

Sustainable synthesis of zinc oxide nanoparticles from spent coffee grounds as a reusable catalyst for Biginelli reaction

M. U. Ismail¹ and H. I. C. De Silva^{2*}

¹College of Chemical Sciences, Institute of Chemistry Ceylon, Rajagiriya 10107, Sri Lanka

²Department of Chemistry, University of Colombo, Colombo 3, Sri Lanka.

* hicdesilva@chem.cmb.ac.lk

Metal oxide nanoparticles (NPs) have attracted significant attention in organic catalysis for the development of recoverable heterogeneous systems. Plant-derived extracts are explored as sustainable alternatives to conventional synthetic capping and stabilizing agents for NP synthesis. Multicomponent reactions (MCRs), particularly the Biginelli reaction, provide an atom-economical route to synthesize 3,4-dihydropyrimidinone (DHPM) derivatives. In this study, zinc oxide (ZnO) NPs were synthesized via co-precipitation using spent coffee ground extract. The optical, structural, and morphological properties of ZnO NPs were characterized using ultraviolet-visible (UV-Vis) spectroscopy, Fourier Transform Infrared (FT-IR) spectroscopy, Powder X-ray diffraction (PXRD), and scanning electron microscopy with elemental dispersion (SEM-EDX) analysis. The catalytic activity and recyclability of ZnO NPs were evaluated for synthesizing DHPM derivatives via the Biginelli reaction, using benzaldehyde and its substituted derivatives. Product structures were confirmed by FT-IR, ¹H-NMR and ¹³C-NMR. UV-Vis analysis revealed a characteristic absorption peak at 372 nm, while the

FT-IR band at 505 cm⁻¹ confirmed ZnO formation. The average crystallite size was calculated as 23.96 nm using Debye-Scherrer's equation, while SEM analysis revealed predominantly spherical particles with an average size of 56.6 nm. The results demonstrated moderate efficiency across substrates. The 4-nitro derivative gave the highest yield (81%), followed by benzaldehyde (69%) and 4-hydroxybenzaldehyde (63%). The 4-methoxy and 4-chloro derivatives yielded 58%. The recyclability of the ZnO nanoparticle catalyst was evaluated over four consecutive cycles, and the catalyst turnover number (TON) was calculated to be 13.0. These results indicate that ZnO NPs possess good catalytic activity, stability, and reusability for the synthesis of Biginelli derivatives.

Keywords:

atom-economical ; 3,4-dihydropyrimidinones ; heterogeneous ; multicomponent.