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REVIEW

An Overview of Physical Properties of Fruit Wastes Mediated Green Synthesized Transition Metal Oxide Nanoparticles and Its Applications

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Different fruit peel wastes have a scope of utilizing in green synthesis of various transition metal oxide nanoparticles (TrMONPs). This articles reveals about the physical properties and their applications of green synthesized TrMONPs mediated through fruit peel wastes. Various transition metal nanoparticles like Ag, Au, Ti, Zn, Cu, Fe, Ni, Pd have been synthesized successfully by peels of mango, banana, pomegranate, apple and other citrus fruits. The SEM analysis revealed that the most of the synthesized silver nanoparticles were spherical in the range of 10 to 15 nm and have wide applications as antimicrobial agents, sensors, conducting materials, *etc*. ZnONPs were synthesized by mediated through most of the fruit peels found in range of 20 to 50 nm (spherical shape) with applications as photocatalyst. TiO_2 nano-particles synthesized were spherical and hexagonal with high optical properties as well as UV absorbent nature targeting applications in cosmetics. The other metals *viz*. Au Cu, Ni, Pd nanoparticles synthesized mediated citrus fruit peels showed crystallinity. SEM analysis confirmed the surface morphology while XRD confirmed the high crystallinity of the metals nanoparticles. FT-IR have been widely used by the researchers for confirmation of new bond formation in nanoparticles. It is also evident that scandium and vanadium from 3*d*-series and other 4*d*-transition metals are less approachable and not used in fruit peel waste mediated green synthesis. The nanoparticles of Ni, Pd, Zr, Cu, Fe and other metals (TrOMNPs) have also been found with optimum surface area and size less than 10 nm for adsorption due to which these have application as catalyst in many organic synthesis. Other applications are biochar, bio-sorbent, carbon dots, edible films, heavy metals absorption, waste water treatment and in medicinal fields. Furthermore, approaches that can aided into development of these nanoparticles is the perspective towards incorporating green methods like laser ablation, sol-gel process, microwave heating, chemical vapour deposition and hydrothermal/solvothermal process into production of fruit wastes derived metal nanoparticles. Recent trends of researchers are towards nanosorbents for the removal of air and water pollutants along with other fields by using fruit and food wastes material.

Keywords: Fruit wastes, Transition metals, Nanoparticles, Catalyst, Nano size.

INTRODUCTION

The vast majority of human activities generate waste [1]. Nevertheless, waste creation has been major concern since ancient times. Increases in both the rate and volume of waste production have occurred in recent years, a trend that can be directly attributed to the expanding human population [2]. Unlike in earlier times, when wastes were merely an irritation to be avoided. As the population was small and there was plenty of land available at that time, appropriate management wasn't major concern. Nowadays, improper disposal of waste has become a serious environmental issue. Garbage or waste is a byproduct of human activity, but inefficient production methods also contribute to this problem by wasting resources [3]. This influx of people into cities resulted in a population explosion, which produced increment in volume and diversity of trash generated in cities. Open garbage dumps and careless trash disposal have become epidemics in populated areas [3,4]. Over the last decade, researchers have been focussing on generating health-beneficial bioactive compounds from the fruits wastes. Wastes from fruit peels are another form of food waste that contributes to the pollution of open spaces. Thus far, the synthesis of nanoparticles from fruit wastes is not common practice, but efforts in this direction have been started [4].

Waste and byproducts management in the fruit and food industries can pose challenges to environmental protection and sustainability. Animal feed and usage as a fertilizer are the two most common traditional waste utilization options. A worldwide implementation of environmentally responsible waste management practices has been facilitated by environmental laws. The waste prevention encompasses three sorts of actions: stringent avoidance, source reduction and product reuse. In contrast, waste prevention entails not only minimizing waste production but also staying clear of potentially hazardous situations [5-9].

Waste comes in all sizes and can be described in a wide variety. Many factors are taken into account when classifying trash, including its physical properties, whether or not it can be recycled or composted, the method used in its production and the extent to which it has an impact on the environment. Waste can be classified into three groups depending on their physical conditions *viz.* solid, liquid and gaseous wastes. It is evident that some countries have different classifications [10, 11]. Listed below are some of the most common types of waste (Fig. 1).

In India, 'fruit processing" industry is a rising star. Poor resource management caused us to waste between 30-35 % of the abundant raw fruit on the market every year. Deficient infrastructure facilities and poor postharvest management practices are the major causes for this loss. India's post-harvest production losses have been estimated at Rs 75,00-1,00,000 crore per year. In comparison to other affluent countries, fruit processing accounts for only about 1.8% of overall horticultural production. India has a tiny number of fruit processing infrastructures that can't handle big amounts of raw materials. Only 5166 fruit and vegetable processing plants are operating in the country as of January 1, 2019. These units don't have a system in place for managing and disposing of military waste. As a result, a lot of wasted resources are expended processing raw fruit in order to extract the intended value product using other ingredients when people do not consume it themselves. In developing countries like India, where food waste is a major problem, these byproducts are often wasted despite their high value and nutritional contents [12-14]. Fruit waste is generally categorized in two types like liquid waste (juice & washed water) and solid waste (peels, seeds, skin, *etc.*). Some fruit have discarded option like 35-50% mango, pineapple 45-50%, orange 40-50% and banana 25-30%. And even though fruit spoilage is a major problem, leading to rodents, flies and unwelcome microbiological growth in the processing facilities. So, certain plans to be made to utilize this waste; alternatively, it should be buried in soil far from the processing facility. Table-1 lists some of the fruit processing facilities available in India [15]. In recent years, scientists have developed several strategies to recycle fruit scraps and cut down on fruit waste. In present study, the synthesis of different metal nanoparticles by using fruits waste, utilization of fruits waste and its applications in various fields has been discussed.

TABLE-1
AVAILABILITY OF FRUIT PROCESSING WASTE IN INDIA [15]

Fruits	Nature of Waste	Approximate waste (%)	Potential quantities of waste (tones)
Mango	Peel & stones	45	3144.5
Banana	Peel	35	823.4
Citrus	Peel, rag & seed	50	606.0
Pineapple	Skin & core	33	24.7
Apple	Pomace, seeds & peel	–	412

Synthesis of metal nanoparticles using fruits waste: Metal nanoparticles have been produced by using both bottom-up and top-down methods. Metal nanoparticles are synthesized from starting through atoms or molecules in the bottom-up approach, while in the top-down approach, metal nanoparticles are produced from bulk metal precursors without any atomic or molecular level control [16]. Metal nanoparticles can be made in a biologically efficient, environmental friendly, cost-effective and non-toxic manner [17,18]. Biogenic systems such as plants extract, fungi, algae and bacteria, among others, are utilized as stabilizing and reducing agents to synthesize metal nanoparticles in this approach [19]. Metal ions are converted into neutral atoms by proteins from these sources, which then

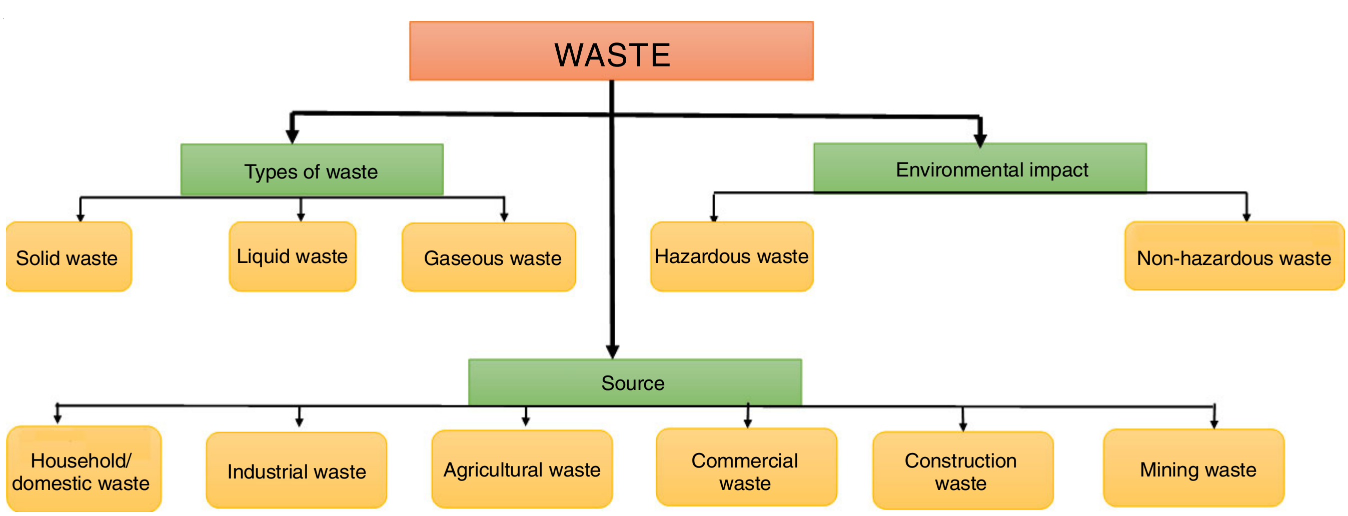

Fig. 1. Classification of waste

stabilize the nanoparticles [20] followed by aggregation and growth. In the physical approach, metal precursors are abraded, melted, evaporated or condensed using any physical energy source like high-energy radiations, thermal or electrical energy.

One of the most common sorts of municipal wastes is fruit trash. Surprisingly, useful organic chemicals can be found in these byproducts. To determine whether the bioactive components present in the fruit wastes could be exploited, they were tried out as reducing and capping agents in the synthesis of nanoparticles [21]. The major capping and reducing agents are polyphenols found in fruit wastes like husks, exterior coverings and shells. Nanoparticles are stabilized by utilizing chemical based on the production of a new C–O group present in the carboxylic acid, aldehyde or ketones. When an electron is transferred to a metal ion, the ions are reduced and nanoparticles are formed as a result. The easy availability of bioactive chemicals has opened up new possibilities for green nanomaterial fabrication. Various methods for preparing and reacting extract solutions with metal ions have been used and aqueous fruit waste extract has been used to manufacture a variety of sizes and forms [22].

Procedure for preparation of extract of fruit wastes: Several methods *e.g.*, maceration, digestion, decoction, infusion, percolation, Soxhlet extraction, superficial extraction, ultrasound-assisted and microwave-assisted extractions [23-25] have been used for the extract production using different types of polar and non-polar solvents. Table-2 represents data regarding synthesis of nanoparticles using different fruit waste under various physiological and environmental conditions. The majority of metal nanoparticles were made up of Ag, Au, Ti, Fe, Pd, Cu and Zn. The characteristic study of respective nanoparticles revealed different functional groups such as carboxylic acid, phenolic groups, amine groups, flavonoids, terpenoids that help them in enhancing reduction & stabilization properties. Fruits like orange, banana, pomegranate, *etc.* were used for the successful synthesis of different morphologically distinct nanoparticles.

TABLE-2
TYPES OF NPs, FRUIT SCRAPS, EXTRACTION METHOD, REACTION'S TIME, SIZE & SHAPE AND FUNCTIONAL GROUPS INVOLVED IN SYNTHESIS OF NANOPARTICLES

NP's type	Fruit scraps	Extraction method	Time of reaction	Size of nano-particles	Shape of nanoparticles	Functional group involved in reduction and stabilization	Ref.
ZnONPs	Jujube fruit waste	Grinded, dried, refluxed at 80 °C (steam extraction)	4 h	29 ± 8 nm	Spherical	Carboxylic group, phenolic group	[26]
AgNPs	*S. lycopersicums* fruit extract	Crushed, separated by centrifugation at 1000 rpm	2 h	10 nm	Spherical	Flavonoids, provitamin A, kaempferol	[27]
PdNPs	Oak fruit bark extract	Dried, chopped, powdered, boiled for 20 min	48 h	5 nm	Spherical	Phenolic hydroxyl groups of flavones, terpenoids and tannis	[28]
AgNPs	*Alpinia nigra* fruits	Dried, heated in double distilled water at 50 °C	2 h	6 nm	Spherical	Phenolic group, hydroxyl group	[29]
AgNPs	*Phyllanthus emblica* fruit extract	Dried, boiled for 10 min	15 min	30 nm	Hexagonal	Alkynyl group, carbonyl group, amide group	[30]
ZnONPs	*Amomum longiligulare* fruit extract	Grinded, diluted, autoclaved at 100 °C	2 h	50 nm	Tetragonal	Amines, carboxylic group	[31]
ZnONPs	*Punica granatum* fruits peels extract	Powdered, extracted in DI water at 65 °C	1 h	S3-32.98 ± 8.63 S4-81.84 ± 24.5 nm	Spherical & hexagonal	Carboxylic group, amines	[32]
AgNPs	Dragon fruit peel extract	Heat and dried, extracted with water at 100 °C, filtered	24 h	25-26 nm	Spherical	Phenolic group, carboxylic acid, amines	[33]
AgNPs	Araza fruit extract	Chopped, heated (60-62 °C) in 50 mL of Milli-Q water for 60 min.	1 h	15-45 nm	Crystalline and spherical	Carboxylic acid, ester group, amide	[34]
CuNPs	*Citrus sinensis* aqueous fruit extract	Washed, squeezed, centrifuged for 10 min at 5000 rpm	15 min	10.2 nm	Crystalline	Flavanones, amino acids, terpenoids	[35]
AgNPs	*Citrus limon, Citrus sinensis, Citrus limetta*	Dried, boiled below 60 °C for 0.5 h, filtered	30 min	9-46 nm	Spherical	–	[36]
AgNPs	*Nauclea latifolia* fruit extracts	Blended, extracted using methanol and water, filtered	24-72 h	12 nm	Irregular	Hydroxyl, carboxyl, phenols, amines, amide groups	[37]
AgNPs	*Glycosmis pentaphylla* fruit extract	Dried in hot- air-woven at 40 °C, crushed, extracted with 100 mL of 80% for 24 h at 30 °C	24 h	17 nm	Spherical	Carbonyl groups	[38]
$CaCO_3$NPs	Orange fruit peels	Washed, chopped, boiled for 30 min, centrifuged for 4000 rpm for 20 min	24 h	80-90 nm	Rhombohedral	Aragonite polymorphs	[39]

CuNPs	*Ziziphus spina-christi* (L.) Willd	Washed with DI water, dried, heated in water bath to 80 °C	1 h	5-20nm	Spherical	Carboxylic group, phenolic group	[40]
CuNPs	*Solanum macrocarpon* fruit extract	Shredded, dried, boiled for 2h in 100 mL distilled water	1 h	35.6 ± 6.24 nm	Hexagonal & spherical	–	[41]
AuNPs	*Annona squamosa* L peel extract	Dried in air, powdered, heated for 30 min at 60 °C, filtered	5 min	5 ± 2 nm	Spherical	Hydroxyl and carbonyl groups	[42]
AgNPs	*Cleome viscosa* L fruit extract	Cleaned by DD water, boiled with 100 mL distilled water for 30 min at 60 °C, extracted	24 h	20-50 nm	Spherical	Phenolic group, amino acids, alkaloids, tannins	[43]
AgNPs	Poemegranate and watermelon peel extracts	Washed, dried in an oven for 3 h at 105 °C, grounded, sieved, filtered	24 h	PPAgNPs: 15-70 nm WPAgNPs: 20-85 nm	Spherical	Flavonoids, phenolic group, amides	[44]
$CoFe_2O_4$	Dragon fruit (*Selenicereus undatus*) peels	Washed with excess water, ethanol and air-dried	24 h	500 nm	Spherical	Phenolic group, carboxylic group	[45]
AgNPs	*Terminalia bellirica* fruit extract	Dried, grinded, powdered dissolved in 200 mL ethanol overnight, extracted	2 h	10-50 nm	Spherical	–	[46]
TiO_2NPs	*Annona squamosa* peel extract	Dried, powdered, 4 g of powder mixed with 40 mL of DD water at 25 °C, extracted	6h	23-25 nm	Spherical	Hydroxyl group	[47]
FeNPs	*Citrus maxima* peels aqueous extract	Milled, boiled for 80 min. at 353 K, filtered	80 min	10—100 nm	Irregular	Hydroxyl group, aldehydes, ketones, terpenoid	[48]
Fe_3O_4	Plantain peel extract	Dried, added 100 mL of DD water & refluxed at 80 °C for 60 min. & filtered	2 h	>50 nm	Spherical	Carboxylic group, amine group	[49]
AuNPs	Banana fruit waste extract	Washed, powdered, boiled, boiled for 5 min in a 500 mL conical flask	20 min.	<20 nm	Spherical	Hydroxyl group, carbonyl group	[50]
TiO_2NPs	Orange fruit waste	50 g of orange peel taken in a beaker & extracted with 150 mL of water at 90 °C for 2 h	3 h	19 nm	Tetragonal	Phenolic group	[51]
NiO nano-crystals	Rambutan (*Nephelium lappaceum* L.) peel extract	Washed, dried in oven at 50 °C, boiled with ethanol & DD water for 10 min, filtered	8 h	50 nm	–	Phenolic hydroxyl groups	[52]

Fruit peel-derived metallic nanoparticles: The bioactive chemicals, *e.g.* amino acids, enzymes, alkaloids, phenolics, tannins, vitamins, and terpenoids are found in fruit and vegetable wastes and act as reducing agents in the formation of metal nanoparticles [53,54]. Some biomolecules act as modelling agents, driving particle growth in one direction while others act like a capping agent, preventing nanoparticles agglomeration. Biosynthesizing nanoparticles from fruit peel waste has recently emerged as a reliable, sustainable and environmental friendly alternative to conventional manufacturing protocols that use toxic chemicals and solvents, thereby reducing the risk to human health and the environment. Because of their unique physico-chemical features and uses in numerous domains of biomedicine and pharmaceuticals, there has been a lot of interest in using nanoparticles. The biogenic nanoparticles are synthesize *via* a bottom-up technique, in which atoms and compounds used as building blocks that self-build to generate the end product. Table-3 displays the types of metal nanoparticles that are synthesized from fruit peel extracts [55-58].

Carbon dots derived from fruit peels: Carbon dots are a sort of photoluminescent material that may be synthesized top-down or bottom-up and are very small in size (< 10 nm) [67,68]. A big carbon structure is broken down in the top-down synthetic approach by utilizing laser ablation, electro-oxidation or acid assisted chemical oxidation in the synthesis process. However, one of the disadvantages of this strategy is that it requires a complex and harsh synthetic environment. Bottom-up technique, which relies on plants and its byproducts rather than chemicals, appeared to be superior to the top-down technique. Fruits peel waste treatment must include the right processes for synthesizing carbon dots as a carbon source, including pyrolysis at high temperatures, oxygenolysis with concentrated acids, polymerization, carbonization and oxidation [69,70]. Applications in heavy metal detection, photocatalytic activity and sensing are only few areas where CDs prepared from fruits peel waste have shown as promising material (Table-4).

Fruit peel derived biochar: Biochar has been produced using several forms of food waste and its yield and physico-

TABLE-3
FRUIT PEEL DERIVED METAL NANOPARTICLES

Name of fruit	Types of nanoparticles synthesized	Reaction time	Shape (size)	Applications	Ref.
Orange, banana, pomegranate and apple	Ag	2 min	Spherical (25 nm)	Antibacterial action against *E. coli*, *Salmonella* sp., Antioxidant activity using DPPH; MCF-7 cell lines showed anticancer activity.	[59]
Lemon	Ag	30 min	Spherical (2-5 nm)	Antifungal action against *Candida albicans* in *Streptococcus mutans*, *Proteus mirabilis* and *Acinetobacter baumannii*	[60]
Apricot	Ag	–	Rod shaped (50 nm)	Antibacterial action against *S. aureus*, *B. subtilis*, *E. coli*	[61]
Cavendish banana	Ag	30 min	Spherical (55 nm)	*K. pneumonia*; Antioxidant activity (DPPH)	[62]
Orange, grapefruit	ZnO	1 h	Hexagonal (12.55 nm; 19.66 nm)	Photocatalytic against MB (methylene blue)	[63]
Banana	Ag	1 h	Spherical (23.7 nm)	Antibacterial action against *E. coli*, *S. aureus*, *B. subtilis* and antifungal against *C. albicans*	[64]
Pomegranate	Ag	24 h	(5-50 nm)	Antibacterial action against *P. aeruginosa*, *E. coli* & *S. aureus*	[65]
Orange	Ag	5 h	(48.1 nm)	Antibacterial activity against *Xanthomonas axonopodispv. citri* (Xac)	[66]

TABLE-4
FRUIT PEELS ARE USED AS A CARBON SOURCE IN THE PRODUCTION OF CARBON DOTS

Name of fruit	Conditions for production	Applications	Ref.
Mango	Hydrothermal treatment; time - 2 h; temp. - 300 °C	Detection of Fe^{2+} ion using cellular labelling	[70]
Pineapple	Hydrothermal treatment; time - 3 h; temp. - 150 °C	Security gadgets that are electronic auantification of Hg^{2+} ion	[71]
Pomelo	Hydrothermal treatment; time - 3 h; temp. - 200 °C	Sensing of Hg^{2+}	[72]
Pomegranate	Hydrothermal treatment; time - 36 h; temp. - 180 °C	Latent prints can be recovered	[73]
Orange	Hydrothermal treatment; time - 10 h; temp. - 150 °C	Photocatalytic activity	[74]

chemical qualities have been thoroughly documented. Biochar is commonly used to remove several sorts of heavy metal containing pollutants from contaminated water bodies. Several studies show that various fruit peel waste can be converted into biochar and these results are summarized in Table-5 [75,76].

Fruit peel derived biosorbents: Biosorbents contain a wide variety of functional groups that can bind to and remove toxins from the environment, however these groups depend on the functional groups and the type of biosorbent [83]. The removal of methylene blue dye from aqueous solutions has been the focus of numerous studies aimed at developing bio sorbents from the peels of fruits including apple, pineapple, and dragon fruit and the details are shown in Table-6.

Fruit peel based edible films/coatings: Edible coatings are thin layers placed to surface of food to enlarged its shelf life and conserve its quality, attributes and functionality at a low-price. Its use can be made more useful by extending its storage life, preventing bacterial spoilage and acting as a carrier

TABLE-5
FRUIT PEEL DERIVED BIOCHAR WITH ITS APPLICATIONS

Name of fruit	Conditions required for formation of Biochar	Applications	Ref.
Banana	2 h of hydrothermal carbonization at 230 °C	Exhibited magnificent lead clarification capability of 193 and 359 mg/g	[77]
Pomegranate	2 h of pyrolysis at 300 °C	Cu(II) adsorption-52 mg/g	[78]
Rambutan	3 h of pyrolysis at 600 °C	Cu(II) adsorbent doses of 0.2 and 0.4 g/L were used to extract Cu(II) from aq. solutions containing 50 and 100 mg/L.	[79]
Pomelo	1 h of pyrolysis at 450 °C	57.637 mg/g of Cr (VI) was adsorbed by 0.05 g of Biochar	[80]
Pineapple	2 h of pyrolysis at 200 °C and heated for 3 h at 650 °C	Oxytetracycline absorption	[81]
Litchi	12 h of hydrothermal carbonization at 180 °C	Malachite green and Congo red have adsorption capacities of 404.4 and 2468 mg/g, respectively.	[82]

TABLE-6
FRUIT PEEL-BASED BIOSORBENTS AND ITS APPLICATIONS

Name of fruit	Time of drying (h)	Temp. (°C)	Applications	Ref.
Dragon fruit	24	104	Methylene blue (MB) was captivated at 192.31 mg/g in a dose of 0.06 g.	[84]
Banana	5	60	Removed 90% cadmium (II) and 90% lead(II) ions	[85]
Apple	24	60	MB was absorbed at a rate of 107.52 mg/g.	[86]
Pineapple	48	72	MB was absorbed at a rate of 97.09 mg/g.	[87]
Grapefruit	24	105	Copper ion adsorption: 52.48 mg/g	[88]

TABLE-7
FRUIT PEEL DERIVED EDIBLE FILMS/COATINGS INCLUDING THEIR APPLICATIONS

Name of fruit	Usable to food items	Valuable effects	Ref.
Orange	Cupcake	In refrigerated storage to one-week, peroxide value increased by 3.60-4.80 (mL.eq./kg fat) & microbial growth decreased.	[91]
Apple	Fresh beef patties	On raw beef patties, lipid oxidation is completely inhibited & microbial development is effectively suppressed. Also, sensory properties of cooked & raw beef patties are unaffected.	[92]
Pomegranate	–	In comparison to control mung bean protein films, films supplemented with pomegranate peel demonstrated greater total, antioxidant &antibacterial activity & also had greater phenolic content	[93]
Orange	Shrimps	With shelf-life extension of roughly 6 days, gelatin coating in combination with orange peel essential oil retained small quality in cold storage.	[94]
Lemon	Strawberry & Tofu	Each ofedible coating agents was greatly reduced in degradation when 0.6% LPEO was added to tofu & 1% LPEO was added to strawberry.	[95]

mould for antimicrobial compounds. Coating might be considered as a viable preservation method for perishable fruits and vegetables during transport, especially those that are vulnerable to microorganisms, insects and pre- and post-harvest conditions. Fish gelatin is widely considered to be a valuable biopolymer material for biofilm formation due to its biodegradability and high myofibrillar concentration of protein. As a result of differences in amino acid sequence, mammalian fruit peel and gelatine based films have reduced water accessibility. Pomegranate peel powder added to gelatin films considerably improved water vapour permeability [89,90] due to the film matrix's inability to fully degrade the peels, resulting in the formation of more heterogeneous microstructure. Pomegranate peel has hygroscopic properties, however the moisture content of film is unaffected because hydrophobic and hydrophilic components counteract them (Table-7).

Conclusion

The green synthesis of a various transition metal oxide nanoparticles (TrMONPs) formed from fruit wastes and their numerous applications has inspired researchers to work towards the fabrication of safe nanoparticles and the repurposing of otherwise useless waste materials. The development of TrMONPs has been vigorously pursued and other fruit byproducts have been utilized as capping and reducing agents. Further selected transition metal nanoparticles like Ti, V, Zr, Cr, Ni, Pd have wider scope for application as catalyst due to size less than 10 nm and larger surface area to volume ratio for adsorption at the outer surface. While other TrOMNPs have comparatively size in the range of 10 to 60 nm have good electrical and magnetic properties due to which they can be used for producing magnetic field or can be used conductor or as super conductor. Nanoparticles formulated using unwanted fruit wastes open up into new methods of sustainable waste managements. Using different green synthesis techniques, it gave information regarding their morphological properties and their wide applications in pharmaceutical fields. Majority of the works were carried out on citrus, *Myrica esculenta*, orange, pomegranate, apple and banana peels waste for synthesizing TrMONPs. Yet, some fruit wastes, such as plum fruits, have been investigated to a lesser extent. The researchers may have been influenced to employ ordinary fruit waste due to its simplicity of manufacturing or seasonal availability.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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