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Comprehensive Account on Role of Catechin-based Polyphenols in Nanocomposite Formulations

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Abstract
Antioxidants play a crucial role in wellness, health maintenance and the prevention of chronic and degenerative disease, in neuroprotection activities and protection against oxidative cellular damage by UV radiation. Polyphenols are secondary metabolites of plants usually involved in defense against ultraviolet radiation or aggression by pathogens. Antioxidant properties, free radical-scavenging abilities and antimicrobial properties of these polyphenols make them useful in the field of biomedical applications; used as food additives, as bio preservatives to improve the shelf life of perishable products. Polyphenols are progressively gaining popularity due to their wide spectrum of biological activities when taken as a dietary input or as supplements. They are also used as anti-diarrheal, antiulcer, anti-inflammatory as well as for the treatment of diseases such as hypertension, vascular fragility, and allergies. Catechins are easily available and extractable polyphenol which possess excellent antioxidant properties and are also able to inhibit lipid oxidation in red meat, poultry and fish. Present study is an insight to the unique properties and applications of catechins derived from Acacia catechu. Present study is a brief account on catechin-based nanocomposites for advanced applications.

Keywords: *Polyphenols, catechin, antioxidant, secondary metabolites, free radical scavenger, antimicrobial properties, nanocomposites*

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INTRODUCTION

In the present scenario, 80% of the world's population is transforming from conventional therapy towards herbal remedies and their active components because of their lesser side effects [1]. One of such transformations is use of natural antioxidants. Synthetic antioxidants most commonly used in food industries include BHT (butylated hydroxyl toluene) and BHA (butylated hydroxyl anisole), which possess various detrimental effects on the human bodies. Contrary to it, there is a class of natural polyphenols which possess excellent antioxidants properties. Polyphenols are secondary metabolites of plants generally used in defense against ultraviolet radiation or aggression by pathogens [2]. According to a British physical-organic chemist Edwin Haslam, the term "polyphenol" should be used as a water-soluble plant phenolic compounds having molecular masses ranging from 500 to 3000–4000 Da and possessing 12 to 16 phenolic hydroxyl groups on five to seven aromatic rings per 1000 Da of relative molecular mass [3].

Polyphenols are inexpensive, simple to use and environment friendly natural extracts obtained generally from natural sources and because of its safety and excess availability they are more exploited compounds. Polyphenols play an important role in food industries as they replace chemical additives in foods and have antimicrobial, bio-preservative properties. They also inhibit lipid oxidation in red meat, poultry and fish which attributes for their use as natural food preservatives in food industries. Usually more than 0.3% concentration of antioxidant is required to inhibit the lipid oxidations [4, 5]. They also have complexing properties towards proteins, which favors their use for treatment of various diseases like inflammation or cancer, anti-ageing purposes in cosmetic formulations [6]. They improve the shelf life of foods and red meat as they inhibit the lipid oxidation and growth of microorganism. They are applied as methods of direct addition, spraying, dipping and coating the polyphenolic compounds [4, 5]. Polyphenolic compounds also have antiviral activities which favour their use against the flaviviruses, West Nile Virus, Zika virus and dengue virus [7]. Polyphenols are mainly categorized on basis of their chemical structure as: phenolic acids, flavonoids, stilbenes and lignans [8].

They are natural antioxidant and like vitamin C, E and carotenoids, they also have anti-inflammatory, cholesterol lowering, antiviral and antibacterial activities [2, 9]. The antioxidant effect of plant polyphenols is due to their phenolic content which is responsible to scavenge free radical and gives singlet oxygen, hydroxyl radicals and superoxide free radicals. Their redox nature allows them to act as reducing agents, hydrogen donors and metal chelators [1, 2]. Antioxidant activities of plant extract, i.e., catechin which is a natural antioxidant extracted from plants has much interest due to its multifunctional properties to prevent oxidation in complex food system. Recently, catechin is exploited for use as an additive and natural reducing system for the fabrication of advanced nanocomposite materials with inherent antioxidant behaviour.

CATECHIN

Catechin is one of the polyphenolic compounds and a secondary metabolite of herbal source which belongs to a group of flavonoids with antioxidant activities and diverse health benefits in mankind [10]. Catechin and epicatechin are major catechins which have dietary importance for human health. Owing to their inherent antioxidant behaviour, they have been extensively used as natural antioxidant for animal food since they inhibit untimely lipid oxidation. Catechins also act as useful ingredient and nutritional supplements. Epicatechin is a monomeric flavanol found naturally in fruits (apples), wine, tea and *Acacia catechu* plant. Catechin and epicatechin have the ability to quench the free radical species such as hydroxyl radicals, peroxyl radicals, superoxide radicals and free radical scavenging ability of epicatechin is ten times more than beta carotene [1–3, 5, 11]. They also have the properties of chelating the transition metal [2, 12]. Prolong heating can change the stability and antioxidant properties of catechin, as at high temperature, catechin changes into catechutannic acid, and therefore, decreases the antioxidant properties. Catechin is stable at pH <4 and unstable at pH >6. Instability in antioxidant reduces the functional value of food [13, 14].

Catechins are pharmacologically active ingredients mainly extracted from natural origin such as olives, grapes, tea leaves, ginger roots, *Bengal catechu*, *Acacia catechu*, apple peel, pomegranate, chocolate and many more [1, 2, 5, 11, 13]. The main chemical constituents of *Acacia catechu* are catechin, epicatechin, epigallocatechin etc. Extract of *Acacia catechu* exhibited antioxidant properties and is found to be an antibacterial, anti-inflammatory, analgesic and anticancer agent and used for Ayurvedic formulation and has pharmacological effect [2, 4, 9, 11, 12]. Catechin extracted from green tea (Camellia plant) is especially EGCG (-) epigallocatechin-3-gallate which has preventive effect against chronic diseases such as heart diseases, diabetes, neurodegenerative diseases, etc. [2, 12, 15].

Catechin-based Nanocomposites

Catechins possess excellent antioxidant properties as well as distinctive chemical, biological and physiological activities; but their instability towards temperature, pH fluctuations and low solubility limits their applicability as that leads to a decrease of its bioavailability properties. Various strategies have been adopted from time to time to improve their stability. Nanoencapsulation is one of such techniques which can increase the stability of catechin and therefore increases its solubility as well as bioavailability [13, 16–18]. Iron and polyphenol can form nanoparticles with size ranging from 50 to 80 nm. The iron-polyphenol nanoparticles are then utilized as Fenton like catalyst for decolourization of acid black 194 in solution, moreover there is no need to adjust the pH value of the solution [19]. Bioavailability of catechins can be improved by encapsulating polymeric system composed of poly-lactic-co-glycolic acid (PLGA) having didodecyldimethyl ammonium (DMAB) which acts as stabilizer, and SMB (Sodium meta borate) as chelator fabricated through modified emulsion-diffusion-evaporation method [20]. Kailaku *et al*. reported that the functional properties of catechin can be improved by nanoencapsulation technology using a non-toxic biopolymer chitosan. Nanoencapsulation of catechin was achieved by mixing chitosan polymer cross linked with poly-anion-sodium tripolyphosphate (Na-TPP). Best particle size of nanoencapsulated catechin is formed by mixing 0.2% concentration of chitosan solution with 0.1% concentration of sodium tripolyphosphate (NA-TPP) [13]. Vairapperumal *et al*. have synthesized Gd-doped $LaVO_4$ nanoparticles with different crystal structure and irregular rectangular nanocrystals by catechin. Magnetic properties shifted para-magnetism to supra-magnetism. Gd-doped $LaVO_4$ nanoparticles have a remarkable role in MRI applications [21].

In the present scenario, new techniques have been explored for the synthesis of nanoparticles using biological methods accompanying with the use of plant extracts, fungi, bacteria or essential oils. These are a favorable alternative to conventional methodologies as they can minimize the use of hazardous substances [22]. Green tea extracts can be used as reducing and stabilizing agent for the biosynthesis of AgNPs in an aqueous solution in ambient environment. Silver nanoparticles are easily aggregated to form larger colloids and usually require capping agents to keep the nanoparticles at nanoscale [23]. Polyhydroxy groups of catechins reduce silver ions to form metallic silver, which ultimately leads to the formation of AgNPs. Choi *et al*. reported in their study that catechin can be used as an eco-friendly reducing and capping agent for green synthesis of gold nanoparticles (AuNPs) without use of any external energy. Catechin being natural extract replaced the toxic reducing agent sodium borohydride $NaBH_4$ which is commonly used for reduction of 4-nitrophenol (4-NP) to 4-aminophenol (4-AP). 4-aminophenol is an important intermediate for the manufacture of antipyretics and analgesics. This results into good solubility and biocompatibility of resultant AuNPs [14]. Chanphai *et al*. reported that chitosan nanoparticles were prepared by ultrasonic methods and characterization was carried out by spectroscopic methods. This study reveals that in tea catechin-chitosan conjugation, there were major changes in the absorption spectra of catechin which can be used to calculate the binding constants. Binding efficiencies of tea catechin with chitosan nanoparticles show that polyphenols-polymer conjugation via hydrophilic, hydrophobic and H-bonding contacts with the order of stability that is EGCG>EGC>C. Chitosan nanoparticles transport tea catechin and increase the bioavailability of these micronutrients [17, 18]. Monika *et al*. have prepared nanocapsules through emulsion solvent evaporation technique using catechin rich extract (CRE). Antioxidant activity was measured using DPPH method. Antioxidants react with methanolic solution of 2,2-Diphenyl-1-Picrylhydrazyl (DPPH) and convert it into α,α-diphenyl-β-picryl hydrazine. The extent of decolouration indicates the scavenging power of the antioxidant [10]. Bhushani *et al*. reported that nanoencapsulation of antioxidant of bioactive compound of Zein, that is, catechin was carried out by electrospraying method which improves the stability of bioactive compounds [24]. Nasir *et al*. successfully synthesized the silver nanoparticle Ag(NP)/catechin/gelatin nanofiber by electrospinning method. Silver nanoparticle/catechin acts as bioactive component. Morphology and structure of bioactive catechin-gelatin nanofiber were characterized by scanning electron microscopy (SEM). Ag(NP)/catachin-gelatin nanofiber has potential application in cosmetic, soap, textile and medical [25, 26]. Riaz *et al*. prepared an active food packaging by apple peel polyphenols (APP) which were incorporated in chitosan to develop a biodegradable film for food packaging for food industries. They improved the physical properties of film by increasing thickness, density, opacity and swelling ratio and moisture content was decreased [27].

Pluta *et al*. have synthesized silver nanoparticles using red tea infusion as a reducing and stabilizing agent. Various spectroscopic techniques (UV-Vis, DLS, SEM and TEM) have been used to confirm the formation of nanoparticles, morphology and particle size of functionalized nanoparticles. Silver nanoparticles' synthesis proceeded most efficiently using the 7 and 10 ml of red tea infusion [22]. Sokmen *et al*. have reported that phenolic compounds, particularly, epigallocatechin gallate (EGCG) form a bilayer of (ca. 6 nm thickness structure) around silver nanoparticles during synthesis and thus establish a protective layer which suppresses particles' aggregation. Sokmen *et al*. have synthesized metallic silver nanoparticles (AgNPs) through green tea extract and microwave power using polyethylene glycol (PEG) as capping agent. Crude aqueous tea extract selectively extracted from crude extract and 2% (w/v) aqueous solutions of catechin extract have been used in microwave assisted AgNP production system. AgNPs were successfully produced by both extracts only with 0.5 ml extract volume. Catechin extract was superior when compared with crude extract as the former produced high concentration of AgNPs at average 15±6 nm particle size. This method is very simple, economical and fast for the production of AgNP. Aggregation was inhibited by the extract and therefore no capping agent was required. Microwave treatment facilitates the nanoparticle production and biomolecules present in tea extract are able to reduce silver ions to silver nanoparticles. Pure catechins display better reductive power and have great capacity[28].

Apart from nanoencapsulation of catechins, catechin based polyphenols are also exploited for the fabrication of hydrophilic networks. Sun *et al*. fabricated keratin-catachin nanoparticles (KE-NPs) and cellulose based hydrogel. The haemostatic abilities of keratin are improved by making keratin hydrogel. There are improved physical properties of Keratin and resulting keratin catechin composite (KEC) hydrogel has good adhesiveness, hem-adsorption and rapid blood coagulation. KEC hydrogel exhibited the capacity of preventing blood loss and served as a novel type of haemostatic material. These hydrogels do not cause secondary damage to the wound and reduce the bleeding amount and time compared with the control groups and cellulose groups [29]. Catechin is well-known as a very powerful antioxidant, containing the effects of anti-inflammation and skin wound healing. Kim *et al*. have synthesized cyclodextrin nanoparticles incorporated into poly(vinyl alcohol) (PVA)/pectin (PT) hydrogel. The composite hydrogels have been evaluated for the induction of re-epithelialization in skin wound. During wound healing test, the catechin nanoparticles-loaded PVA/PT hydrogel showed faster healing of the wound made in rat dorsum than the CTEC gel [30].

CONCLUSION

Plant polyphenols are having extensively increasing attention due to their powerful antioxidant properties and their efficient effect in prevention of various oxidative diseases such as cardiovascular, cancer and neurodegenerative diseases. Antioxidant activities of plant extracts are usually linked to their phenolic contents. Hydrogen donating characteristics of the phenolic compounds are responsible for the inhibition of free radical induced lipid ability to scavenge free radicals. The antioxidant activities of catechin are believed to be responsible for anti-inflammatory, tissue protectant, antihypertensive and anti-diarrheal effects. Encapsulation of polyphenols has shown to protect and increase stability, solubility as well as bioavailability of these dietary compounds and to enhance their anticancer activity. Thus, inherent nutraceutical behaviour of these polyphenols can be used for the fabrication of advanced nanocomposites with improved properties and applicability.

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