Biological Forum – An International Journal **15(3): 199-202(2023)**

ISSN No. (Print): 0975-1130
ISSN No. (Online): 2249-3239

Photocatalytic and *in vitro* Antioxidant Activity of *Terminalia chebula* Fruits Mediated ZnO Nanoparticles

***Rathika R.*[1*], *Mahmoodah Parveen K.*[2], *Suganthi, P.*[3], *Kannadasan N.*[4], *Srinivasan K.*[5]**

[1]*Assistant Professor, PG & Research Department of Zoology, Government Arts College, Affiliated to Periyar University, Dharmapuri, (Tamil Nadu), India.*

[2]*Assistant Professor, PG & Research, Department of Chemistry, Jamal Mohamed College (Autonomous), Affiliated to Bharathidasan University, Tiruchirappalli, (Tamil Nadu), India.*

[3]*Research Assistant, KIRND Institute of Research and Development Pvt. Ltd., Tiruchirappalli (Tamil Nadu), India.*

[4]*Assistant Professor, Department of Zoology, Karur Velalar College of Arts and Science for Women, Affiliated to Bharathidasan University, Karur, (Tamil Nadu), India.*

[5]*Guest Lecturer, PG & Research Department of Zoology, Government Arts College, Affiliated to Periyar University, Dharmapuri, (Tamil Nadu), India.*

(Corresponding author: Rathika R.)*

(Received: 10 January 2023; Revised: 12 February 2023; Accepted: 20 February 2023; Published: 22 March 2023)

(Published by Research Trend)

ABSTRACT: The synthesis of metallic nanoparticles using phytocompounds has drawn a lot of interest from scientists and the pharmaceutical sector. The phytocompounds from *Terminalia chebula* are the most promising in Southeast Asian nations for treating a wide range of human illnesses. Zinc oxide (ZnO) nanoparticles made from *T. chebula* fruit are created through a downstream process. The deterioration of the dye compounds was caused by green ZnO, which increased the surface area on the electron hole. The green synthesised ZnO NPs *in vitro* antioxidant capabilities demonstrated a substantial inhibitory activity of the tested agents against free radicals. According to our findings, ZnO nanoparticles produced by *T. chebula* fruits exhibited enhanced scavenging and reducing activities against reactive oxygen radicals as well as efficient photodegradation of methylene blue dye.

Keywords: *T. chebula*, ZnO, Photocatalytic Degradation, Antioxidant

INTRODUCTION

Nanotechnology is considered as the newly emerging field which produces different nanoproducts such as nanoparticles, nanopowders, nanotubes, nanowires and nanocolloids (Suganthi *et al.*, 2019). Nanoparticles (NPs) are the smallest particles (10-9m) with wide applications and unique properties such as electronical agent, catalytic factor, pharmaceutical, chemical, environmental, agricultural and remediation industries (Athif *et al.*, 2020). Metallic nanoparticles received a great attention among the researchers due to the alternative utilization of bulk (whole) materials, increased surface area, anionic & cationic properties, binding efficiency and compound affinity. Utilization of metallic nanoparticles in the consumer products and drug are double during the last decade (Suganthi *et al.*, 2015). Excessive utilization of metallic nanoparticles leads to various pollution and health hazards (Suganthi *et al.*, 2016).

Green synthesis representing the utilization of either microbes, fungal and plant compounds for the nanoparticles synthesis (Rai *et al.*, 2011; Zahir *et al.*, 2014). Particularly during metallic nanoparticles synthesis, the researchers giving preference to the phytocompounds. Phytocompounds interact with the metal ions during synthesis which resulted in the stable configuring compound synthesis by downstream and condensation process (Amarnath *et al.*, 2013). Moreover, phytocompounds are less toxic to the non-targeted species. Finally, the green synthesised compound are considered as the cost effective, eco-friendly and easy to access (Dhandapani *et al.*, 2014). The environmentally harmful waste byproducts are not produced during the environmentally hazardous green synthesis of the nanoparticles, which lowers the possibility of contamination (Varada and Lalit 2022).

Zinc oxide (ZnO) nanoparticles are the most promising metallic nanoparticles with enhanced antioxidant, antimicrobial and photocatalytic degradation abilities (Maham *et al.*, 2017). ZnO NPs are widely used in clinical products such as sprays, wound dressings, ointments, drug carrier. The plant *Terminalia chebula* belongs to Combretaceae family used for the treatment of various human ailments (cardiac and hepatic disorders, Fever, TB, indigestion) in different nations such as Nepal, India, Sri Lanka, China, Vietnam and

Malaysia (Ravi Shankara *et al.*, 2019; Singh and Kumar 2013). There are several medical benefits of plant material, including antibacterial, antitussive, antispasmodic, antiasthmatic, and antihelminthic qualities (Gilani *et al.*, 2008a & 2005a). Our study was aimed to analyze the *Terminalia chebula* mediated ZnO nanoparticles (GZnO) photocatalytic (dye degradation) and in vitro antioxidant activities.

MATERIAL AND METHODS

Terminalia chebula fruits purchased from the local Ayurveda shops and confirmed their species with standard taxonomical aids. Dried *T. chebula* fruits are ground well and sieved. Zinc oxide (CAS 1314-13-2, mol. wt. 81.39) are purchased from Sigma-Aldrich. By downstream process, the *T. chebula* mediated ZnO nanoparticles are synthesized (Results not included) and used for this study.

A. Antioxidant studies: DPPH, FRAP assay

Scavenging and reducing activity of Green synthesised ZnO NPs are analysed by in vitro DPPH and FRAP assay (Jeeva *et al.*, 2011). 1mgL^{-1} of green synthesised ZnO NPs are added to the DPPH (1ml) solution and kept the mixture in dark (30min) for incubation. Finally, the absorbance of the solution is measured at 520nm and the values are calculated by using the following formula. The ferric acid reducing ability of the nanoparticles are analysed by FRAP assay. 0.2ml of green synthesized ZnO NPs are added with the TPTZ solution (3.8ml) and the solution are incubated for 30 mins at room temperature. The absorbance of the solution is measured at 570nm and the values are calculated by using the following formula.

$$\text{Inhibiton}(\%) = \frac{(\text{Abs. of control} - \text{Abs. of sample})}{\text{Abs. of control}} \times 100$$

B. Photocatalytic activities

T. chebula mediated ZnO NPs photocatalytic activities on the basic dye methylene blue (MB) are studied by standard irradiation method. 100mg of green synthesized ZnO NPs mixed with 6mgL^{-1} of MB dye and kept the solution in the magnetic stirrer for 60min in dark environment for the adsorption attainment. The solution is exposed to sunlight with 80-130Klux (120min) for the photocatalyst mechanism. During irradiation, 2ml of mixture collected at for every 15min and centrifuged. By using UV visible spectrophotometer, the dye degradation nature of the solution measured between 600-700nm. The degradation efficiency rate (R) of the MB dye by green synthesised ZnO is calculated by using the following formula. C_0 is the initial concentration absorbance and C_1 is the final concentration absorbance.

$$\text{Degradation efficiency rate (R)} = \frac{C_0 - C_1}{C_0} \times 100\%$$

RESULTS AND DISCUSSION

The antioxidant activities of the green synthesized ZnO nanoparticles results showed the increased concentration (50, 100, 200 and 400µg/ml) of the nanoparticles showed increased scavenging activities against the oxygen radicals efficiency (Table 1). The oxidation mechanism of the nanoparticles are significantly enhanced due to the first order reaction kinetics (IC50=280µg/ml) between the tested methylene blue dye and the green synthesized ZnO nanoparticles. The active phyto metabolites present in the GZnO reducing the ROS which resulted in high antioxidant activities in treated groups.

Table 1: DPPH and FRAP activity of green synthesized ZnO nanoparticles.

Sr. No.	Conc.	DPPH scavenging activity (%)	FRAP reducing activity (%)
1.	50µg/ml	8.99	2.53
2.	100µg/ml	15.78	3.57
3.	200µg/ml	32.16	9.78
4.	400µg/ml	67.53	21.57

The metal oxide nanoparticles showed an effective photocatalytic activity against different type of dyes (Thakur *et al.*, 2019). It has the remarkable unique kinetics on scavenging and reducing mechanism against the reactive oxygen species whereas *T. chebula* (Retz.) plant extract total antioxidant IC50 values observed at 14µgmL^{-1} (Saha *et al.,* 2016). Similar results are reported by Pfundstein *et al.,* (2010); Bajpai *et al.,* (2005); Zhu *et al.* (2019).

The methylene blue (MB) dye degradation efficiency of the *T. chebula* mediated ZnO NPs are observed at every 15min for 2 hrs. From the solution mixture kept in dark, the samples are collected and absorbance are measured. Fig. 1 showed the increased percentage of the degradation efficiency of the *T. chebula* mediated ZnO NPs concentrations (0, 50, 100, 200 and 400mg) are observed. Doubling of irradiation time increased eventually leads to the increased MB degradation rate.

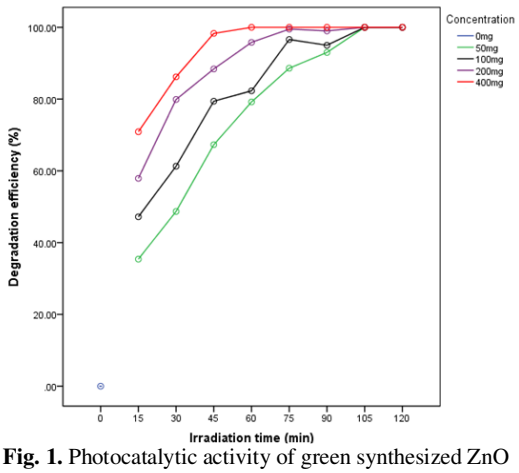

Fig. 1. Photocatalytic activity of green synthesized ZnO nanoparticles.

The phytocompounds present in the ZnO NPs enhanced the substantial surface area eventually developing many active sites for the catalytic and oxidative activity of the nanoparticles. During recombination of ZnO NPs with MB dye, the irradiation enhances the catalytic activity by GZnO protonation. This counterbalance activity increased redox potential of the intermediate compounds. Finally, the photodegradation activity of

the GZnO obeys the pseudo first order kinetics reaction which resulted photoexcitation of the trapped electron. The radical and hydroxyl groups developed the electron hole which degraded the MB dye and finally dye are degraded. The following reaction explained the photocatalytic activity of the green synthesised ZnO nanoparticles.

$$\begin{aligned} \mathrm{GZnO - MB} \xrightarrow{\mathrm{hv}} & \mathrm{GZnO(h^+) - MB(e^-)\, MB(e^-) + O_2} \\ & \rightarrow \mathrm{MBO + O_2^- GZnO(h^+)} \\ & + \mathrm{H_2O/OH^-} \\ & \rightarrow \mathrm{GZnO + OH^- OH^- + MB} \\ & \rightarrow \text{Dye degradation products} \end{aligned}$$

Moon *et al.* (2018) explained the dye degradation process by biosynthesised nanoparticles as various small byproducts (sulphates, carbon dioxide and water molecules) were released from degradation of dye molecules. Mahmoud and Mohammad (2015) evidenced the photocatalytic activity of the green synthesised metallic nanoparticles.

In photocatalytic studies, the peaks observed between the 350-400nm represented the presence of nanoparticles in the dye degradation activity (Gola *et al.,* 2021; Shan *et al.,* 2021) reported the photocatalytic degradation efficiency of M. fragrans mediated ZnO NPs, an effective photocatalytic agents led to the 88% breakdown of methylene blue dye in about 140 minutes. Albeladi *et al.* (2020); Muraro *et al.* (2020); Chandhru *et al.* (2020) reported the photocatalytic activity of the green synthesized oxide nanoparticles against the various toxic dyes such as methyl orange (MO), Rhodamine B dye, Auramine O, Congo red, Tymol blue and Phloxine B.

CONCLUSIONS

Nowadays, Green synthesis of metallic nanoparticles considered as the high efficient, cost effective and eco-friendly products and also a sustainable product for chemical based nanoparticles. *Terminalia chebula* mediated titanium dioxide (GZnO) nanoparticles DPPH and FRAP antioxidant studies revealed the enhanced oxygen radical scavenging activities. GZnO nanoparticles exhibited the dye degradation (photocatalytic) activities against methylene blue dye.

FUTURE SCOPE

The advantages of green synthesis ZnO NPs over chemical and physical approaches are cost-effectiveness, environmental friendliness, and efficiency in scaling up for large-scale synthesis.

Acknowledgement. The authors are thanking to the PG & Research Department of Zoology and Botany, Government Arts College for their support during this study.

Conflict of interest. None.

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How to cite this article: Rathika R., Mahmoodah Parveen K., Suganthi P., Kannadasan N. and Srinivasan K. (2023). Photocatalytic and *in vitro* Antioxidant Activity of *Terminalia chebula* Fruits Mediated ZnO Nanoparticles. *Biological Forum – An International Journal, 15*(3): 199-202.