

ABSTRACT

In present scenario, greener and eco-friendly protocol (i.e. after the term ‘green chemistry’ was first coined by Paul Anastus) is getting special attention in the scientific community due to their broad spectrum of advantages over conventional synthetic tools like using non-hazardous chemicals, easy separation techniques, offering high to excellent yield and many more. A diversified approach towards the synthesis of multiple value-added heterocycles opens up a new horizon towards organic catalysis and for this purpose, development of natural resources as effective catalyst is what green chemistry aimed to do. Using environmentally benign reaction medium, we worked diligently on synthesis of some crucial heterocyclic scaffolds such as pyrans, tetraketones, biaryl skeleton, *N*-heterocycles (i.e. disubstituted benzimidazoles, pyrazoles, aryl benzazoles and dihydroquinazolinones (DHQs)) along with substituted β -amino alcohols starting from plant poly phenols.

It all initiated with designing a facile approach towards the synthesis of β -amino alcohols those which have wide applications in pharmaceutical field. They are also used as chiral auxiliary in asymmetric synthesis. Synthesizing the same starting from substituted trans-cinnamic acids using an unconventional greener medium coconut juice abbreviated as ACC (água-de-coco do Ceará), has established itself as a novel one due to non hazardous, renewable feedstock, and energy efficient experimental method. It was followed by density functional theory (DFT) based computational study to investigate the mechanistic path for generating desired product in last step i.e. the main key step for the synthesis of β -amino alcohols.

ACC was further explored as reusable catalyst for synthesis of diols and 4*H*-pyrans offering both ease of operation and impressive yield. Thereafter, tetraketone and substituted tetrahydrobenzo[*b*]pyran systems of high economic and industrial importance compounds were synthesized under in ethanol using water extract of tamarind seed ash (WETSA) as catalyst, involving one step multicomponent coupling reactions (MCRs). Desired products were separated using simple precipitation technique with excellent yield and less reaction time. Recyclability of the catalyst upto four cycles channelized the objectives of green chemistry successfully.

Then, we developed *ex-situ* multi-walled carbon nanotubes-zirconia (MWCNTs-ZrO₂) nanocomposite from natural feedstock ACC, which acted as a green, environmentally friendly, and nontoxic solvent. Moreover, we utilized the aforementioned nanocomposite for synthetic chemistry for the first time. Multiple heterocyclic scaffolds like as pyrazoles, disubstituted benzimidazoles, 2-arylbenzazoles, and DHQs were synthesized using the

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MWCNTs-ZrO₂ as sustainable and heterogeneous catalyst under green reaction medium. The catalyst was recovered easily by filtration and offered reusability upto four cycles without any significant loss in catalytic activity.

Biaryl skeletons are crucial moiety present in drug molecule with anti-inflammatory, anti-microbial property as well as in chiral ligands and electron conducting materials. Suzuki-Miyaura cross coupling reaction is known for synthesizing biaryl scaffolds in wide range. This time, we focused on generation of *in-situ* Pd nanoparticles for the formation of bi-aryl building block starting from aryl boronic acids and aryl halides, where water extract of banana stem ash (WEBSA) was explored as co-solvent and base. The use of WEBSA bestowed this protocol with few added advantage as short reaction time, eco-friendly medium, recovery of catalyst with further use and less rigorous reaction set up.

In conclusion, this work sheds light on construction of green and eco-friendly protocol for diversity oriented synthesis of biologically and economically value added heterocyclic scaffolds. Incorporating the aspect of sustainable and heterogeneous catalysis with broad substrate scope and ease of operation also enhances it's practical acceptability to the scientific community in future.

Keywords: coconut juice (ACC); β -amino alcohols; multicomponent coupling reactions (MCRs); diols; 4*H*-pyrans; water extract of tamarind seed ash (WETSA); tetraketones; tetrahydrobenzo[*b*]pyrans; multi-walled carbon nanotubes-zirconia nanocomposite (MWCNTs-ZrO₂); disubstituted benzimidazoles; pyrazoles; 2-aryl benzazoles; dihydroquinazolinones (DHQs); water extract of banana stem ash (WEBSA); biaryl skeletons; Pd nanoparticles.