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Review Article

A Review on Bio-Synthesized Co_3O_4 Nanoparticles using Plant Extracts and Their Diverse Applications

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Abstract: Developing a rapid, reliable and eco-accommodating methodology for the synthesis of metal/metal oxide nanoparticles (NPs) is an important step in the area of nanotechnology. Cobalt oxide nanoparticles (Co_3O_4 NPs) have been widely studied due to their potential applications including, antibacterial, antifungal, electrochromic sensors, heterogeneous catalysis, and energy storage devices. Due to the large rate of perilous chemicals employed in the physical and chemical production of these NPs, green methods employing the use of plants, fungus, algae, and bacteria have been adopted. However, plant-mediated synthesis of metal NPs has been developed as a substitute to defeat the restrictions of conventional synthesis approaches such as physical and chemical methods. Biomolecules, such as tannins, saponins, proteins, amino acids, steroids, enzymes, flavonoids, and vitamins from several plant extracts have been used as a stabilizing and reducing agents for the synthesis of Co_3O_4 NPs. Recently, several attempts were made to develop plant-mediated synthesis methods to produce stable, cost-effective, and eco-accommodating Co_3O_4 NPs. In this review, a comprehensive study was conducted on synthesis, characterization, and various applications of Co_3O_4 NPs produced using various plant sources.

Key words: Green synthesis, Plant extracts, Nanotechnology, Co_3O_4 NPs, Applications.

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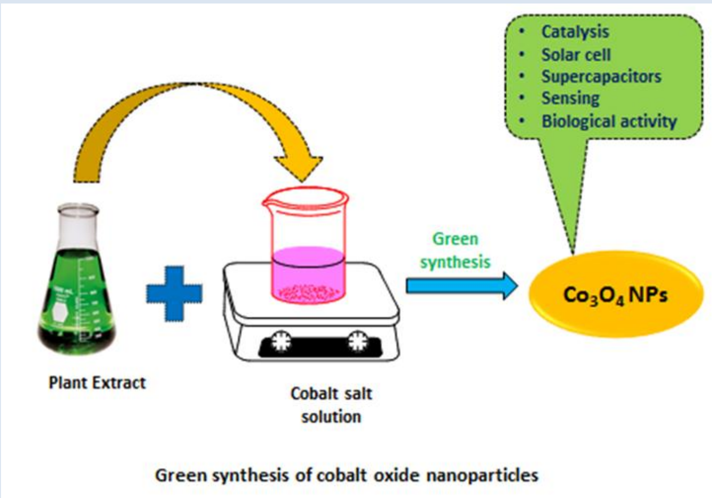
Graphical Abstract:



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Biography

Trupti Pagar was born in Surgana (Maharashtra), in 1997. She completed her B.Sc. (2019) degree from Arts, Science and Commerce College, Surgana, Savitribai Phule Pune University, Maharashtra. At present she is pursuing her M. Sc. degree in organic chemistry from Department of Chemistry, G.M.D Arts, B.W Commerce and Science College, Sinnar (Savitribai Phule Pune University) Maharashtra, India. Her area of research interest is synthesis of nanomaterial and their applications in biological activity

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Shreyas Pansambal obtained his PhD (2019) degree from Pune University in Chemistry under the supervisor of Dr. K. K. Deshmukh. Later on, he is working as an Assistant Professor at S.N. Arts, D.J.M. Commerce and B.N.S. Science College, Sangamner, Savitribai Phule Pune University, Maharashtra. He has been actively involved synthesis of heterocyclic compounds using nanocatalyst.

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1. Introduction

Nowadays, noble metal nanomaterial are well known for their creation and miraculous applications in the field of solar cells, space industry, biomedicines, sensors, catalysis, cosmetics, health care, fuel cells, electrochemistry, biotechnology, energy device, agriculture, imagines, food technology, optics and optical devices, pharmaceuticals, textile industry, and water treatment [1-32]. Among the metal oxide nanoparticles, Co_3O_4 NPs has attracted numerous research interests due to its low cost and good electro-activity. However, cobalt is a transition metal and is capable of exhibiting several possible oxidation states (Co^{2+}, Co^{3+}, and Co^{4+}). Therewithal, Co_3O_4 is a known multifunctional, antiferromagnetic p-type semi-conductor with spinel crystal structure [33]. The direct optical band gaps of Co_3O_4 NPs were about 1.48 and 2.19 eV [34] and it can be used as photocatalyst to degrade several organic pollutant using visible light. Cobalt can be found in different spin states in its oxide forms such as low, high, and intermediate spin. These spin states make the physics of the Co_3O_4 attractive from a fundamental viewpoint and in spintronic applications [33]. Hence, Co_3O_4 NPs finds immense applications (Figure 1) in areas such as field effect transistor [34], energy storage [35], catalysis [36], anode material in Li-ion rechargeable batteries [37], electrochromic sensors [38], solar cells [39] and photocatalyst [40]. In addition, Co_3O_4 NPs have antibacterial, anticancer, antioxidant, antifungal, and enzyme inhibition properties due to their superior biomedical applications [41-42].

Heretofore, several physical and chemical techniques have been applied for the synthesis of Co_3O_4 NPs. Table 1 summarizes these synthetic routes of Co_3O_4 NPs. Nevertheless, the aforementioned synthetic routes are accompanied by certain disadvantages including, being costly, time and energy consuming, need of special instruments for experimental work and being environment unfriendly that may create a negative impact on the environment [1,4-5]. To overcome the problem of an energy imbalance and toxic wastes, some greener and eco-accommodating methods have been proposed. Also, there is a need to develop and utilize safer synthetic methods, which should be simple, nontoxic, clean, and efficient with low cost. Biological resources such as plants and microorganisms can be used in a swift, effective, simple, and economical way to produce the desired Co_3O_4 NPs. Plant mediated biosynthesis of Co_3O_4 NPs has been successfully done. Hence, biosynthesis of NPs is evolved into a significant offshoot of nanotechnology. Plant derived materials are used for the fabrication of NPs, and is credible alternatives to the physical and chemical methods [4]. The use of plant extract eliminates the laborious and complicated protocols of physicochemical methods [5].

The aim of the present review is to focus on the current knowledge concerning the capability of plant materials for biogenic synthesis of Co_3O_4 NPs and presents a database that future researchers may be based on the green synthesis of Co_3O_4 NPs using plants material sources.

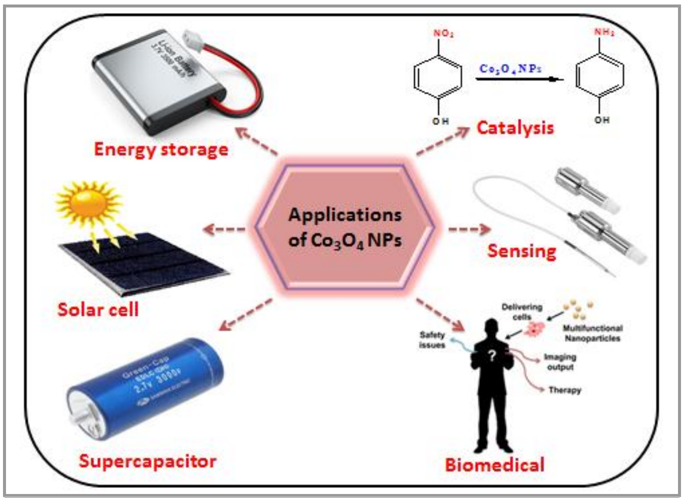


Figure 1. Various applications of Co_3O_4 NPs.

Table 1. Various methods for synthesis of Co_3O_4 NPs

Entry	Name of the Synthetic Process	Reference
1	Hydrothermal reaction	[43]
2	Thermal decomposition	[44]
3	Solution combustion	[45]
4	Microwave assisted	[46]
5	Microemulsion method	[47]
6	Spray pyrolysis	[48]
7	Vapor deposition method	[49]
8	Sono-chemical	[50]
9	Co-precipitation	[51]
10	Mechanochemical processes	[52]
11	Ionic liquid assisted method	[53]
12	Reflux method	[54]
13	Polyol	[55]
14	Pulsed laser deposition	[56]
15	Sol gel	[57]
16	Template method	[58]
17	Solvothermal	[59]
18	Irradiation technique	[60]
19	Chemical reduction method	[61]
20	Casting technique	[62]
21	Wet synthesis	[63]

2. Green Synthesis of Co_3O_4 NPs

Currently, green synthesis of metal oxide NPs has been an emerging research area in the field of green chemistry and nanotechnology. The significance of the green synthesis (Figure 2) over chemical and physical methods is: one pot and clean synthesis, environmentally gracious, cost effective, swift, facile and easily scaled up for large scale syntheses of NPs, furthermore there is no need to use high amount of temperature, energy, pressure and toxic chemicals [1, 4-5]. Using plant material for synthesis of Co_3O_4 NPs has received a great deal of attention due to its environmentally gracious, simple, rapid, non-toxic, and economical protocol which provides a single step technique for the green synthesis processes [33,36]. stabilizing the Co_3O_4 NPs by combination of biomolecules such as amino acids, saponins, enzymes, proteins, steroids, phenols, tannins, vitamins, sugar and flavonoids, which are already presented in the plant extracts having medicinal importance and are environmental benign [40,46]. Some plants are already reported to facilitate Co_3O_4 NPs (Table 2). Various parts of plant such as leaf, latex, flower, root, seed, peel, and fruit can be used for synthesis of Co_3O_4 NPs with different morphologies and sizes by biological approaches. The water soluble chemical components are mainly accountable for generation and stabilization of NPs. Thereafter, the synthesized NPs need to be characterized using numerous techniques.

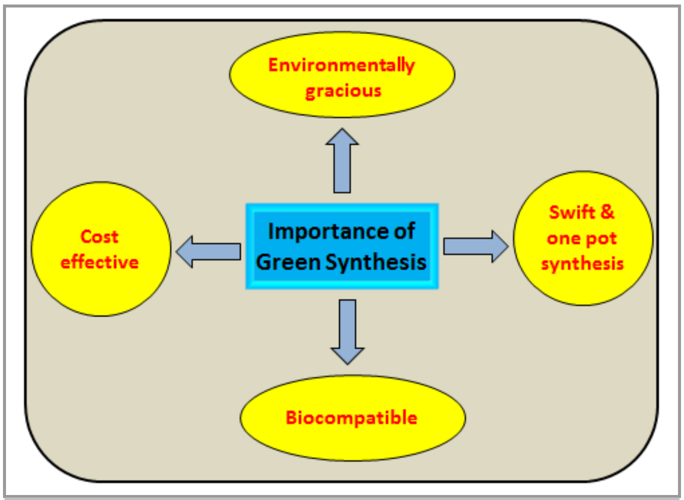


Figure 2. Importance of green synthesis of metal oxide nanoparticles.

Table 2. Green synthesis of cobalt oxide NPs using different plant source with morphology and size.

Entry	Name of Plants	Part	Shape	Size	References
1	Aspalathus linearise	Leaf	Quasi spherical	3.57 nm	[33]
2	Azadiracta indica	Leaf	Quasi spherical	1-7 nm	[36]
3	Colotropis giganta	Leaf	Spherical	50-60 nm	[39]
4	Colotropis procera	Latex	Quasi spherical	3-5 nm	[64]
5	Euphorbia heterophylla	Leaves	Spherical	69.75 nm	[40]
6	Gingko	Leaf	Irregular	30-100 nm	[38]
7	Helianthus annus	Leaves	Plate	-	[65]
8	Hibiscus Rosa-sinensis	Flower	Regular	40.05-61.37 nm	[42]
9	Manihot esculenta crantz	Root	Octahedron	36 nm	[66]
10	Moringa oleifera	Leaf	Cubic	38 nm	[67]
11	Piper nigrum	Seed	Spongy triangular	30-60 nm	[68]
12	Punica granatum	Peel	Granular	46 nm	[69]
13	Punica granatum	Peel	Spherical	43.78-73.10 nm	[70]
14	Sageretia thea	Leaf	Cubic	20.03 nm	[41]
15	Sechinum edule	Fruit	Irregular	39 nm	[71]
16	Taraxacum officinal	Leaves	Spherical	50-100 nm	[72]

3. Protocol for Biogenic Synthesis of Co_3O_4 NPs

The use of plants as the production assembly of Co_3O_4 NPs has drawn peculiar attention, because of its rapid, clean, ecofriendly, non-pathogenic, and economically affordable protocol, providing a one pot synthesis protocol. The reduction and stabilization of Co_3O_4 NPs by combination of biomolecules such as tannins, proteins, amino acids, enzymes, polysaccharides, sugars, alkaloids, phenols, saponins, terpenoids and vitamins which are naturally occurred in the plant extracts having medicinal values and are eco-accommodating. A few numbers of plants are reported to facilitate Co_3O_4 NPs synthesis are presented in Table 2. The protocol for the NPs synthesis involves: collection of the part (leaf, latex, flower, root, seed, peel, and fruit) of plant of interest from the available sites, washing them twice or thrice with ordinary water to remove the epiphytes and associated debries. Then they are washed with the sterile distilled water to remove unwanted entities if any. These clean and fresh plant parts are either dried or grinded to form the fine powder using domestic blender or used directly to obtain extract. For the plant broth preparation, around 10 g of the dried powder or fresh material is boiled with 100 mL of deionized distilled water. The resulting extract is then filtered thoroughly until no insoluble material appeared in the broth. Then addition of cobalt metal salts precursor into plant broth follow the reduction of cobalt ion to form Co_3O_4 NPs. There is no need to add external chemicals such as reducing agent or stabilizers, simply extract is mixed with cobalt salt solution and the phytochemical present in extract acts as a stabilizing agent for the synthesis of Co_3O_4 NPs. The detailed protocol of green synthesis of Co_3O_4 NPs by *Aspalathus linearis* leaves extract is described by authors reported in [33]. The synthesized Co_3O_4 NPs solution is further centrifuged to separate the NPs, and then washed thoroughly with suitable solvents. A fine powder of NPs is obtained and collected for further characterization purposes.

4. Characterization Techniques for Co_3O_4 NPs

Generally, two main approaches are used for the synthesis of nanoparticle: top down and bottom up approach. The top down approach deals with material size reduction of particles via the physical and chemical process to produce fine nanoparticles. The size, shape, overall physiochemical properties, and surface structure are processed throughout the process [1,5,73]. Bottom-up approach deals with engineering at atomic, molecular level [1]. Co_3O_4 NPs of different size, shape, surface area are characterized by various techniques such as UV-visible spectroscopy (UV-vis), thermogravimetric analysis (TGA), powder X-ray diffraction (XRD), Brunauer-Emmett-Teller (BET), Fourier transform infrared spectroscopy (FT-IR), photoluminescence (PL), dynamic light scattering (DLS), energy dispersive spectroscopy (EDS), Zeta potential, scanning electron microscopy (SEM), transmission electron microscopy (TEM), and atomic force microscopy (AFM) (Figure. 3). Crystal size, plane, structure, phase and composition are also determined from spectroscopy technique like UV-vis, XRD, FT-IR, DLS, EDS and Raman. Element composition is the determination by EDS analysis. DLS analysis estimates the size distribution and quantifies the surface charges of NPs. BET can be used to analyze the surface area and pore size of the NPs. XRD analysis can identify the crystallite size of NPs. FTIR spectroscopy identifies the surface residues and functional groups like flavonoid, phenol, hydroxyls which attach to the surface of nanoparticles during the course of their synthesis for their efficient reduction and stabilization.

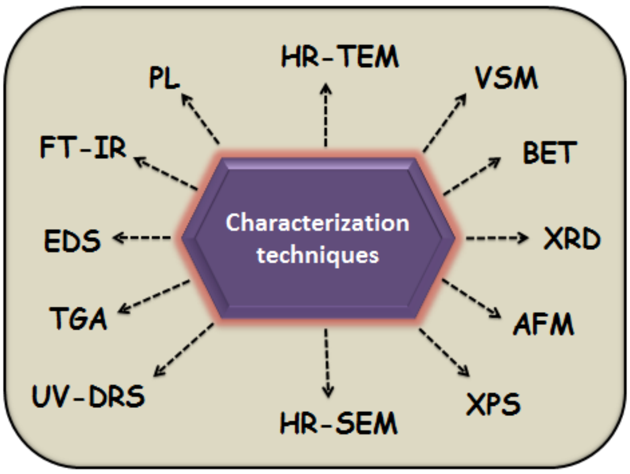
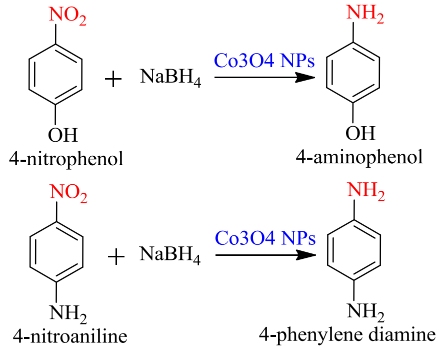


Figure 2. Characterization techniques for NPs.

5. Various Applications of Biogenically Synthesized Co_3O_4 NPs

Co_3O_4 NPs have many applications in various fields. However, the sensing, photocatalytic, and antimicrobial activities of the biosynthesized Co_3O_4 NPs are very prominent nowadays. Accordingly, we have described their potent applications as guidance to new researchers for future prospects (Table 3).

Judhit Vijaya *at al.* [36] synthesized 15 nm Co_3O_4 NPs using *Azadirechta indica* leaves extract and reported for the antimicrobial activity against gram positive and gram negative bacteria. Also, they applied Co_3O_4 NPs as a catalyst for the hydrogenation 4-nitrophenol and 4- nitroaniline (Scheme 1).



Scheme 1. Catalytic hydrogenation 4-nitrophenol and 4- nitroaniline using Co_3O_4 NPs.

Sharma and co-workers demonstrated the *Calotropis gigantea* aqueous extract mediated Co_3O_4 NPs were in the size of 50-60 nm and investigated electro-catalytic reduction of I_3^- to I^- ions in dye sensitized solar cells and thermal decomposition of ammonium perchlorate [39].

Sharma and co-workers reported biosynthesis of Co_3O_4 NPs using latex of *Calotropis procera* were in the size of 3-5 nm and analyzed the antibacterial activity of the prepared nanoparticles [64]. Yulizar and co-workers synthesized Co_3O_4 NPs using *Euphorbia heterophylla* leaf extract and studied the photocatalytic activity of synthesized NPs. The synthesized NPs are spherical shape with the size ranges from 69.75 nm. They evaluated the photocatalytic activity of green synthesized Co_3O_4 NPs using methylene blue dye and results indicate Co_3O_4 NPs exhibits good photo catalytic activity (degraded up to 63.10%) under visible light irradiation [40]. Liu and co-workers described biosynthesis of Co_3O_4 NPs using leaf extract of *Gingko* were in the size of 30-100 nm. They successfully utilized Co_3O_4 NPs as direct electrochemical sensing interface for non-enzymatic detection of H_2O_2 and glucose [38]. Saeed and co-workers synthesized plate shape Co_3O_4 NPs using *Helianthus annus* leaf extract and studied the photocatalytic activity of synthesized Co_3O_4 NPs using methyl orange dye [65].

Raji and co-workers reported the biosynthesis of Co_3O_4 NPs using *Hibiscus Rosa-sinensis* flower extract and also studied the size dependent antibacterial and antifungal activity of the Co_3O_4 NPs. The average particle size of the Co_3O_4 NPs was reported at the range of 40.05-61.37 nm. They showed that, Co_3O_4 NPs possess significant antibacterial activity against the gram positive and gram negative bacteria such as *Staphylococcus aureus*, *Streptococcus mutans*, *Klebsilla pneumonia*, *E. coli* and antifungal activity against *Aspergillus flavus*, *Aspergillus niger*. From the antibacterial and antifungal studies, it was found that Co_3O_4 NPs can be useful for developing decent fungicidal and antibacterial formulations [42].

Matinise and co-workers described green synthesis of cubic Co_3O_4 NPs using leaf extract of *Moringa oleifera* were in the size of 38 nm and reported as an electrode material for supercapacitors [67]. Saravanakumar and co-workers synthesized spongy triangular Co_3O_4 NPs using *Piper nigrum* seed extract and studied the photocatalytic activity (degraded up to 41%) of the synthesized Co_3O_4 NPs using crystal violet dye [68]. Moreover, Bibi and co-workers synthesized Co_3O_4 NPs using *Punica granatum* peel extract and studied the photocatalytic activity of the NPs. The synthesized NPs were spherical with the particle size of 43.78-73.10 nm. They evaluated the photocatalytic activity of the green synthesized Co_3O_4 NPs using Remazol Brilliant Orange 3R (RBO 3R) dye and results indicate Co_3O_4 NPs shows excellent photocatalytic activity (degraded up to 78.45%) [70].

Khalil and co-workers reported the biogenic synthesis of cubic Co_3O_4 NPs using *Sageretia thea* leaf extract and also studied the biocompatibility and biological applications of Co_3O_4 NPs. They showed that, the biogenic Co_3O_4 NPs have potent antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*. These synthesized Co_3O_4 NPs significant cytotoxic potential against axenic leishmanial promastigote and amastigote cultures. Herewith, they also reported the significant DPPH radical scavenging potential, moderate antioxidant capacity, reducing power and enzyme inhibition activities of bioinspired Co_3O_4 NPs. From the biological activity results, it can be suggested that Co_3O_4 NPs can be used for treatment of various diseases especially leishamniasis [41]. Golder and co-workers demonstrated fruit extract mediated green synthesis of Co_3O_4 NPs using *Sechium edule* were in the size of 39 nm and reported as an electrochemical sensor for H_2O_2 sensing [71]. Bilal and co-workers synthesized Co_3O_4 NPs using *Taraxacum officinal* leaves extract and studied the photocatalytic activity of synthesized NPs. The synthesized Co_3O_4 NPs is spherical shape with the size ranges from 50-100 nm. They evaluated the photocatalytic activity of biosynthesized Co_3O_4 NPs using methyl orange and direct yellow 142 dyes. The synthesized Co_3O_4 NPs catalyzed up to 96.24% and 93.37% degradation of methyl orange and direct yellow 142 dye, respectively, within 60 min. From the results, Co_3O_4 NPs could be a potential candidate to degrade toxic azo dyes [72].

6. Conclusion

This review has summarized the recent research studies in the field of biogenic synthesis of Co_3O_4 NPs by using some plant parts material. This literature surveys exhibited that major portion of work on biosynthesis of NPs of gold, silver, and zinc oxide NPs in comparison with the Co_3O_4 NPs. Hence, particularly special attention of scientific community is required to explore this simple, swift, robust, clean, eco-accommodating, and commercially affordable method for fabrication of Co_3O_4 NPs through this green chemistry bottom to top approach. Even though the green synthesis has many limitations like controlling the size and shape of these NPs and formed which is not a major issue for other conventional routes of synthesis, this one pot green synthesis still needs few developments which should

Table 3. Various applications of Co_3O_4 NPs synthesized using different plant extracts

Entry	Name of Plants	Applications	References
1	Azadirechta indica	Catalytic hydrogenation & antibacterial activity	[36]
2	Calotropis gigantea	Dye sensitized solar cell	[33]
3	Calotropis procera	Antibacterial activity	[58]
4	Euphorbia heterophylla	Photocatalytic activity	[34]
5	Gingko	Sensing (detection of glucose & H_2O_2)	[32]
6	Helianthus annus	Photocatalytic activity	[59]
7	Hibiscus Rosa-sinensis	Antibacterial & antifungal activities	[36]
8	Moringa oleifera	Supercapacitors	[61]
9	Piper nigrum	Photocatalytic activity	[62]
10	Punica granthum	Photocatalytic activity	[64]
11	Sageretia thea	Antibacterial, antileishmanial, antioxidant & enzyme inhibition applications	[35]
12	Sechinum edule	H_2O_2 sensing	[65]
13	Taraxacum officinal	Photocatalytic activity	[66]

be further looked into. Moreover, much more plant species should be exploited and evaluated in future era towards facile and rapid biogenic synthesis of metal oxide NPs. Further research needs to improve the applications, use of plant material for synthesis of NPs and highlighting the perfect mechanism behind the fabrication of Co_3O_4 NPs.

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Conflict of interest

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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