Crossian Resonance

A Multidisciplinary Research Journal **ISSN 0976-5417**
(A refereed Biannual Published in June and December)
Vol. 15, No.2, December 2024
https://research.holycrossngl.edu.in/

Evaluation of Antioxidant Potentials of Benzimidazoles Synthesized from *Ortho*-Phenylenediamine and Benzaldehyde Derivatives *Via Phyllanthus emblica* Fruit Extract

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ABSTRACT

The present investigation concentrates the antioxidant potentials of two benzimidazole derivatives synthesised from o-phenylenediamine, 3-nitrobenzaldehyde and 3-fluorobenzaldehyde using Phyllanthus emblica fruit extract. Benzimidazoles synthesised from Phyllanthus emblica extract are confirmed from the spectral studies. Antioxidant property of benzimidazole derivatives is identified from DPPH assay process. The IC_{50} of control ascorbic acid and benzimidazoles synthesised from o-phenylenediamine and 3-nitrobenzaldehyde and ortho-phenylenediamine and 3-fluorobenzaldehyde are 97.12, 80.23 and 92.23 µg/mL. The results suggested that the two synthesised benzimidazole derivatives show very good antioxidant activities. Thus, the green synthesised benzimidazole derivatives make a path for future research in clinical diagnosis as antioxidants and therapeutic agents.
***Keywords**: Antioxidant activity, Benzimidazole. Green synthesis, Phyllanthus emblica extract*

1. Introduction

Chemical reactions carried out without solvents enhance efficiency, selectivity, manipulation ease, and often avoid toxic and volatile solvents. For the increasing economic and environmental concerns in recent years, it is now essential for chemists to search for as many environmentally benign methods as possible. The edible plants, fruits, roots and leaf extracts can be used in biocatalytic transformation for organic reactions [1,2]. Benzimidazole is a heterocyclic aromatic organic compound, beneficial pharmacophore and a prerogative structure in medicinal chemistry. Benzimidazole and its derivatives act as therapeutic agents and its shows various biological activities *viz* antiulcer, antihypertensive, analgesic, anti-inflammatory, anti-viral, antifungal, anticancer, and antihistaminic agents [3-5].

Nowadays, fruit and plant extracts have been recognised as viable organic solvent for synthesizing compounds of pharmaceutical interest [6]. It is used as solvent and catalyst in numerous reactions [7]. The broad application of various fruit extracts is attributed to its harmless, cost-effective, and environmentally friendly properties. Additionally, bioactive

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compounds isolated from the waste of fruits and vegetables play a substantial role in organic synthesis [8].

According to the review of literature, the present study concentrates on the natural acid catalysed solvent free synthesis of benzimidazole derivatives from *o*-phenylenediamine, 3-nitrobenzaldehyde and 3-fluorobenzaldehyde using *Phyllanthus emblica* fruit extract. *Phyllanthus emblica* often known as Indian gooseberry has become extremely important in indigenous traditional medical systems. The fruit of *Phyllanthus emblica* contains various phytochemicals, fatty acids, glycosides and phosphatides. This extract is used as the reagent for benzimidazole derivatives synthesis. The synthesised benzimidazole derivatives are confirmed by spectral analysis (absorption and FT-IR). The benzimidazole derivatives show good antioxidant potentials.

2. Materials and Methods

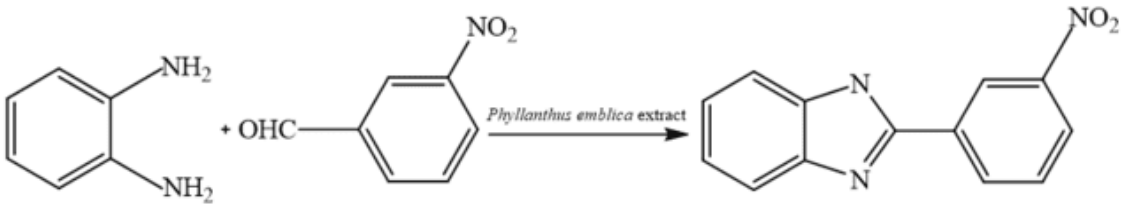
Fresh and ripened *Phyllanthus emblica* fruit was purchased from the market. The chemicals *o*-phenylenediamine, 3-nitrobenzaldehyde and 3-fluorobenzaldehyde were procured from Merck.

2.1. Preparation of *Phyllanthus emblica* extract

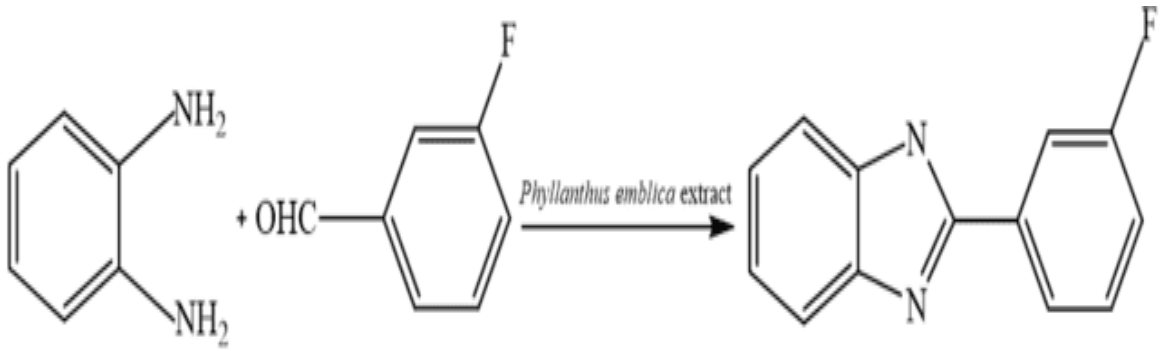
About 50 g of ripened *Phyllanthus emblica* fruit was washed thoroughly by water and cut into small pieces. The pieces were grinded by a mixer grinder and filtered *via* Whatman filter paper. The collected filtrate was centrifuged around 15 min. The collected centrifugate was then used for synthesis of benzimidazole derivatives.

2.2 Synthesis of Benzimidazole Derivatives from *Phyllanthus emblica* extract

Equal quantities of *o*-phenylenediamine, 3-nitrobenzaldehyde and 5 mL of the extract were placed in a mortar and grinded well for 1 h using a pestle. The corresponding benzimidazole product was separated out as a pale orange paste (**Scheme 1**). The product was washed by ice water and purified by recrystallization with a minimum amount of ethanol. Similarly, benzimidazole from *o*-phenylenediamine and 3-fluorobenzaldehyde was also synthesised (**Scheme 2**).



Scheme 1. Synthesis of benzimidazole from o-phenylenediamine and 3-nitrobenzaldehyde



Scheme 2. Synthesis of benzimidazole from o-phenylenediamine and 3-fluorobenzaldehyde

2.3 Characterization Methods

The UV-Visible spectra of benzimidazoles were taken from the Shimadzu UV-1800 spectrophotometer. FT-IR spectrum of the samples was analysed by Shimadzu IR Affinity spectrophotometer using KBr pellet method.

2.4 Antioxidant Activity

Antioxidant efficiency of benzimidazole derivatives was assessed by the DPPH assay method. Antioxidant properties of benzimidazole derivatives at different concentrations (10 - 500 µg/mL) were determined and the results are compared with the control ascorbic acid.

% of Inhibition = [(Abs of control - Abs of test)/Abs of control] x 100

A plot of % inhibition *vs* concentration was drawn and the IC_{50} value was calculated from the plot.

3. Results and Discussion

The role of *Phyllanthus emblica* extract for the synthesis of benzimidazoles from *o*-phenylenediamine and 3-nitrobenzaldehyde and *o*-phenylenediamine and 3-fluorobenzaldehyde is analysed. The benzimidazole derivatives are characterized by absorption and IR spectral methods. *Phyllanthus emblica* extract contains citric acid and ascorbic acid, the acids present in the extract catalysed the preparation of benzimidazole derivatives.

3.1 Absorption Spectral Studies

The synthesized benzimidazoles from *Phyllanthus emblica* extract is first analyzed by absorption spectral studies. The UV-Visible spectrum of benzimidazoles is carried out in ethanol. The benzimidazole synthesised from *o*-phenylenediamine and 3-nitrobenzaldehyde displays bands at 255.5, 262, 268.5 and 285 nm **(Figure 1)** whereas the benzimidazole synthesised from *o*-phenylenediamine and 3-fluorobenzaldehyde displays bands at 262, 264.5, 268 and 305 nm due to π-π* and n-π* transitions **(Figure 2)**.

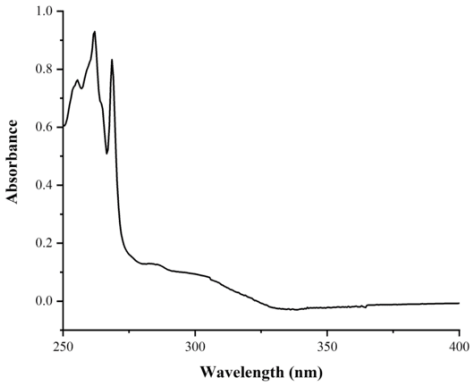


Figure 1. UV spectrum of benzimidazole from *o*-phenylenediamine and 3-nitrobenzaldehyde

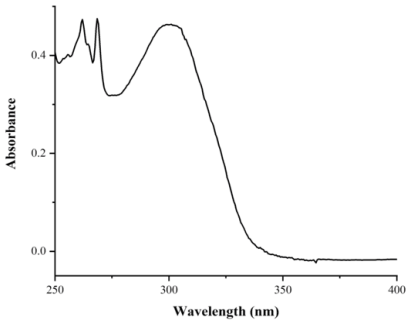


Figure 2. UV spectrum of benzimidazole from *o*-phenylenediamine and 3-fluorobenzaldehyde

3.2 FT-IR Spectral Studies

The FT-IR spectrum of benzimidazole derivative synthesised from *o*-phenylenediamine and 3-nitrobenzaldehyde using *Phyllanthus emblica* extract displays bands in the region 3402, 2980, 2899, 2378, 1924, 1651, 1396, 1328, 1263, 1051, 881, 659 cm^{-1} respectively **(Figure 3).** The broad band occurs at 3402 cm^{-1} indicates N-H stretching of secondary amine. The band at 2980 cm^{-1} indicates aromatic C-H stretching. The band forms at 1651 cm^{-1} demonstrates C=N stretching of benzimidazole ring. The band at 1328 cm^{-1} represents C-N stretching of the benzimidazole ring. The band at 1396 cm^{-1} indicates NO_2 stretching of an asymmetric ring. The band at 1263 cm^{-1} represents NO_2 stretching of the symmetric ring. The band at 659 cm^{-1} indicates N-H stretching. The formation of benzimidazole derivatives from *o*-phenylenediamine and 3-nitrobenzaldehyde is thus confirmed from the FT-IR spectral information.

FT-IR spectrum of benzimidazole derivative synthesised from *o*-phenylenediamine and 3-fluorobenzaldehyde using *Phyllanthus emblica* extract displays bands at 3466, 3194, 2526, 2382, 1687, 1627, 1589, 1517, 1346, 1220, 1132, 1082, 974, 910, 738, 696, 553 cm^{-1} respectively **(Figure 4).** The broad band occurring at 3466 cm^{-1} represents N-H stretching of the imidazole ring (N-H). The band formed at 3194 cm^{-1} indicates C-H

stretching of the aromatic ring. The bands arise at 1687 and 1627 cm^{-1} demonstrates C=N stretching of benzimidazole ring. The bands at 1589 and 1517 cm^{-1} indicates aromatic C=C stretching. The bands occur at 1346 and 1220 cm^{-1} represent aromatic C-N stretching of the ring. The band at 1132 cm^{-1} indicates C-F stretching. The band at 1080 cm^{-1} represents aromatic C-H in-plane bending. The bands occur at 974 and 910 cm^{-1} indicate aromatic C-H out-plane bending. The bands at 738 and 696 cm^{-1} represent aromatic ring vibration. The band occurs at 553 cm^{-1} indicates C-N bending. The formation of benzimidazole derivatives from *o*-phenylenediamine and 3-fluorobenzaldehyde is thus confirmed from the FT-IR spectral information.

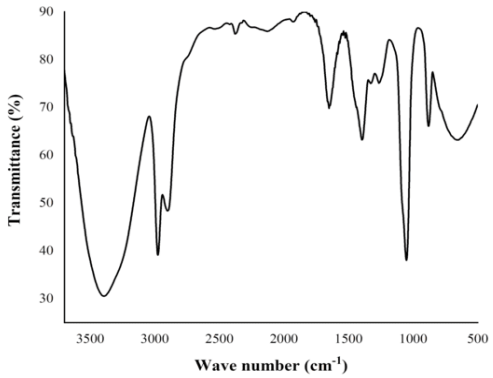


Figure 3. FT-IR spectrum of benzimidazole from *o*-phenylenediamine and 3-nitrobenzaldehyde

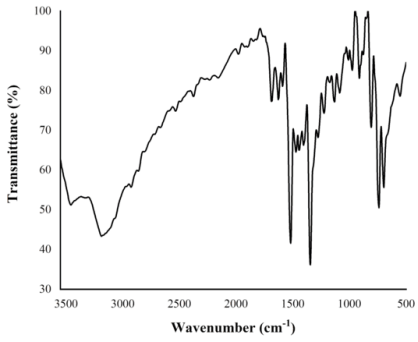


Figure 4. FT-IR spectrum of benzimidazole from *o*-phenylenediamine and 3-fluorobenzaldehyde

3.3 Antioxidant Potential of Benzimidazoles

Antioxidant potentials of benzimidazoles at various concentrations are presented in **Tables 1** and **2**. The synthesized benzimidazoles derivatives shows good antioxidant activity compared to that of ascorbic acid. The IC_{50} of ascorbic acid and benzimidazoles synthesised from *o*-phenylenediamine and 3-nitrobenzaldehyde and *o*-phenylenediamine and 3-

fluorobenzaldehyde are 97.12, 80.23 and 92.23 µg/mL. The antioxidant potential of benzimidazole derivatives mainly depends on the substituents present in the compounds. Thus, the present study confirms the antioxidant efficacy of two benzimidazole derivatives and these compounds can be used as good free radical scavengers.

Table 1. Percentage inhibition of benzimidazole from *o*-phenylenediamine and 3-nitrobenzaldehyde

Concentration (µg/mL)	**Percentage of Inhibition (%)**
Ascorbic acid	97.12
500	67.9489
250	60.7174
100	50.4621
50	47.5975
10	32.5338
IC_{50}	**80.23 µg/mL**

Table 2. Percentage inhibition of benzimidazole from *o*-phenylenediamine and 3-fluorobenzaldehyde

Concentration (µg/mL)	**Percentage of Inhibition (%)**
Ascorbic acid	97.12
500	72.9722
250	68.3493
100	59.1348
50	51.178
10	43.5401
IC_{50}	**92.23 µg/mL**

4. Conclusion

The antioxidant activities of two benzimidazole derivatives synthesised from *o*-phenylenediamine, 3-nitrobenzaldehyde and 3-fluorobenzaldehyde using the extract of *Phyllanthus emblica* has been investigated by DPPH assay method. The structure of the synthesised benzimidazole derivatives is confirmed from UV-Visible and FT-IR spectral techniques. The synthesised benzimidazoles show very good antioxidant activities when compared to ascorbic acid. This solvent-free method is eco-friendly and avoids the usage of harmful materials, making it a greener method for synthesizing benzimidazoles. The

antioxidant properties of benzimidazole derivatives are expected to stimulate further interest in biomedical applications.

References

1. Patiño-Ruiz D.A., Meramo-Hurtado S.I., González-Delgado A.D., Herrera, A. Environmental sustainability evaluation of iron oxide nanoparticles synthesized via green synthesis and the coprecipitation method: A comparative life cycle assessment study. ACS Omega. 2021; 6: 12410 - 12423.
2. Redha A.A. Review on extraction of phenolic compounds from natural sources using green deep eutectic solvents. Journal of Agricultural and Food Chemistry.2021; 69: 878 - 912.
3. Prakash D.P., Moon S.A. Review on synthesis of benzimidazole from o-phenylenediamine. International Journal of Advance Research and Innovative Ideas in Education. 2024; 10: 2395 - 4396.
4. Panda J., Padhy G.K., Patro C.S., Raul S.K., Behera A.K. Synthetic approaches of benzimidazole derivatives: A review. Rasayan Journal of Chemistry. 2023; 16: 2009 - 2018.
5. Ebenezer O., Oyetunde-Joshua F., Omotoso O.D., Shapi M. Benzimidazole and its derivatives: Recent advances. Results in Chemistry. 2023; 5: 2020 - 2022.
6. Patil S., Jadhav S.D., Mane S.Y. Pineapple juice as a natural catalyst: An excellent catalyst for Biginelli reaction. Int. J. Org. Chem. 2011; 1: 125 - 131.
7. Pramanik T., Pathan A.H. Exploring the utility of fruit juices as a green medium for Biginelli reaction. Res. J. Pharm. Bio. Chem. Sci. 2014; 5: 444 - 449.
8. Sagar N.A., Pareek S., Sharma S., Yahia E.M., Lobo M.G. Fruit and vegetable waste bioactive compounds, their extraction, and possible utilization. Compr. Rev Sci. Food Saf. 2018; 17: 512 - 531.