetic drugs.

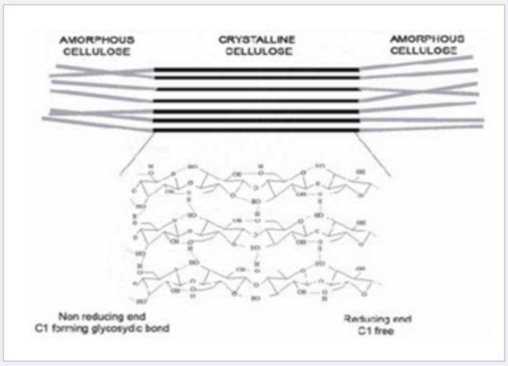

Figure 3: Crystalline structure of cellulose.

Chitin

Chitin, unbranched polysaccharide with unique structural, mechanical and thermal properties, is a derivative of glucose with an amino group substituted at carbon 2 (Figure 2), comprises repeating monomer chains of N-acetyl-D-glucosamine [12]. The balanced existence in its molecule of the hydrophilic hydroxyls and acetyl amino group, combined with hydrophobic pyranose rings, gives to chitin amphoteric character, allowing uptake of both polar and non-polar liquids effective, for example, for oil

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separation [13]. This natural polymer, widely distributed in nature as a supporting structure of crustaceans and insects, surrounds the plasma membrane of bacteria, plant, fungal and algal cells and provides mechanical support and strengthening. In the same way of cellulose, chitin is organized into amorphous and crystalline structures that may be separated each from the other [14] (Figure 4). However, unlike cellulose, chitin has an innate rigidity and its molecule can be partially or totally de-acetylated to produce chitosan (Figure 2).

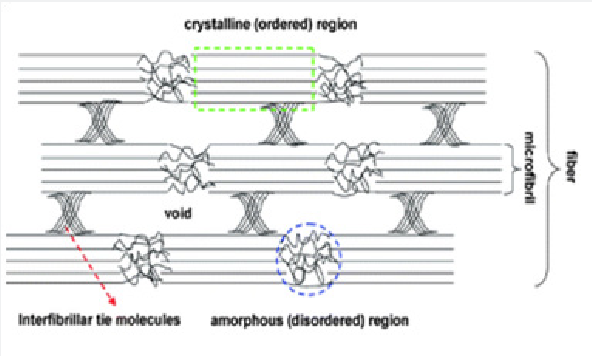

Figure 4: Schematic crystalline structure of chitin [28].

According to Raabe et al. [15], chitin chains in lobster cuticle are arranged in an anti-parallel fashion forming alpha-chitin, which in turn forms nanofibrils assembled into honeycomb shaped arrays. The crystalline portion of these nanofibrils, covered by positive charges, may be used as a filler to strengthen the fiber composite structures and to produce micro/nanoparticles with electronegative polymers. Such complexes can be exploited as delivery carriers. Moreover, although not yet recovered in mammals, chitin has a remarkable compatibility with living tissues and has shown to increase and accelerate the skin healing process without formation of hypertrophic scars and keloids when used as nonwovens, tissues. These tissues offer micro/nano scale channels for the migration of host cells and nutrients into the matrix structure [16]. their antibacterial and skin regenerative effectiveness shown by these scaffolds seem to be connected with ability to increase the production of defensins and to balance the production of metalloproteinases (MMPs) [17].

PHAs

PHAs are a family of biological polyesters containing (R)-hydroxyalkanoic acid monomer units. Consisting of short 3-5 carbon atoms chain-length or medium 6-14 carbon atoms medium-chain length [18,19], they are produced from natural sources, like PLA [20], by bacterial fermentation methodology (Figure 5). Unlike PLA, their production is based on biotechnology as they are produced by bacteria. In particular, PHAs are synthesized as deposit inclusions into cytoplasm of gram-positive and gram-negative bacteria, but they are also ubiquitous in plants and animals. Their accumulation is a natural way for bacteria to store carbon and energy, when nutrient supplies are unbalanced.

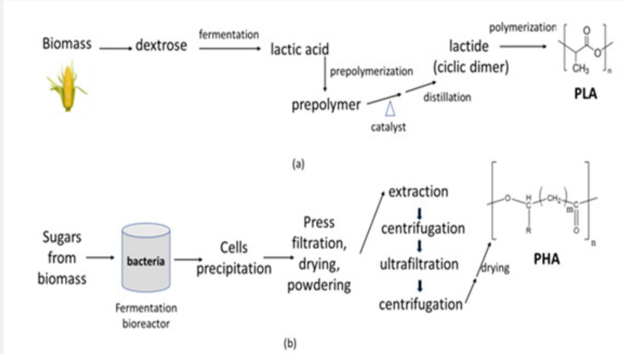

Figure 5: Production of PLA (a) and PHAs (b) by industrial technology.

An important distinguishing feature of PHAs is their biodegradability that depends not only on the microbial activity of the environment, but also on the exposed surface area, moisture, temperature and molecular weight, as well as crystallinity and nature of monomer units. Since PHAs are completely biodegradable into water and carbon dioxide by environmental microorganisms and easily thermo processable, they are attractive. as biomaterials for applications in conventional medical devices, drug delivery and tissue engineering [18-22]. However, the mechanical properties and processability of these polymers mostly depend on the composition of the monomeric units of these polymers and molecular weight.

PLA

PLA is produced by polymerization of lactide obtained by the fermentation of starch to lactic acid (Figure 5). The production technology of PLA is already at a mature stage and the price of this bioplastic is only slightly above the price of petro-based commodities; hence, its access and expansion in many different markets is forthcoming. Regarding the biocompatibility, PLA was widely used in biomedical field, for instance in surgical applications, thanks to its bioresorbability. This polymer is quite rigid and brittle; therefore its properties must be modulated by using proper additives to make it more flexible, such as other bioplastics or renewable plasticizers [23-24], as well as natural fibers [25-26] to make composites or nano-composites. PLA can be used in fiber formulations for tissue based beauty masks, both woven and nonwoven, but also in beauty mask produced by using plastic films or sheets as a support. Moreover, it can be applied in rigid or biodegradable containers for cosmetic packaging, beauty masks, as well for other cosmetic uses. PLA results compostable in industrial composting plants in dependence of dimension and thickness of PLA items; hence, compostable PLA based products can be easily disposed in the organic fraction of waste. Unlike PHA, PLA is currently not compostable in home composting conditions or in marine environment.

Conclusion

Textiles and nonwoven tissues made of petrol-derived polymer fibers, interact with the skin functions in a dynamic

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pattern, and could provoke injury and inflammation. The safety aspects of polymer fibers, related to their biological characteristics, such as biodegradability, biocompatibility and antigenicity, indeed, are of great importance for biomedical applications [20,23,27]. Therefore, the cosmetic market is driven by the necessity to use bio-based polymer fibers and nonwoven tissues, characterized for their protective effectiveness and free of undesirable side effects such as cytotoxicity or allergic, sensitizing and irritating reactions. These natural tissues, the production of which has been increased by the use of nanotechnology, are made of bio-fibers able to sense and hinder different stimuli elicited from outside and inside the skin. The production of these fibers and non-woven tissues is the aim of the PolyBioSkin project that specifically is focused on the development and fabrication of innovative sanitary, cosmetic and wound care products provided with innovative behavior and functionality. Of course, the success of the proposed products will be connected with the advanced composite materials produced, their human safety and effectiveness, to be tested both in vitro, and in vivo, taking care of the environment natural equilibrium.

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