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LYCOPENE – BACKGROUND, PERSPECTIVES AND CHALLENGES IN DERMATO-COSMETIC FORMULAS

BY

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Abstract. In the dermato-cosmetic formulation, the antioxidants have gained an almost permanent place based on strong scientific arguments. The most of the antioxidant ingredients enriched formulas are intended for antiaging skincare products, even though in a wide variety of presentation forms, these have no special indications and are limited to daily skincare routine for healthy skin. Nevertheless, there is an increasing interest for antioxidant compounds also for dermatocosmetic products with medical indications for acne, rosacea, and seborrheic dermatitis. Therefore, finding new resources or fructification of any discovered source of antioxidant compounds have led to increasing the researches related to performance, stability, efficiency and quality of extraction methods. Special attention to indigenous plant resources, economical and rich in antioxidant active ingredients is as well inherent but also incipient, if we have to consider the amount of well-known but still unexploited active ingredients.

Keywords: antioxidant effects; dermatocosmetic products; lycopene characterization; lycopene extraction methods.

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1. Introduction

There is a series of long-established antioxidants, such as vitamin C, E, resveratrol, Q10 coenzyme used to combat and / or treat various dermatological pathologies. Combating oxidative stress is a goal taken into consideration by many specialists, and antioxidants play an extremely important role (Kumar *et al*., 2020; Turcov *et al.*, 2020a, 2020b). New ones are gaining ground in dermatocosmetic formulas, various polyphenols or even synthetic antioxidants, such as idebenone (Fig. 1). Based on the number of antioxidants used in food supplements, there is a need for a further in-depth analyze of antioxidant potential of other similar compounds.

Lycopene is one of these potential compounds. Although it has a significant history in literature and in food supplements manufacturing, for reasons incompletely understood yet, its presence in dermatocosmetic formulas is bellow expectation suggested by its proven properties (Fernández-García *et al*., 2012). Currently, lycopene is first-line as a coloring ingredient in food products and supplements and, secondary, as an antioxidant.

The aim of this paper is to synthesize literature data on the use of lycopene in dermatocosmetic, to evaluate extraction methods from indigenous sources and to identify barriers to the inclusion of lycopene as a top antioxidant for dermatocosmetic formulations.

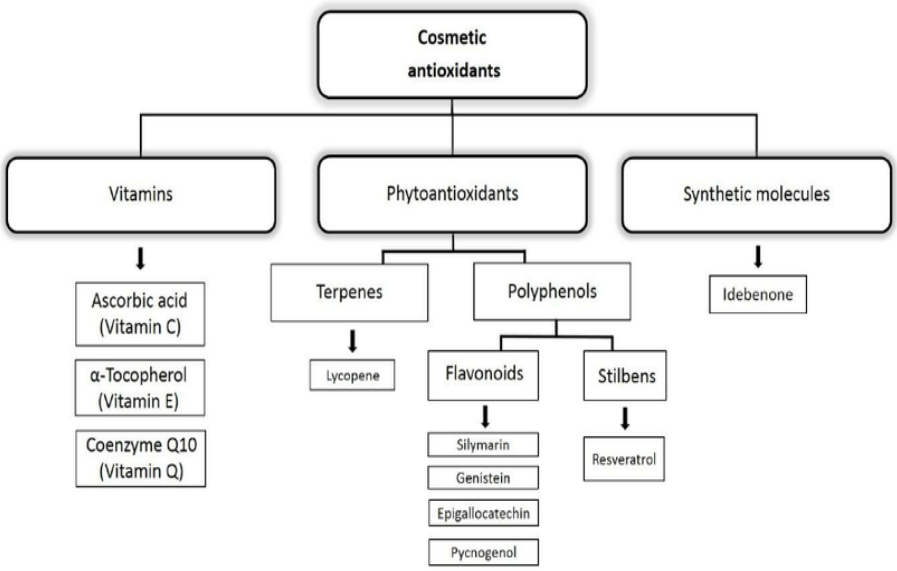

Fig. 1 – The most frequently antioxidants used in the dermatocosmetic products (according to Montenegro, 2014).

2. Lycopene - General Characteristics

Lycopene is a fat-soluble pigment from carotenoid group, responsible for red color of tomatoes and many fruits. It is an acyclic non-polar compound with $C_{40}H_{56}$ formula. His structure has 11 conjugated and 2 unconjugated double bonds. Almost 95% of lycopene found in nature is in trans-isomer form, considered more stable than cis-isomer lycopene (Abreu *et al*., 2011). In the same time, for the lycopene intake, the bioavailability is considered to be higher in cis-form than in trans-form, cis-isomers presenting a higher solubility in bile acid micelles and being easier to incorporate by chylomicrons (Urbonaviciene *et al*., 2018). Lycopene is a lipophilic antioxidant, lipid-soluble, the predominant carotenoid found in human plasma.

Some sources rich in lycopene that can be used for its extraction is presented in Table 1.

Table 1
Food Sources of Lycopen

Source	Type	Amount (mg/100 g wet weight)
Apricots	fresh	0.005
Apricots	dried	0.86
Chilli	processed	1.08 – 2.62
Grapefruit	Pink, fresh	3.36
Guava (*Psidium guajava*)	Pink, fresh	5.40
Tomatoes	fresh	3.1 – 7.74
Tomatoes (*Lycopersicon esculentum*)	Wholed, peeld, processed	11.21
Tomatoes juice	processed	7.83
Tomatoes paste	canned	30.07
Ketchup	processed	16.60
Pizza sauce	From pizza (not canned)	32.89
Salsa	processed	9.28
Spaghetti sauce	processed	17.50
Wattermelon (*Citrullus lanatus*)	Red, fresh	4.10

3. Extraction Methods of Lycopene

The highest known concentration of lycopene in lycopene-riches formulation is 30,000 - 60,000 ppm, difficult however to be safely dosed for human intake (Sabio Rey, 2005). As far as dermatocosmetic products are concerned, there are no studies developed to establish the most efficient and safe concentration for external use.

In traditional methods of extraction, organic solvents are used, as lycopene is apolar, but these solvents present toxicity in different levels, thus other methods, like using supercritical fluids shall be considered (Sabio Rey, 2005; Choksi and Joshi, 2007).

Modern, advanced protocols of extraction for lycopene result in higher concentration of lycopene extracted, than conventional simple solvent extraction. The most considerable amounts of lycopene are present in the outer part of pericarp (skin and seeds), while the jelly part contains mainly beta-carotene (Hussain *et al*., 2017).

A remarkable patent which presented a great industrial application proposed a method using polar solvents (water, ethanol, polyoles) for extraction of other compounds than lycopen, crystals, present in chromoplasts, the location of lycopene in plants.

The vegetal material used for extraction are tomatoes and tomato-by-products, among which tomatoes paste (up to 38.8%, (Hussain *et al.*, 2017)) is by far the most rich in lycopene.

Still, the maximum radical scavenging activity was determined in raw tomatoes (Hussain *et al*., 2017).

Therefore, there are other different methods of extraction aimed to optimise the amount of lycopene obtained in safe, risk-free conditions.

Table 2

Extraction Methods of Lycopen from Vegetal Material

Source	Type	Extraction and Analytical characterization methods	Amount	References
Tomato by-products (*Lycopersicon esculentum Mill.*)	Peel, seeds, pulp	HPLC-grade solvents (hexane, methanol, methyl-t-butyl ether, tetrahydrofuran, isopropanol) Supercritical fluid extraction with CO_2	Five times higher lycopene content in peel > seeds, pulp	(Urbonaviciene *et al.*, 2018)
Tomato products (paste and pulp)	Fresh and cooked for several hours	Hydrolytic Enzyme-aided extraction (enzyme pectinase)	Increased by 90.6 μg/g (188%)	(Hussain *et al*., 2017)
Tomato peel (tomato processing waste)	Blended and partially dehydrated	Enzymatic pre-treatment and surfactant assisted extraction (3 extraction cycles)	643±17 μg/g	(Papaioannou and Karabelas, 2012)
Tomato concentrate	Processed	Lipid extraction by solid-liquid extraction	500-1000 ppm, free of organic solvent	(Sabio Rey, 2005)

Tomato juice	Fresh	Layered double hydroxides (LDHs) (green extraction-inorganic compound-metal cations)	No quantitative analyse performed	(Carbajal Arizaga *et al.*, 2018)
Tomato skin and seeds	Tomato-processing wastes	Hydrophilic and lipophilic solvents (distilled water, ethanol, isopropanol, ethyl lactate, ethyl acetate, n-hexane) + Microwave and Ultrasound Assisted extraction	Lycopene content higher in lipophilic solvents and lower in hydrophilic solvents	(El-Malah *et al*., 2015)
Cherry tomato/ watermelon	Fresh	Benzene/Methanol/Aceto e-Ethanol-Hexane/Acetone-Petroleum ether/Hexane	88.87 mg/kg / 74.53 mg/kg /	(Lilwani and Nair, 2015)
Red tomatoes (*Lycopersicon esculentum Mill*)	Fresh	Ultrasound assisted extraction (UAE) with the aid of RSM (Response Surface Methodology)	All-trans lycopene 5.11±0.27 mg/g dry weight, enhanced by 75.93% with RSM compared to other optimised conventional technology	(Eh *et al*., 2012)
Tomato skin, seeds and part of the pulp	Fresh	Ultrasound assisted extraction (UAE) under moderate pressure	Extraction Yield increased with 143% compared with control	(Luengo *et al.*, 2014)
Tomato skin	Fresh	Optimised separation through dynamic adsorption/desorption	30.4 fold increased lycopene content in lycopene oleoresin	(Liu *et al*., 2010)

4. Mechanism of Lycopene Action

Lycopene has an acyclic molecular structure, with multiple conjugated double bonds which, additional to its high hydrophobic property manifests an antioxidant effect expressing in quenching singlet oxygen and scavenging free radicals. Moreover, after deactivating free radicals through electron transferring, the new lycopene radical formed is stable due to the same large

number of double carbon bonds, and can be further stabilized by resonance (Sabio Rey, 2005).

On the other hand, its unsaturated structure makes lycopene instable to certain reaction like oxidation or to heat and light. Therefore, its important antioxidant activity is strongly diminished in these specific conditions.

Lycopene's antioxidant action is exerted through:

- Quenching singlet oxygen (the most dangerous RO species generated in the skin as a consequence of UV exposure (Caramori Cefali *et al*., 2015). Lycopene capacity of quenching singlet oxygen is twice that of β-carotene and ten times higher than that of α-tocopherol.
- Scavenge free radicals
- Prevent formation of free radicals
- Electron transfer
- Hydrogen atom transfer (Fig. 2)

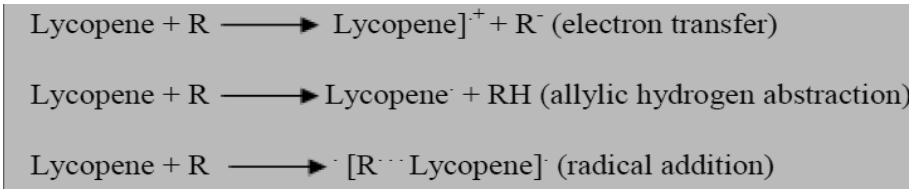

Fig. 2 – Lycopene's antioxidant mechanism (Kaur and Kaur, 2015).

Studies show some important effects of lycopene involvement:

- Modulating growth factors (insulin like growth factor 1 (IGF-1), vascular endothelia growth factor (VEGF), epidermal growth factor (EGF), platelet derived growth factor (PDGF) (with key anti-tumor role))
- Signaling pathways (androgen pathway, cytokine pathway (IL-4, IL-6), growth factor)

There are also presented non-oxidative mechanism of lycopene (Sgherri *et al*., 2015):

- Gap junction communication
- Cell cycle regulation
- Modulation of gene expression
- Improvement of hormone and immune response

5. Biological Effects of Lycopene

Lycopene has no pro-vitamin A activity, owing to the absence of the β-ionone ring at the end of its structure, but it is considered to have the highest scavenging capacity of singlet oxygen, among the main carotenoids found in human plasma (like α-carotene, β-carotene, β-cryptoxanthin, lutein, zeaxanthin),

reducing the rate of free radicals. This activity stands as an evidence of the strong antioxidant activity, which is associated with the contribution in preventing or treatment in different pathologies.

Existing studies show different types of involvement in various oxidative stress-related diseases, as shown in the Table 3.

Table 3
Biological Effects of Lycopene

Type of participation	Pathology	Suggested biochemical mechanism of action	References
Reduce occurrence	Esophageal, gastric, prostate, lung cancer	Prevent or minimise the effect of free radicals	(Kaur and Kaur, 2015)
Assist in the treatment	Pancreatic, colon, rectum, breast, endometrial, lung cancer, leukaemia	Modulating the epigenome, Gene function regulation (reversing abnormal gene activation regulation or silencing, Antimetastatic activity, Apoptosis, Cell-cycle arrest, Cell-Cell communication and progression, Carcinogen metabolism	(Sgherri *et al*., 2015)
Reduce risk	Myocardial infarction	Barrier against LDL (low density lipoprotein) oxidation	(Kaur and Kaur, 2015)
Male infertility	Reproductive system	Reduce lipid peroxidation, Improve sperm quality (concentration, motility, morphology), Protects the viability, osmotic resistance and DNA integrity during cryopreservation	
Prevents insulin resistance and delay complications	Diabetes	ACE (Angitensin Cinverting Enzyme) inhibitor in vitro studies, inhibits MDA-LDL formation, prevents ox-LDL uptake by macrophage	
Bone health	Osteoporosis, bone tumour, joint inflammatory disease	Decreases serum Ca, P, alkaline phosphatise (ALP) and IL-6 concentration, Enhances serum estrogen level, bone mineral density, bone mineral content.	
Prevention	Neurodegenerative diseases	Prevents neuro-inflammation and cognitive impairment, Enhances activity of superoxide dismutase and glutathione	(Butnariu and Giuchici, 2011)
Prevention	Prevents Atherosclerosis and Blindness	Counteract lipid oxidation	(Stoica *et al*., 2018; Dasgupta and Klein, 2014)

Anti-UV damage Protection, support healing	Skin disorders (infections, seborrheic dermatitis)	Antimicrobial, anti-inflammatory action, Enhance regeneration,	(Dasgupta and Klein, 2014; Petyaev *et al*., 2019)

Despite such observations, molecular mechanism in some of the pathologies above are not fully understood. Moreover, significant lower risk of tumors, diabetes or bone diseases is shown to be in associating lycopene with vitamin E, selenium, chlorophyll or other compounds, rather than administration of one antioxidant alone (Kaur and Kaur, 2015; Sgherri, *et al*., 2015).

6. Lycopene in Dermato-Cosmetics

The presence of lycopene in dermato-cosmetic formulation is still an aspiration, while in cosmetic and beauty products is poor, both under existing products and research topic of interest as well. If we consider the large number of lycopene-based food supplements, we can easily assume that the beneficial biological effects of lycopene, proven or suggested, are convincing enough and certainly compelling for pharma industry. However, the idea of including it in cosmetic or dermato-cosmetic formulas is rarely but continuingly explored and some experimental formulas are analyzed (Table 4).

Table 4
Experimental Studies Evaluating Lycopene for Topical Use

Assay type	Objective	Lycopene concentration	Result	References
Emulsion new formula	Combating skin aging acceleration	0.58 mg/100 g sample of phyto-cosmetic	Promising antiaging product (Stable, safe, biocompatible product, cumulative effect on lycopene concentration in the skin), Sensitive in indirect/direct light and high temperature	(Caramori Cefali *et al*., 2015)
Correlation between	Establish lycopene	0.0055-0.021 detection	Lycopene is expected to be	(Darvin *et al*., 2008)

lycopene cutaneous concentration and skin roughness	involvement in maintaining skin smooth texture	through resonance Raman spectroscopic measurements, on an 0.33cm^2 skin surface structure	an efficient protective compound against negative action of free radicals in the skin	
Hyaluronidase inhibition evaluation,	Comparative measurements reported to Solanum *Lycopersicum L*. extract	Measurements for concentrations between 5.21-166.67 μg/mL, both for lycopene as for Solanum *Lycopersicum L*. extract	Higher antiaging activity than Solanum *Lycopersicum L*. extract	(Djohab *et al*., 2019)
Lycopene antioxidant like a nanoemulsion component	Evaluating the "harmlessness" of propolis-lycopene association	Good tolerability and effectiveness of product, good compliance, Reduction in collagenase activity	Obtaining and characterization a nanoemulsions based on lycopene and propolis: (20-35)% lycopene + (27-35)% propolis + (53-30)% H_2O (v / v)	(Butnariu and Giuchici, 2011)

In cosmetic products lycopene has an antioxidant role. Another experimental formulation present new concepts for encapsulating lycopene in new more stable and better absorbed products (Butnariu and Giuchici, 2011) (Table 5).

Table 5

Presentation Forms of Lycopene Based Cosmetic Products

Main ingredients	Presentation form	Indications
Calendula, lycopene, papaya enzymes	Gel mask	Hydration, antiaging, exfoliation
Aloe, Vitamin E, C, β-carotene, lutein, lycopene, zeaxanthin, CoQ10, astaxanthin	Cream	Anti (stress, environmental aging, wrinkles, UV damage) protection
Tomato extract (lycopene), sea berry oil, camellia seed oil	Oil	Antioxidant booster

Vitamin A. C, K, lycopene	Soap bar	Oily, sensitive, acne prone skin
Vitamins, plant extracts (including tomato)	Gel	Dehydrated, oily blemished-skin
Aqueous micro dispersed-lycopene	Skincare range of products (cream, gel, serum, men's moisturiser, scrub)	Hydration, renewing, protection
Lycopene applied to cosmetic beauty line	Cream, balm, essence, shampoo, soap, serum	Full body daily skin care, all skin type
Organic lycopene based products	All types of face and body products	Daily routine and treatments (anti-acne, peelings)
Others	Foam scrub, lotions, lipsticks, cleansers,	Daily care, Beauty, Face and body

7. Challenges in the Use of Lycopene in Dermatocosmetic Products

Lycopene is sensitive to heat, oxidation and light, like other carotenoids, due to its unsaturated chemical structure (Choksi and Joshi, 2007; Tang *et al*, 2015; Djohab *et al*., 2019). Experimental formulation showed that special nano liposome-encapsulated lycopene represents a protected form with higher antioxidant activity than free lycopene (Stojiljkovic *et al*., 2018).

Furthermore, oral administration has its challenges: the classical dosage issue and using the right form: although the most (95%) lycopene in tomatoes is all-trans and the most stable form, the cis-isomer is the most bioavailable (key property linked to optimum absorption, metabolism, transport and tissue distribution, bioactivity), *in vitro* and *in vivo* (the higher solubility of *cis*-isomers in bile acid micelles and the facile incorporation chylomicrons are probable explanations) (Urbonaviciene *et al*., 2018).

8. Conclusions

Lycopene is scientifically proven an effective antioxidant with numerous biological benefits. The extraction resources are notoriously rich and extremely accessible. Nevertheless, lycopene as an ingredient for topical use in dermato-cosmetic formulation is considered under-explored (Caramori Cefali *et al*., 2015).

Still, in recent years, several manufacturing companies have been developing new, attractive and promising range of products, consisting in extremely wide variety of presentation forms, textures, formulations and benefits. Among all these, no dermato-cosmetic formula, only cosmetics.

These findings support the idea of continuing researches in order to provide solutions to current dilemmas: ways to improve stability, more affordable pure lycopene, the most effective combination of formulas.

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LICOPEN – BACKGROUND, PERSPECTIVE ŞI PROVOCĂRI ÎN FORMULELE DERMATO-COSMETICE

(Rezumat)

În formulele dermato-cosmetice, antioxidanţii şi-au câştigat un loc aproape permanent, cu puternice argumente ştiinţifice. Cele mai multe formule ce conţin antioxidanţi sunt dedicate îngrijirii anti-aging, iar produsele, deşi într-o mare varietate de forme de prezentare, nu au indicaţii speciale şi se limitează la îngrijirea de rutină a pielii sănătoase.

Totuşi, există un interes în creştere pentru compuşi naturali cu acţiune antioxidantă şi pentru produse cu indicaţii medicale cum sunt acneea, rozaceea, dermatita seboreica. Astfel, găsirea unor noi resurse şi fructificarea oricărei surse de ingrediente antioxidante au dus la creşterea numărului de studii ce vizează performanţa, stabilitatea, eficienţa şi calitatea metodelor de extracţie.

Atenţia specială asupra resurselor vegetale indigene, avantajoase şi bogate în ingrediente active antioxidante este pe cât de firească pe atât de incipientă, dacă ne gândim la cantitatea disponibilă de ingrediente active cunoscute dar neexploatate.