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Biomass reforming into hydrogen production with ZrO_2 using ReaxFF

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In recent decades, the sustainability field has received much attention for future energy conversion and storage because it provides clean, efficient, and sustainable energy for modern society. Among the sustainable-based resources, biomass is the most abundant renewable material and is produced several million tons worldwide and consists of three major components, cellulose (38-50%), hemicellulose (23-32%), and lignin (15-25%). It has been considered a great potential replacement for the hydrogen (H_2) energy produced from fossil fuels. Various thermochemical processes have been applied to valorise biomass into H_2 such as pyrolysis, gasification, aqueous phase reforming and catalytic reforming. Gasification is a fundamental process and can be done by treating biomass with water at supercritical conditions. However, the purity of H_2, formation of a higher amount of char, larger hydrocarbons and undesirable gases are the major challenges. Therefore, the use of a catalyst during the supercritical water gasification stage offers a potential route to produce high H_2 yield from biomass. Transition metal-based catalysts such as Ni, Co, Fe, CeO_2, ZrO_2, TiO_2, ZnO, and Al_2O_3, and their combinations, provide adequate support for the gasification process. Among these catalysts, ZrO_2 exhibits ubiquitous properties, including acidic and basic properties, redox characteristics, good thermal stability and stable at supercritical conditions. The ZrO_2 has been applied successfully for the gasification of biomass to reduce the tar formation, carbon deposition and enhance the H_2 yield from biomass intermediates. However, understanding of such gasification process with ZrO_2 catalyst at the molecular level is missing. Therefore, the present work mainly focuses on the investigation of the catalytic activity of ZrO_2 with biomass (cellulose and lignin) to unravel the mechanism at the molecular level for the generation of H_2 using the reactive force field (ReaxFF) molecular dynamics method.

The cleavage of cellulose and lignin molecules were investigated with ZrO_2 nanoparticle to find pathways towards the generation of H_2 by employing temperature from 300 K to 1100K. Three different sizes of ZrO_2 nanoparticles were considered with radius such as 2.0 nm, 3.0 nm and 4.0 nm to study the influence of the size effect. The obtained results have shown that cellulose undergoes ring-opening and dissociation which leads to the formation of a series of alcohols and thus results in the generation of H_2. Similarly, lignin displayed different cleavage pathways including C-O, O-H, C-C and C-H on the surface of the ZrO_2 to generate H_2. In order to confirm the catalytic activity of the ZrO_2 nanoparticle, ReaxFF simulations were performed without ZrO_2 catalyst by considering cellulose and lignin with water (500 K) and the results revealed that no reactions occurred. These results demonstrated that ZrO_2 can be a potential catalyst that enhances the generation of H_2 under supercritical water conditions.

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