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Sustainable hydrogen production and CO_2 mitigation from acetic acid dry reforming over Ni/Al_2O_3 catalyst

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ABSTRACT

Dry reforming of acetic acid (DRA) was first-time investigated on $10\%Ni/Al_2O_3$ at varied temperatures within 923–973 K employing several CO_2:CH_3COOH ratios of 1.5:2; 1:1; and 2:1. Depending on NiO particle size and location on support surface or inside the porous support structure, the H_2 reduction of NiO phase to active metallic Ni^0 form was evidenced at different reduction temperatures within 550–950 K and the estimated degree of reduction was about 73.68%. Weak, medium, and strong basic centres were evidenced on $10\%Ni/Al_2O_3$ via CO_2 desorption measurement. Increasing DRA temperature improved both CH_3COOH and CO_2 conversions with the corresponding apparent activation energy of 100.71 and 58.50 kJ mol^{-1}. As CO_2 partial pressure was increased from 0 to 40 kPa, the initial CH_3COOH conversion was noticeably enhanced from 30.5% to 97.8% whilst H_2/CO ratio always remained less than unity (0.73–0.77). Notably, DRA was a two-step process and barely negligible CH_4 intermediate product was evidenced since it was promptly reformed by CO_2 into syngas. The Ni^0 active form was not susceptible to oxidation during DRA and carbon deposited on spent catalyst surface was heterogeneous in nature with the evident co-existence of amorphous and graphitic carbons.

1. Introduction

Hydrogen has been broadly recognized as an efficient alternative and green energy for the promising substitution of draining fossil fuels, inducing substantially rising crude oil price worldwide since H_2 reportedly has great energy capacity of 120.7 kJ g^{-1} [1] and water is a sole by-product from H_2 combustion [2]. Apart from being efficiently used in fuel cells and hydrogen vehicles for environmentally friendly transportation, H_2 is also an important feedstock for petrochemical and gas processing industry for synthetic fuels and essential chemicals production [3]. At present, the majority of H_2 fuel is produced through natural gas steam reforming, coal gasification, and liquid fossil fuel

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