

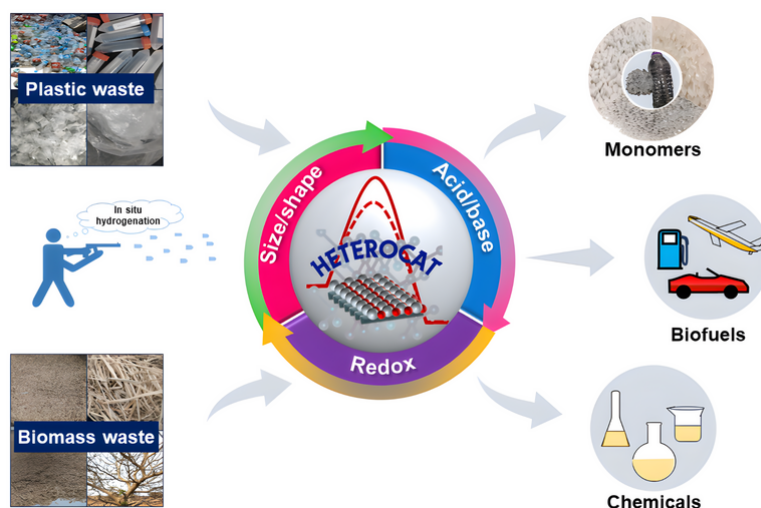
# Advanced heterogeneous catalysis for plastic/biomass waste valorization



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Both plastic and biomass are complex polymers, consisting of strong carbon-carbon and carbon-heteroatom linkages, which make them highly recalcitrant to degradation/recycling at ambient conditions. Heterogeneous catalysis, a key pillar for the chemical industry, can provide alternative routes to activate plastic/biomass structures towards desirable products with low energy consumption and negligible waste generation (Figure 1). Metal oxides and supported metals are widely used heterogeneous solid catalysts in the chemical industry. The unique characteristics of these catalysts are tunable acid-redox properties, high thermal/chemical stability, and facile recovery/reusability, enabling them as appealing catalysts for plastic/biomass waste valorization. The particle size/morphology of these catalysts play a vital role in optimizing catalytic active sites (acid-redox) to achieve higher reaction rates, selectivity, and yield in any chemical reaction. The fine-tuning of the particle size/morphology in the nanoscale range (1-100 nm in one dimension at least) offers remarkable properties, including high specific surface area, abundant acid-redox sites, and highly enriched surfaces (corners and edges). Hence, the application of nanostructured metal oxides and supported metal nanoparticles in plastic/biomass waste valorization has been a focal research topic in recent years.

The activity, selectivity, and stability of a catalyst are the key driving forces for the sustainable development of a catalytic process in the chemical industry. Among them, the catalyst's selectivity is considered the most important factor to make any catalytic process/technology economically viable. The high selectivity of a catalyst towards a desirable product reduces tedious workup procedures as well as the use of solvents to recover/purify the product, thus it eventually minimizes the waste generation and process cost. In the case of plastic/biomass valorization, achieving high selectivity towards a particular product is challenging because both plastic and biomass are rigid, complex polymeric molecules, and their cleavage can lead to the formation of different products.



*Figure-1: Heterogeneous Catalysis for Plastic/Biomass Waste Valorization*

The heterogeneous catalysts, including nanostructured metal oxides and supported metal nanoparticles, typically contain various active sites with non-uniform dispersion and inhomogeneous local geometries. This will not only lead to uncontrolled cleavage of carbon-carbon or carbon-heteroatom (oxygen, nitrogen, etc.) in plastic/biomass but also to non-specific activation of the obtained intermediates, resulting in lower yields of the desirable products. It will have serious implications on the practical feasibility of the catalytic process for plastic/biomass waste valorization. Hence, significant efforts should be made towards improving the existing catalyst development methods and/or designing novel synthesis methods for advanced heterogeneous nanocatalysts with structural homogeneity and optimum specific active sites (acid, redox, or both). A promising approach is to use porous carbon as a support for stabilizing metal nanoparticles, which can lead to improved dispersion and uniform coordination structure of metal active sites as well as enhanced diffusion properties to provide higher reaction rates and product selectivity. In this regard, a great opportunity lies in using plastic/biomass waste as a precursor for preparing porous carbon materials and using them to develop multifunctional metal-based nanocatalysts for the conversion of plastic/biomass waste into monomers, fuels, or chemicals.

Hydroprocessing of plastic/biomass waste in the presence of hydrogen is an efficient way of producing fuel-grade hydrocarbons. However, using hydrogen gas requires extreme reaction pressure and temperature conditions, which in turn not only increases the process cost drastically but also poses safety ambiguities. Hence, the sustainable strategy is to use safe liquid hydrogen carriers, such as methanol, ethanol, or butanol by means of in-situ hydrogen generation and utilization for plastic/biomass waste valorization. Concurrently, facile workup procedures should be developed for separating the byproducts generated from the hydrogen carriers to make the process practically feasible.

#### SP HeteroCat Lab:

The “SP HeteroCat Lab” at the Department of Chemistry, IITH, aims to design and develop sustainable heterogeneous catalytic methods for plastic/biomass waste conversion and diverse nitrogenous chemical synthesis. Our current work is focused on the catalytic recycling/valorization of two types of plastic waste: PET to value-added monomers and polyolefin plastic to fuels (petrol, diesel, or jet fuel) and porous carbon materials. The conversion of lignin, a waste product from 2G ethanol and pulp/paper industries, to fuels, chemicals, and functional carbon materials is another key focus of our research. The third research topic is to develop facile catalytic C-N coupling strategies for biomass-based N-heterocycles (drug motifs) using safe hydrogen carriers. The key to efficient and selective plastic/biomass waste valorization is to develop new heterogeneous catalytic materials with structural uniformity and the optimal amount of specific active sites. Thus, we strive for a deeper understanding of the catalysts at the nanoscale range that can provide us with valuable insights for the rational design of novel bifunctional nanostructured metal-based catalysts with optimum catalytic active sites (mainly acid-redox properties) for plastic/biomass waste conversion and diverse nitrogenous chemicals.

The success of any research group primarily depends on the research scholars. The SP HeteroCat group is fortunate to have enthusiastic, dedicated, and motivated research scholars, and their commitment and perseverance to work on challenging problems stimulate the group research endeavours at IITH. We aim to continue our commitment to excellence, innovation, and the pursuit of knowledge in the field of Heterogeneous Catalysis, with the ultimate goal of developing industrially relevant processes for plastic/biomass waste conversion towards a sustainable society.



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## Catalytic dye degradation by hydrogel-silver nanoparticle nanohybrids

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Water pollution has emerged as a critical global issue, driven by the industrialization of sectors like paints, food, leather, printing, and textiles that extensively use dyes[1]. These industries generate over  $7 \times 10^5$  tons and nearly 10,000 varieties of dyes and pigments, with more than 10,000 tons consumed annually[1]. Approximately 10–15% of these dyes are discharged untreated into water bodies, posing a significant threat[2,3]. The release of vast amounts of untreated dyes into the aquatic environment is a considerable concern, given their high toxicity and carcinogenicity to microbial populations and mammals[4,5]. It is imperative to treat wastewater effectively to eliminate these hazardous dyes before entering aquatic ecosystems.

Several approaches have been utilized for dye degradation, encompassing chemical[6], biological[7], photocatalysis[8], and catalytic reduction using metal nanoparticles (NPs)[9]. However, using bare metal NPs has some drawbacks compared to stabilized counterparts. The elevated surface energy of naked metal NPs prompts agglomeration, resulting in a notable decline in catalytic activity. Furthermore, these nanoparticles are not easily recyclable through a straightforward centrifugation process. Both challenges can be mitigated by immobilizing metal NPs onto a solid support. Different supporting materials, including dendrimers[10], polymeric microgels[11,12], bulk hydrogels[13], and inorganic substances like reduced graphene oxide[14], are employed for this purpose.

