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Catalytic Decomposition of Formic Acid into H_2/CO_2 Gas: A Distinct Approach

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Abstract : Finding a sustainable alternative energy to fossil fuel is an urgent need as various environmental challenges in the world arise. Therefore, formic acid (FA) decomposition has been an attractive field that lies at the center of the biomass platform, comprising a potential pool of hydrogen energy that stands as a distinct energy vector. Liquid FA features considerable volumetric energy density of 6.4 MJ/L and a specific energy density of 5.3 MJ/Kg that qualifies it in the prime seat as an energy source for transportation infrastructure. Additionally, the increasing research interest in FA decomposition is driven by the need for in-situ H_2 production, which plays a key role in the hydrogenation reactions of biomass into higher-value components. It is reported elsewhere in the literature that catalytic decomposition of FA is usually performed in poorly designed setups using simple glassware under magnetic stirring, thus demanding further energy investment to retain the used catalyst. Our work suggests an approach that integrates designing a distinct catalyst featuring magnetic properties with a robust setup that minimizes experimental & measurement discrepancies. One of the most prominent active species for the dehydrogenation/hydrogenation of biomass compounds is palladium. Accordingly, we investigate the potential of engrafting palladium metal onto functionalized magnetic nanoparticles as a heterogeneous catalyst to favor the production of CO-free H_2 gas from FA. Using an ordinary magnet to collect the spent catalyst renders core-shell magnetic nanoparticles as the backbone of the process. Catalytic experiments were performed in a jacketed batch reactor equipped with an overhead stirrer under an inert medium. Through a distinct approach, FA is charged into the reactor via a high-pressure positive displacement pump at steady-state conditions. The produced gas (H_2+CO_2) was measured by connecting the gas outlet to a measuring system based on the amount of the displaced water. The uniqueness of this work lies in designing a very responsive catalyst, pumping a consistent amount of FA into a sealed reactor running at steady-state mild temperatures, and continuous gas measurement, along with collecting the used catalyst without the need for centrifugation. Catalyst characterization using TEM, XRD, SEM, and CHN elemental analyzer provided us with details of catalyst preparation and facilitated new venues to alter the nanostructure of the catalyst framework. Consequently, the introduction of amine groups has led to appreciable improvements in terms of dispersion of the doped metals and eventually attaining nearly complete conversion (100%) of FA after 7 hours. The relative importance of the process parameters such as temperature (35-85°C), stirring speed (150-450rpm), catalyst loading (50-200mgr.), and Pd doping ratio (0.75-1.80wt.%) on gas yield was assessed by a Taguchi design-of-experiment based model. Experimental results showed that operating at a lower temperature range (35-50°C) yielded more gas, while the catalyst loading and Pd doping wt.% were found to be the most significant factors with P-values 0.026 & 0.031, respectively.

Keywords : formic acid decomposition, green catalysis, hydrogen, mesoporous silica, process optimization, nanoparticles
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