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Hair surface engineering: Combining nanoarchitectonics with hair topical and beauty formulations

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ABSTRACT

In this review article, the recent approaches to nanoarchitectonics on hair are briefly overviewed. Different types of nanomodifications can be used for the cosmetic and medical applications to provide the aesthetic improvement or treatment of hair and scalp diseases. This article will discuss in detail the various aspects of nanostructures using to enhance the effect of daily care products, to increase the efficiency of penetration of active substances into the hair structures responsible for their regeneration. In general, nanocoatings are promising in the field of application for improving the structure and aesthetics of the hair shaft, as well as for imparting various beneficial properties to hair or for protection from harmful environmental factors. Finally, the approaches of nanoarchitectonics and regenerative medicine will be overviewed from the point of effective methodology for the regeneration of hair follicles creation.

1. Introduction

Nanoarchitectonics

Nanoarchitectonics, a novel concept of directed and tunable fabrication of nanostructured materials through combining self-assembly and self-organization, has recently emerged [1], forming paradigm by combining nanotechnology with the different research fields including biological sciences [2]. For biomedical applications, the nanoarchitectonics methodology is beneficial to produce a range of biocompatible, biodegradable and non-toxic multi-functional materials and nanomechanical structures from nanoscale unit components through the selection and combination of chemical, physical and bio-related processes [3]. Surface modification can be carried out using the method of layer-by-layer deposition of various polyelectrolytes, both natural and chemically synthesized. This method has found wide application in techniques for the formation of functional surfaces, cell modification and the fabrication of carriers for drugs [4–6]. Among numerous other examples, the special interest of nanoarchitectonics is in biomedicine, where a plethora of methods and materials has been developed in recent years. In particular, though largely unnoticed, the hair-related aesthetic part of human health and well-being, directly and indirectly determining the quality of life and the level of personal anxiety, can be easily improved through nanoarchitectonics approaches, resulting in a novel direction of hair surface engineering, which we cover in this review article.

The structure of hair

Human skin, apart from areas of glabrous skin (e.g., non-hairy skin on the palms), is covered with follicles, which produce pigmented thick terminal and short, thin non-pigmented vellus hair [7]. In terms of physiology, the presence of hair on the human head and body does not currently play a significant physiological role, being primarily a rudiment, however the overall condition of hair can be an indicator of human health [8,9], furthermore, the good appearance of hair is a prerequisite of high self-esteem in majority of people. Hair is a skin appendage that consist of the hair shaft, a protein filament (around 20–180 μm in width) and root grows from follicles found in the dermis [8]. The human hair shaft has prominent microstructures that can be divided roughly into three zones observed in TEM imaging of hair cross sections (Fig. 1A) [10]. These are the cuticle, an outermost layer that protects the intermediate cortex, and the medulla, a disorganized and open area at the fibre's center [8]. The hair cuticle formed from dead flat cells in several layers laid out overlapping one another as roof shingles (Fig. 1B) [10]. The essential function of the cuticle is protection of the inner cortex. Stability and physical toughness of the cuticle cells is partly provided by keratin filaments, which are held together by disulfide and

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hydrogen bonds. Internal layers composed of elongated keratin-rich cortical cells, which are joined together by an intercellular cement rich in lipids, and keratin-associated proteins that provide the elastic properties of the cortex of the hair shaft [8,10].

Melanin is located in the cortex of hair shaft and provides the specific hair colour. This pigment is produced by skin melanocytes located in the basal layer of the epidermis. The ultimate hair colour is determined by the combination of eumelanin (it is the most abundant melanin in humans, confers brown or black tones) and pheomelanin (responsible for ginger tones in the hair) [11]. Melanin absorbs UV light, and thereby protects the tissues of the deep skin layers from UV radiation-induced damage. Melanin itself can serve both as an anti-oxidant and free radical scavenger (typical for eumelanin) and pro-oxidant (pheomelanin produces free radicals in response to UV radiation), depending on various external factors and potentially its state of aggregation [11].

The medulla of mammalian hair is the innermost layer of the hair shaft that can be continuous, fragmented or absent without altering the hair strength. In the human hair the medulla consists of either single or multiple, unstructured, heavily vacuolated horizontal cells in column on the centre of the hair cortex. Medulla cells develop large inter and/or intracellular spaces during their terminal differentiation. These are air-filled, and provide insulating capacities to hair. [12].

The hair follicle is a complex miniature organ that undergoes regular cycles of involution and regeneration during its lifetime [13]. The hair follicle of the skin consists of the hair bulb, the inferior segment of the hair follicle, follicular dermal papilla and the hair matrix. The hair follicles are attached to sebaceous gland and arrector pili muscle. The dermal papilla of the hair follicle is a niche for mesenchymal cells population and epithelial progenitor cells that regenerate the cycling portion of the hair follicle and generate the hair shaft [14,15]. The hair follicles undergo life-long cyclical transformations, progressing through phases: the anagen (growing phase, lasts 2–7 years), the catagen (involution or regression phase during ca. 3 weeks), and the telogen (resting stage, ca. 3 months) [16]. In addition, the hair follicle regulates the immune response against pathogens, sebum production, thermoregulation, neurogenesis, angiogenesis and wound healing [17]. Arrector pili muscle is a tiny smooth muscle bundle that attaches to the base of follicle at one end and to dermal tissue on the other end. The hair stands vertically after contraction of the arrector pili muscles, for example, when the skin is exposed to cold [17].

Hair cosmetology and pharmacology

Most common interest in hair is focused on hair growth, hair colouration and care, making it among the most important parts of cosmetic and pharmaceutical industries. Hair is also an important biomaterial as an alternative reinforcement for fibre reinforced polymer composites. In the field of material science, human hair found applications as a nano-composite biological fiber [18].

For hair care, the cosmetic industry offers daily care products (shampoos, creme rinse, conditioner), styling products (fixation spray, gels and mousses), products to stimulate hair growth (niacin, panthenol) and dyes. Cosmetic detergents and adjuvants serve to cleanse and to neutralize an excessive negative charge, prevent static electricity in shampoo-washed hair [9,19]. Regular washings lead to mechanical damages of the hair structure, such as partial or complete removal of the endocuticle or the entire cuticle cells. Solar irradiation intensifies the damages to hair cuticle, for instance, peeling of the cuticle layers and the complete cuticle removal, thus contributing significantly to hair fragility and opacity [20]. Permanent hair dyes often inflict a harmful action on cells and organisms that expressed in hypersensitivity reactions [21], cytotoxic and genotoxic effects in the cells [22].

Zinc pyrithione is the most common shampoo active additive for dandruff, seborrheic dermatitis and other more serious hair and scalp disorders treatment. Having also antibacterial properties, it is effective against a number of gram-positive pathogens. For the management of hair loss, the shampoos containing panthenol to aid in hair growth and maintaining of the moisture content have been developed. [23]. A similar pharmaceutical effect is demonstrated by niacin (nicotinic acid), which is regarded as a promising compound to treat female alopecia [24].

Hair disorders

In ancient tombs, the basic structure of the keratin filaments of human hair may remain unchanged over centuries and its essential structural features are extraordinarily stable. However, as a person ages, a change in state of the growing hair occurs that is expressed in decrease of their density (hair loss or alopecia), hair graying, thinning tends (fine-coarseness) and dryness/oiliness of the scalp and the hair [25]. The hair disorders are associated with a number of possible reasons. Nutritional deficiency, in particular, fatty acid deficiency, zinc deficiency, chronic starvation, niacin deficiency and caloric restriction may affect the hair

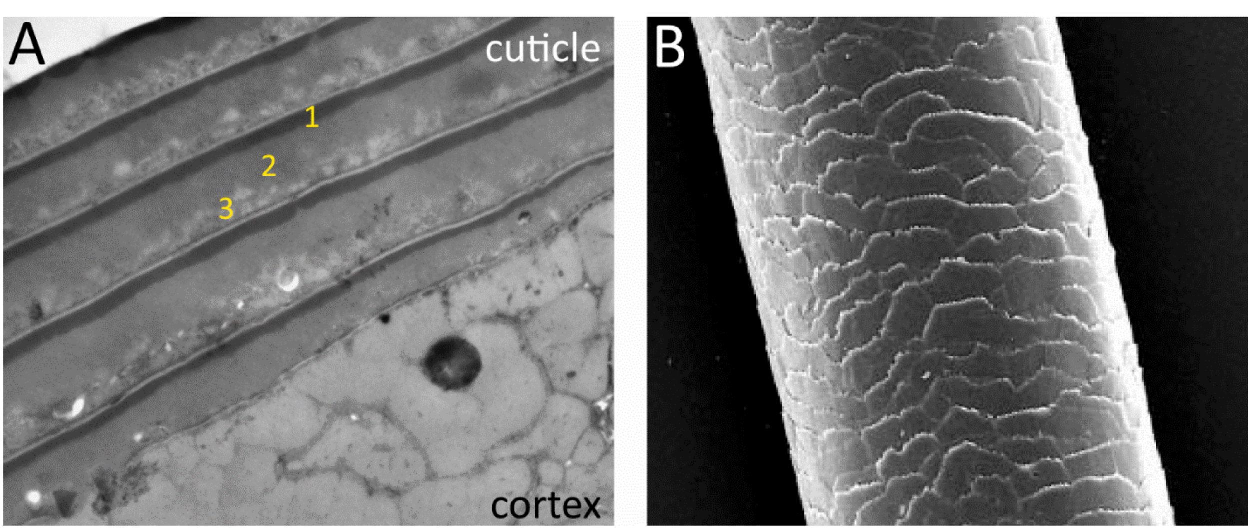

Fig. 1. A - TEM of cross-section of human hair with five overlapping cuticle cells. Each cell has a dense A-layer (1), beneath a less dense exocuticle (2) and the lower layer is endocuticle (3). × 35,000, B - SEM of hair fiber × 400 (Adapted from Ref. [10]. The images are reproduced under Creative Commons Attribution 4.0 International License).

structure and growth. Acute telogen effluvium [26] have potential associations with androgenetic alopecia, diffuse alopecia, female pattern hair loss, and alopecia areata (autoimmune alopecia) [27]. Telogen effluvium quite commonly accompanies the pregnancy, significantly stressful physical or psychological events [28]. The hair follicle structure and hair cycle are subject to hormonal regulation by various hormones such as androgens and their prohormones [29]. Hormonal dysfunction in a combination with genetic disorders are the most common causes of male-pattern hair loss. Hair loss is common and affects up to 50% of males [27]. Other common causes of hair loss are chemotherapy and several drugs [30], HIV [31], hypothyroidism [32], fungal infection [33], autoimmune disorders (e.g., systemic lupus erythematosus) [34], radiation therapy and sarcoidosis [28].

While most of the cosmetic difficulties are temporary and manageable, the problem of hair loss has no fundamental solution. The current procedures for hair loss or, for example, fungal infections treatment need to be frequently repeated, which stimulates the development of new drug formulations.

Nanotechnology-based formulations for hair

In recent years, nanotechnology-based formulations have attracted attention in the cosmetic and pharmaceutical fields. Currently, nanomaterials are used as a functional coatings and carriers that find application in treatment and protection of the hair shaft from external damage, to facilitate penetration of specific active compounds via the follicular pathway [35]. The use of nanostructured materials for topical drug delivery offers controlled release for a prolonged period of time, increasing their retention and avoiding the side effects and irritations caused by the excessive introduction of drugs. These nanomaterials include protein-based [36], natural [37–39] and synthetic polymers nanomaterials [40,41], lipid-based [42, 43], metallic [44,45], silica nanoparticles [46,47], dendrimers [35], clay [48] and carbon nanotubes [49] etc. When using nanomaterials for hair modification, it is necessary to accurately determine the safety for each specific method of their use, as well as take into account the differences in different ethnic groups [50].

Here we overview the using of new approaches of nanoarchitectonics in hair care and treatment.

The search for articles for presented review was carried out in the MEDLINE database through PubMed using the terms that are reflected in the keywords and their combination and related terms.

2. Methods and materials in nanoarchitectonics of human hair

2.1. Nanomaterials for hair cosmeceuticals

Hair daily care

The main purpose of cosmetic products for hair care is maintaining cleanliness and visible health of hair. The visible damage of hair shafts may be induced by mechanical stress (hair brushing and styling), chemical stress (hair dyeing and bleaching), heat stress (iron curling, the hair drying), or photoaging (UV radiation and visible light damage) [51]. The development of novel nanomaterials for hair care has focused on enhanced effectiveness of hair cosmetics through an immediate or long-term effect, better interaction and targeting. In addition, encapsulation of cosmetic components into nanomaterials allows for the delivery of insoluble compounds.

Many carriers for hair nanocosmetics are based on using micro and nanoemulsions, niosomes and liposomes that contribute to the targeted distribution and improved stability of bioactive components [52]. Complex cosmetic products for daily hair care contain not only washing surfactants, but also components for increasing hair silkiness and preservation of hair moisture. Silicone oil, widely used in cosmetic preparations for lubrication, is added to shampoos for a conditioning effect. Silicone oil is designed to target the hair shaft and not for a long-term effect on the hair skin. In conventional shampoos, this oil has high affinity and deposition on the hair scalp rather than penetrate the hair shaft (given its hydrophobic characteristics), which results in a greasy touch after application. From the surface of hair shaft silicone oil easily washed off with the surfactants of the shampoo [53]. Incorporating of silicone oil inside nanomaterials facilitates its diffusion into hair shaft during washing without inflicting any damage to hair cuticle. The oil-in-water emulsification with nonionic surfactants (i.e., Span 80 and Tween 80) was used to prepare thermodynamically stable nanoemulsions with droplet size in the range of 100–700 nm. X-ray analysis together with SEM indicated an increased deposition of silicon from nanoemulsion onto the surface of the hair shaft [54]. Cationic nanoemulsions (emulsions with a droplet diameter less than 100 nm) significantly improved the appearance of dry hair, making it shiny, less brittle and non-greasy [55]. Some authors demonstrated the ability of cosmetic nanocomposition of oxides, silicates, hydroxides, phosphates and carbonates to prevent the appearance of greasy on hair [56].

The adsorption and lubricating properties of a polycation-PEG graft copolymer, PLL-g-PEG, on human hair surfaces were characterized using XPS, AFM, and fluorescence microscopy. Bleached hair samples, PLL-g-PEG-treated hair samples, and subsequently shampooed hair samples were studied. AFM techniques have shown that the adsorption of PLL-g-PEG on the hair reduced the frictional force between the hair sample surfaces and the AFM tip, suggesting that the adsorbed graft copolymers can act as a boundary lubricant on the hair surface (i.e., have conditioning effect) [40]. Epoxy silicone nanoemulsions (with an average particle size of 100–250 nm) prepared by a microfluidization method have been used to treat damaged hair (e.g., hair exposed to chemical processes, such as hair lightening/bleaching, relaxing, dyeing, permanent waving and hair smoothing). Applying of this epoxy silicone nanoemulsion provideed greater strength, elasticity, and fatigue resistance upon repeated brushing compared to the untreated control [57]. Solid lipid nanoparticles (SLN) with the size range from 50 to 1000 nm are gaining the popularity in cosmeceutical and pharmaceutical industries. They are composed of a single layer of shells and the core is oily or lipoidal in nature. SLNs *per se* have UV resistant properties and may using as carrier for 3,4,5-trimethoxybenzoylchitin and vitamin E [58] to enhance UV protection of hair. The hair shaft exposition to ultraviolet radiation leads to the degradation of the cuticle proteins. Weathering of cystine in hair is primarily a photochemical reaction proceeding mainly through the C-S fission producing cystine S-sulfonate residues as a primary end product. Due to photo-protective effect of melanin, protein photooxidation has less intensity in black hair than in light hair [25,59].

Recently, to reduce the photochemical destruction, a nanoarchitectonics-based protective hair treatment based on halloysite nanotubes/keratin composites was successfully developed [48]. Clay nanotubes coated with hydrolyzed keratin produce micrometer-thick protective coating on hair shaft (Fig. 2) [48]. During this study hair samples have been exposed to UV irradiation for times up to 72 h to explore the protection capacity of this coating by monitoring the cysteine oxidation products. This approach presents a promising strategy for a sustainable medical coating on the hair, which remediates UV irradiation stress. In addition, it has previously demonstrated that HNTs can be used as carriers for hair dyes (Fig. 2C, 2D) [60]. The suitability of halloysite nanotubes for daily haircare cosmetics is related to their biocompatibility and low toxicity toward both cells [61–63] and organisms [60,64] safe for the environment that promises their natural origin [48]. A similar coating was recently reported for sustainable anti-lice protection for agricultural (goats), wild aquatic (capybaras) and domestic (guinea pigs) animals, where the insecticide (permethrin) was loaded into pristine or hydrophobic halloysite [65].

Hair colouring

All natural hair colors are due of melanin hair pigments. Melanins are produced by melanocytes in the bulb of the follicle and packed into granules found in the fibers. During aging melanin production decreases

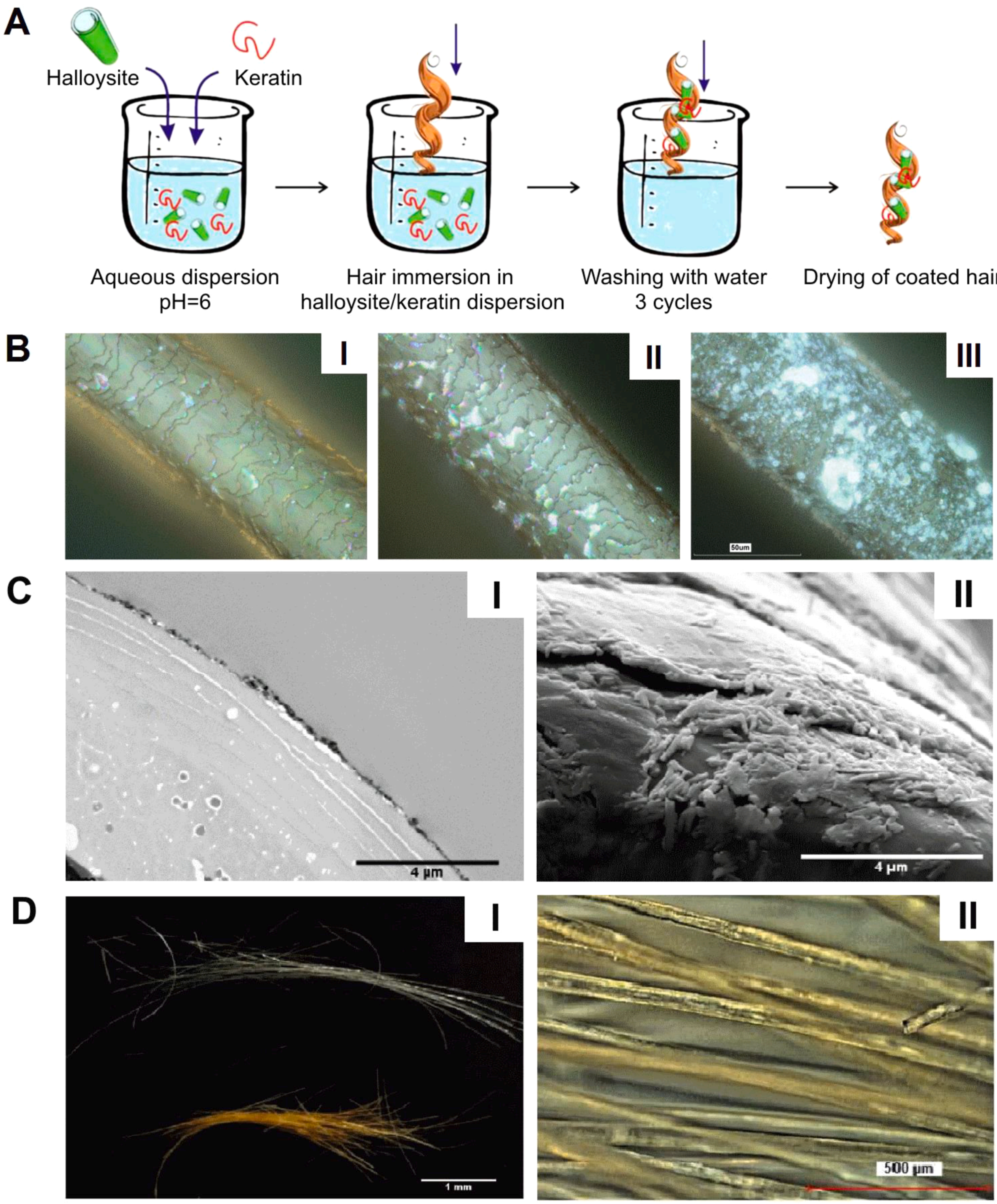

Fig. 2. A - Schematic representation of the hair treatment protocol based on halloysite/keratin aqueous dispersions; B - 3D measurement laser scanning microscopy of untreated hair (I), hair treated by keratin solution for 60 min (II), hair treated by the halloysite/keratin mixture for 60 min (III). (Reprinted from Ref. [48]. The images are reproduced under Creative Commons Attribution 4.0 International License.); C - TEM image of cross-section of coated with darker halloysite visible at the edge (I) and SEM image of the same area (II). D - Color treatment of hair with lawsone-halloysite complex: Grey hair (upper image, I) and treated with an aqueous dispersion of lawsone-loaded halloysite gives bright ginger color (bottom image, I). High definition images of hair coated with lawsone-loaded halloysite (II) (Reproduced from Ref. [60] with permission from the Royal Society of Chemistry).

or stops, gray hair appears causing concern in people. The following chemicals are commonly used in hair dyes: ammonia, peroxide, p-phenylenediamine (PPD), diaminobenzene, toluene-2,5-diamine, resorcinol, etc. [66]. Permanent dyes are the most effective in hair coloring but often have negative side effects are skin rashes, itching, hair loss, dandruff, irritation, cancer [67], asthma, allergic response [68,69], eye sight weak, etc. [66]. Nanoarchitectonic approaches can reduce the unwanted properties of hair dyes. p-Phenylenediamine (PPD) is a derivative of aniline that is used as an ingredient in hair dyes [69] and is occasionally applied as a substitute for henna. PPD-incorporated NPs were formed by the ion-complex formation between the cationic groups of PPD and the anionic groups of poly(γ-glutamic acid) (PGA). To reinforce PPD/PGA ion complexes glycol chitosan was added. PPD-incorporated NPs can reduce the side cytotoxic and allergic effects of PPD dye [70]. Polydopamine (PDA) allows to achieve black colors (i.e., natural Asian hair colors) in human hairs in the presence of ferrous ions and involved three kinds of deposition mechanisms (i.e., innate binding ability of polydopamine, metal-assisted self-assembly of polydopamine, and metal-related bridging between keratin surface and polydopamine). The dyed hairs demonstrate resistance to conventional detergents, as well as different kind of metal ion allow to finely adjusting the shades of hair color. In addition, dyeing methods with PDA showed much greater viability in skin toxicity tests on mice and much less hair loss [71]. The method of hair dyeing based on in situ polymerization of dopamine catalyzed by copper sulfate and hydrogen peroxide on the hair surface to form a polydopamine (PDA) coating for was developed (Fig. 3) [72]. Human hair can be dyed by PDA in as little as 5 min with significant durability of PDA-based hair dye which barely faded after continuous washing with shampoo (30 times). In addition, remarkable antibacterial properties were demonstrated which could effectively prevent the occurrence of bacterial inflammation on the scalp [72].

Encapsulation of natural macromolecular compounds within nanomaterials is of particular interest. The silk derived from silkworm *Bombyx mori*, consists mainly of sericin and fibroin proteins. Sericin is a globular protein, soluble in water and has a wide range of molecular weight ranging from 10 to over 300 kDa has been incorporated into conditioning agents as cationic nanoparticles and proven as useful component in repairing damaged cuticles, restoring gloss and texture [36]. Sericin nanoparticles in hair dye formulations help to maintain the hair colour along the washes. The dye formulations containing 3% of sericin nanoparticles and the placebo formula without addition of nanoparticles were applied to the hair locks for evaluation of the proposed active agent's effect in colouring. The dye was previously mixed with a hydrogen peroxide and applied on the hair for 30 minutes. After this time the locks were rinsed with running water and dried. Subsequent data and images analysis of damaged hair treated with sericin nanoparticles clearly showed the recovery of the hair's healthy appearance after the sericin nanoparticles application to its surface [36]. In general, lipid capsules can easily penetrate the stratum corneum without assistance or traverse the epidermis through shunt pathways inside the follicles. Melanin from cuttlefish ink was encapsulated into nanolipid bubbles for delivery inside the follicles for white hair photoepilation [73].

A synthesis of gold nanoparticles (GNPs) inside the hair cortex have been successfully used to dye white hair a deep brown colour lasting during 16 days. [44]. In this study, hair fibers treated with alkaline solution of $HAuCl_4$ gave different shades (ranging from pale yellow to deep brown) in a time-dependent manner, with colour remaining even after repeated washing. GNPs also caused an intense red fluorescence of dyed locks exposed to the blue light. [44]. The composition and method for emitting of visible light using quantum dot luminescent nanoparticles was developed for artificially coloured or pigmented hair and dark skin [74]. The method described in the patent provide a broad palette of different fashionable color shades from the red range to the brown and black range according to the type and composition of its ingredients. Two-dimensional and three-dimensional modifications of carbon are used in hair cosmetology. Water-based graphene suspensions find application as hair dye. Graphene oxide hair formulations mixed with chitosan can provide brown to black colors. The graphene hair coating has demonstrated the strong resistance to repeated shampooing and thus obtain the characteristics of a permanent hair dyes [75]. However, many researches reveal that graphene oxide leads to lung damage [66]. Carbon nanotubes have been modified to increase their affinity for hair. This nanotubes-based composition provides a wash-resistant colour without destruction of the keratin fibers [49]. Therefore, nanotechnology can provide new possibilities for hair dyeing, in particular enhancing long-lasting effect of dye while decreasing the toxicity of pigment components.

2.1.2. Nanomaterials for biomedical applications

Treatment of Pediculosis and Scabies. Pediculosis represents the lice infestation by blood-feeding ectoparasitic insects of the order *Phthiraptera*. Millions of children each year get head lice, this state known as pediculosis. Head lice have evolved resistance to many of the currently used pediculicides moreover insecticides and acaricides are dangerous toxic compounds and always be used with precautions, since they may harm effect to health of humans and animals that induce demand an effective new treatment. [76]

Momordica charantia Linn. (*Cucurbitaceae*) is liana grows in tropical

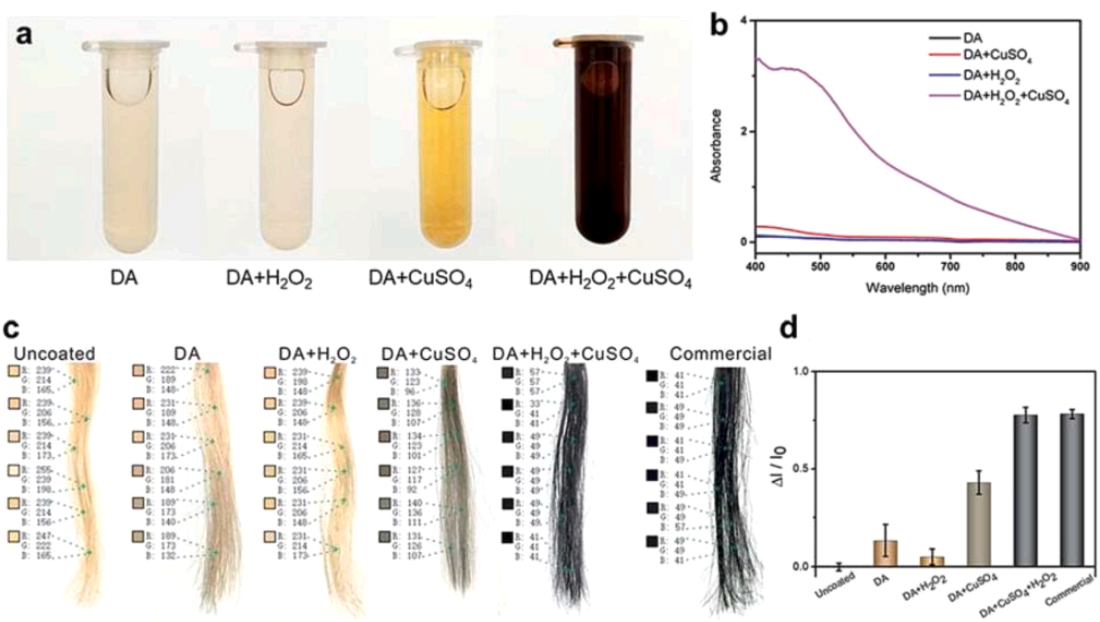

Fig. 3. (a) The colours and (b) UV/Vis absorbance spectra of various dopamine solutions. (c) The digital picture and (d) variations in the RGB intensity of uncoated hair, dopamine dyed, dopamine + H_2O_2 dyed, dopamine + $CuSO_4$ dyed, dopamine + H_2O_2 + $CuSO_4$ dyed, and commercial product-dyed hair. (Reproduced from Ref. [72] with permission from the Royal Society of Chemistry).

areas, where it is used for food and as the medicinal plant. *M. charantia* has been reported to have antibacterial, antihelmintic and anti-mycobacterial properties. An aqueous extract of the leaves of this plant was combined with ZnO NPs for evaluation of the anti-parasitic activity of synthesized ZnO NPs. The SEM image demonstrated that the synthesized nanoparticles were spherical in shape with a size of 21.32 nm. The results of GC-MS analysis indicated the presence of the major compound of Nonacosane (a pheromone of several insects) in the *M. charantia* leaf extract. Cattle tick, head lice and mosquito larvae were exposed to varying concentrations of the ZnO NPs and leaf extract for 24 h. Received data revealed that synthesized ZnO NPs in combination with *M. charantia* extract possess excellent anti-parasitic activity against the adult of *Pediculus humanus* capitis, and the larvae of *Anopheles stephensi, Culex quinquefasciatus* and *Rhipicephalus (Boophilus) microplus* [76].

Plant extract from another species, *Lawsonia inermis,* used in the synthesis of silver nanoparticles (Ag NPs) was tested against human head lice (*Pediculus humanus capitis De Geer* (*Phthiraptera: Pediculidae*)), and sheep body lice (*Bovicola ovis Schrank* (*Phthiraptera: Trichodectidae*)). To determine the potential of pediculocidal activity the contact method was conducted and impregnated method was used with slight modifications to improve practicality and efficiency of tested materials of synthesized Ag NPs against *B. ovis*. Eventually were revealed that synthesized Ag NPs are highly stable and had significant adulticidal activity of *P. humanus capitis* and *B. ovis*. [45].

It has previously been noticed that most of the neurotoxic anti-lice treatments are not reliably ovicidal and over time become useless due to drug resistance [77]. The development of resistance is excluded if the treatments based on a mechanical mode of action the weakness of which is necessity of tedious combing. An oily single-phase lotion composed by deterpenated Protium heptaphyllum resin as an active ingredient concentrated in grapeseed oil as lipophilic support was designed with the other ingredients such as latent surfactants, Laureth-4 and Comamide MIPA (Eur-Amid N2®), and Citrus aurantium dulcis peel wax a natural antioxidant with antimicrobial properties consists of a complex of special esters. Authors explained the efficiency of this new lotion by mechanical action in the plant's resin from Protium heptaphyllum, easily distributed along the hair and on the parasites with help of Eur-Amid N2®, by decreasing the surface tension. As a result, the chitin of parasite's exoskeleton becomes accessible to the resin. A single application of the new lotion made of semi-crystalline polymers and plant extracts was effective in elimination of head lice infestation in 15 min and completely inhibit eggs hatching [41].

Dandruff treatment. Dandruff and seborrheic dermatitis appear in more than 50% of adults. It should be noted that the regular use of shampoos as hair cleansing agents and conditioners as surface lubricating products, give only temporary reducing of flakes and sebum on the scalp [78].

Shampoos and conditioners, enriched with zinc and chitin nanofibril complexes were formulated. According to results obtained *in vitro* and *in vivo* chitin nanofibrils, (CN) complexing with zinc pyrithione nanoparticles, possess activity in reducing hair flakes and sebum, ameliorating the hair shine and manageability when compared to conventional formulations. CN have repairing activity on both the proteins of cortex and surface scales. Anti-dandruff and anti-oily activity of zinc-CN is explained an important role of zinc in metabolism of free fatty acids [23]. Zinc pyrithione improves both effects and may have antimicrobial activity as demonstrated previously [79].

Antibacterial coatings of hair. Nanoparticles with antibacterial properties have several mechanisms of action. For example, they can completely change the metabolism of bacterial cells, altering the permeability of their membrane, which leads to an imbalance of electrolytes, inactivation of various proteins, including enzymes, and disrupting gene expression [80–82]. The main mechanisms determining antibacterial properties are oxidative stress, metal ion release, and non-oxidative mechanisms [83].

Quite a lot of research has been done on the creation of antibacterial coatings on natural fibres. Typically, hair can also be modified to provide antibacterial properties.

Hair can be coated with compounds containing metal nanoparticles and semiconducting materials which have been reported as antibacterial materials. Antibacterial properties of such coatings have previously been demonstrated in work devoted to coating natural fibres with ZnO-TiO2 [84]. Gold nanoparticles has strong antifungal and antibacterial properties which make this material perspective for the cosmeceutical industries [85]. Silver nanoparticles have similar properties, although in this case there are limitations due to their toxic properties [86].

Another approach to creating multifunctional hair coatings with antibacterial properties is treatment with non-metallic nanocoatings. Polydopamine (PDA) nanoparticles was found to possess antibacterial properties with simultaneous hair colouring (Fig. 4A) [87]. It was revealed that PDA-colouring of hair can prevent the scalp from different bacterial infections. PDA exhibits redox-dependent properties that allow it to act as a pro-oxidant and antioxidant. The prooxidant properties of the molecule leading to the formation of reactive oxygen species allow the antibacterial properties of polydopamine nanoparticles to be manifested [88,89].

Chitosan, the chitin derivative, which has recently been extensively used in biomedical and cosmetic fields. Chitosan possess safety and absence of toxicity in in vitro and in vivo as well as biodegradability and low immunogenicity [90]. Its effectiveness in hair care was demonstrated in the study of hair growth promoting in the treatment of hair loss in patients with alopecia [91].

The antibacterial properties of chitosan are based on interaction between phosphoryl groups on chitosan and lipopolysaccharide on bacterial cell membrane (Fig. 4B) [92,93].

2.2. Nanoformulations for hair de novo regeneration

2.2.1. Delivery to hair follicle

Non-cosmetic issues for example hair loss cannot be solved by treatment on the hair shaft with drugs. Therefore, hair follicles regulating hair growth represent a target site of particular therapeutic interest [56]. The hair follicle is one of the pathway for the drugs transportation through the skin [94], in this way the intensity of substances penetration depends on the density of the follicles in a certain area of the skin. For topically applied substances, the hair follicle represents a distribution point.

Lipid nanosystems for HF drug delivery and the alopecia treatment. Targeted delivery to the hair follicles depends on the lipophilicity of the drugs. In this way, lipid carriers have advantage due to their chemical nature that may facilitate such interaction, reason why they have been extensively studied for HF targeting. [94]. The delivery systems presented lipid-based nanoparticles such as nanostructured lipid carriers (NLC), solid lipid nanoparticles (SLN) [42], liposomes [43], transferosomes [95], niosomes [96] and ethosomes [97] have been developed to intense the skin permeation of various active molecules [94]. Particle size influence on drug delivery in such a way that the smaller nanoparticles demonstrate greater depths of penetration, for example, the size range below 100 nm by promoting accumulation in the hair follicles allowing the delivery of drugs at the level of the capillary bulb. Particles around 200 nm have been more suitable for drug delivery to the isthmus part of the hair follicle [98]. When particle size decreases the surface area and hence, the dissolution rate, affecting the drug pharmacokinetic profile (accumulation area, release and distribution, metabolic transformation, etc.) increase. Solid lipid nanoparticles encapsulating minoxidil, with diameter of 190 nm, demonstrated a better accumulation in skin layers in relation to commercial products

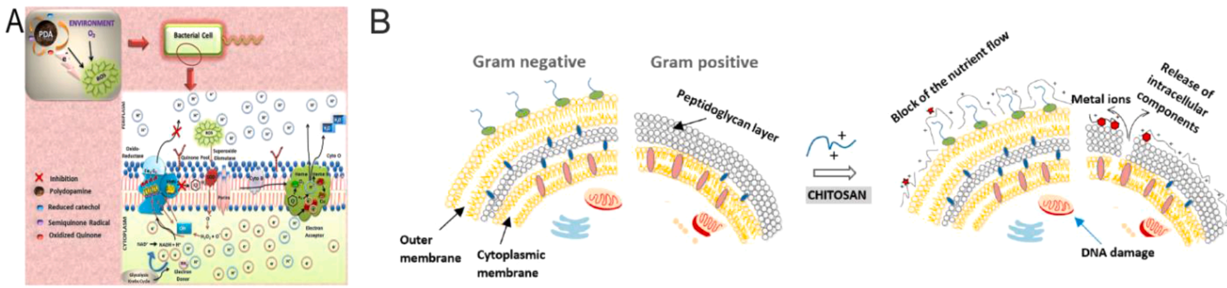

Fig. 4. Antibacterial properties of non-metallic nanoparticles for hair treatment. A - reactions occurred during electron transfer from polydopamine; B - Mechanisms of antibacterial action of chitosan. (Reprinted (A) from Ref. [87], (B) from Ref. [93]. The images are reproduced under Creative Commons Attribution 4.0 International License).

[99].

Also, for the distribution of drugs, the chemical structure of nanolipid carriers is of importance. NLC were developed to over-come some potential limitations associated with SLN as low loading capacity for a number of active compounds and stability of the loaded particles due to the relatively high water content. SLN may form crystalline networks [100], leading to drug leakage during storage [101]. Wang et al. (2017) developed and characterized minoxidil-loaded NLC (MXD-NLCs) and SLN (SLN -NLCs) for topical treatment of alopecia. The authors noticed that MXD-NLCs had a more pronounced permeation and retention profile than MXD-SLNs, moreover the release of minoxidil from NLC was faster than from SLN, and skin irritation tests showed that no erythema was developed after MXD-NLCs [42]. In another study, NLC were loaded with clobetasol propionate for targeting drug delivery to the epidermal layer [102]. The finasteride-loaded NLC presented good physical and chemical stability in storage [103] which can be attributed to the presence of oleic acid in the composition. Oleic acid promotes an amorphous form in the solid lipid matrix and, as a result, decreases the crystallinity of the particles, thereby improving the encapsulation efficiency [104].

The advantage of liposome-based delivery systems is the increased permeability of skin and tissues for these carriers. Liposomes form a phospholipid film on the skin surface that interacts with the sebum facilitating follicular drug distribution. The finasteride-loaded vesicular systems incorporated into 2% w/w methyl cellulose gel demonstrated significantly higher permeation of FNS through excised abdominal mice skin compared corresponding solution and conventional gels. Liposomal-FNS formulations were quite stable for 2 months in the refrigerated conditions [43]. Transfersomes are ultra-deformable or elastic carriers possessing a higher hydrophilicity than liposomes, with tend to be more resistant to fusion with the skin lipids. The elasticity has been achieved by incorporation of a surfactant as edge activator into the lipid bilayer structure (Fig. 5A) [105]. Remarkable feature of transfersomes is ability to carry large molecules across intact mammalian skin if applied non-occlusively. Transfersomes have been used as drug carriers for peptides, vaccines [106], anticancer drugs [105] and a range of small therapeutic molecules [107], both in vitro and in vivo. In vivo tests in mice with a formulation on the base of minoxidil and caffeine encapsulated in transfersomes prolonged permanence of the cells in the anagen phase, promoted the hair length and weight [107].

The novel drug delivery systems, called niosomes, are formed as bilayer of non-ionic surface-active agents [108]. Niosomes represent small unilamellar vesicles (SUV) (10–100 nm), large unilamellar vesicles (LUV) (100–3000 nm), and multilamellar vesicles (MLV) with more than one bilayer (Fig. 5B) [109]. The vesicle holds hydrophilic drugs within the space inside the vesicle, while hydrophobic drugs are embedded within the bilayer itself, therefore they can be used for a wide range of substances [109].

Finasteride niosomes have been formulated for effective treatment of androgenetic alopecia and the *in vitro* permeation and *in vivo* deposition studied using hamster flank and ear models. The results demonstrated the perspectives of niosomes for successful delivery of finasteride in topical application [110].

Lipid nanoemulsion, squarticles, was investigated as nanotechnology-based formulation for topical administration of minoxidil (medication for hair growth enhancement); this nanocariers was fabricated from sebum-derived lipids such as squalene and fatty esters. Cationic squarticles have larger average diameters than anionic ones. Lipidic nature and the nano-size facilitate the interaction and fusion between squarticles and the sebum favoring targeted delivery of minoxidil into follicular [111]. In comparison study two different nanosystems, nanostructured lipid carriers (NLC) and nanoemulsions (NE) with Nile red was demonstrated the advantages of NE. Nile red-containing NE contributed to the relatively homogeneous distribution of red fluorescence over the treated skin. Squarticles in the form of NE were detected in both the stratum corneum and follicular ducts. Higher intensity of Nile red fluorescence was detected in the case of NE-treated skin [111].

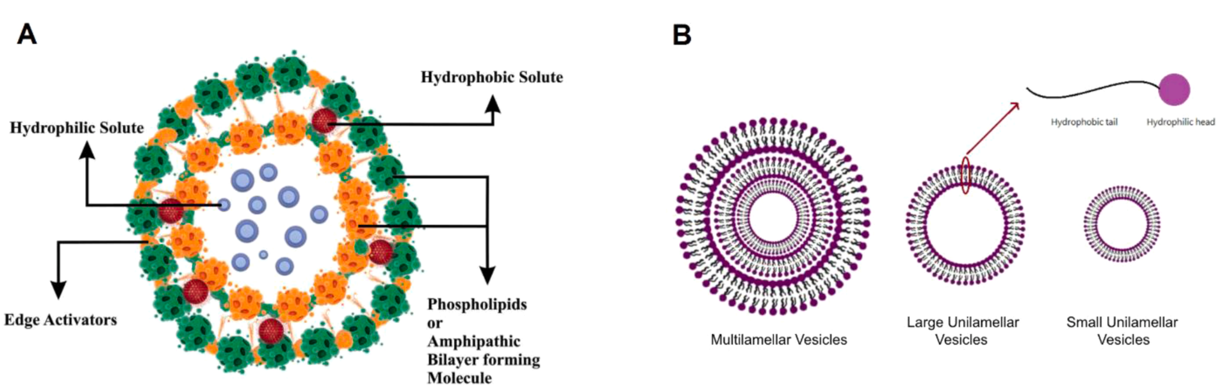

Fig. 5. A - Structure of transfersomes; B - Typical vesicle size of niosomes. (Reprinted (A) from Ref. [105], (B) from Ref. [109]. The images are reproduced under Creative Commons Attribution 4.0 International License).

The methods of regenerative hair medicine involve the transplantation of autologous hair follicular stem cells (HFSCs) and dermal papilla cells (DPCs), as well as using of signal molecules, growth factors and topical drugs into regions of hair loss [37,112].

Polymeric nanoparticles for the treatment of alopecia. Biodegradable and biocompatible polymeric nanoparticles for the treatment of alopecia employed gelatin, alginate [37], chitosan [38,39], albumin, acquired from natural source, and synthetically produced polyvinyl alcohol [113], poly(ε-caprolactone) [47], poly(lactide-co-glycolide) copolymers, poly(amino acids) and poly(lactide), polymethacrylates [114]. Polymeric nanoparticles have the ability to protect encapsulated drugs from degradation for several months [115], increased regular distribution and controlling drug release [39,116]. The mechanism of drug release from these polymeric nanoparticles occurs through the relaxation of the polymer chains or by the polymer degradation (for example, as a result of fermentative hydrolysis in biological systems) leading to the release of substances from carriers. Moreover, the aqueous phase interacts with the particles during the drug release that also leading to the relaxation of the polymeric wall [117]. The pharmacodynamics and penetration depth of polymer nanoparticles into tissues, as in the case of lipid nanoparticles, depend on their size and chemical structure. Polymer nanoparticles (NP) with smaller sizes (less than 50 nm) may reach a deeper penetration into the hair follicles (HF), while NP with a size of about 5 μm accumulate in the infundibulum of the HF [118].

Drugs delivery of poorly water-soluble compounds can be realize using halloysite nanotubes [62], lipid nanocarriers, or nanocrystals [119]. Due to the increased kinetic solubility of nanocrystals, which facilitates passive penetration through the skin and some another special properties, nanocrystals represent a very promising drug delivery system not only for oral and topical application but also for hair follicle targeting. The curcumin nanosuspension (nanocrystals with a size of about 300 nm) was incorporated into the different gel bases (polar, non-polar hydrogels and oleogels), fabricating the gels that contained 1 % (w/w) curcumin. The *ex vivo* pig ear model was used for the analysis of the efficacy of hair follicles penetratration, as well as the passive, dermal penetration. As a result, hair follicles were able to efficiently absorb nanocrystals that reached the lower part of the infundibulum. Humectant-contained hydrogels exhibited the highest level of passive penetration, however, these hydrogels penetrated the hair follicles with less efficiency. [119].

Nanoscale biomimetic ECM. Another aspect of regenerative medicine is the reconstruction of matrices and scaffolds which provide favourable microenvironment for directing and maintaining cell fate for tissue regeneration [63,120] because long-term conventional 2D culture could also impair the HF inductive capacity of cells [113]. In addition, hair follicle reconstruction requires transplant a rich pool of hair inducing stem cells [121], represented by hair follicle stem cells (HFSCs) as well as dermal papilla cells (DPCs). HFSCs were coated with gelatine and alginate using layer-by-layer (LbL) self-assembly technology acting as nanoscale microenvironment to regulate hair follicle stem cell fate. Then transforming growth factor (TGF)-β2 was loaded into coating matrix. (TGF)-β served as a sustained-release signal molecule in reconstructed biomimetic ECM that allowed for the stable proliferation of HFSCs and maintenance of their stem properties both *in vitro* and *in vivo*. Thus, LbL gelatine-alginate coating saturated with TGF-β2 mimicked the microenvironment that regulates stem cell fate for tissue regeneration during HF cycling. Co-transplantation of 50% LbL-HFSCs and 50% LbL-(TGF-β2)-HFSCs with neonatal mouse dermal cells into skin of nude mice has led to improvement of HF regeneration [37].

The combination of epidermal keratinocytes, dermal cells and β-catenin-expressing CD133+DPCs with micro/nanoscale hydrogels could contribute to HF regeneration). The nanogel encapsulates a single cell by LbL self-assembly and further form dermal papilla cells spheroids by physical crosslinking on nanogel-coated cells [122].

The DP cells from postnatal skin retain the ability to direct epithelial cells to form hair follicles. However, after a few passages cultured *in vitro* in ordinary (2D) conditions DP cells lose their trichogenic properties [123]. Experimental results indicated that hydrogel from human placenta can provide a favourable biomimetic 3D microenvironment to regulate the morphology and functions of DPCs by reactivating key genes that are not expressed under 2D conditions [124].

Nanofiber scaffolds for HF regeneration. Chitosan/polyvinyl alcohol nanofiber scaffold was obtained for the fabrication of efficient and functional multicellular spheroids without tends to lose the function with the increasing of cell passage. The cellular nanofiber structure is mimic the growth microenvironment and extracellular matrix microenvironment of DPCs *in vivo*, which facilitates aggregation of DPCs enhances secretory activity of DP microtissues and restores their intrinsic properties. Further transplantation of cultured DP microtissues and epidermal cells into BALB/c nude mice demonstrated that this composite structure has enhanced HF-inducing ability [113].

The hair follicle regeneration was observed in a diabetic wound healing model by the test of regenerative potential of multidomain peptide (MDP) hydrogels. These hydrogels elicit a mild inflammatory response with promotes angiogenesis that can also support newly formed HF in vivo [125].

Multifunctional scaffolds have serious advantages allow to achieve at once many challenges of regenerative process including inhibition of bacterial infections during wound healing. Zn-doped hollow mesoporous silica/polycaprolactone electrospun membranes is a promising multi-functional material with antibacterial activity for the treatment of infected wounds and accelerating the skin wound healing, angiogenesis promoting and hair follicles regeneration [47].

In general, hydrogels and matrix porous scaffolds could be suitable materials to support stem cells for organogenesis and regeneration of HFs, with main function to provide a specific microenvironment and mimic a stem cell niche in which HF stem cells are present in an undifferentiated and self-renewable state.

As mentioned above, nanoparticles smaller than 50 nm can passively penetrate the skin through the SC lipid matrix, reaching the deepest layers of the SC, the stratum granulosum, and hair follicles [126]. Therefore, metallic nanoparticles which sizes are smaller than 10 nm have great pharmaceutical interest favourable potential carriers of drugs to stimulate hair growth. Silver and iron nanoparticles were synthesized in alcoholic *Blumea eriantha* DC extract (named BEAgNPs and BEFeNPs, respectively) and have been estimated as a hair growth promoter. The qualitative and quantitative analysis of hair growth dynamic, histopathological studies, skin thickness, and length of the hair follicle after treated naked skin of mice with BEAgNPs/BEFeNPs was performed. The best results were obtained with silver nanoparticles, biological potential of which was comparable to Minoxidil [127].

3. Summary and short perspectives

In conclusion, it should be noted that nanocosmeceuticals are a very new and insufficiently studied aspect of nanomedicine. The possible toxicity of nanomaterials, which in some cases does not depend on the chemical nature of nanocomponents, should be studied carefully before using such materials in products for widespread and mass use in society. In the field of hair and scalp alteration products and treatments, nanoarchitectonics may have the potential to evolve towards imparting new properties to hair that can be an effective protective coating or a way to restore lost hair. From this point of view, nanoarchitectonics can be combined with tissue engineering to create unique hair restoration methods.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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