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CNFs-supported Pd series catalysts comparison for hydrogen evolution from additive free formic acid decomposition

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Searching for a safe and efficient H_2 generation/storage material has become a serious challenge toward a fuel-cell-based H_2 economy as a long-term solution. Herein we report the development of Pd nanoparticles catalysts supported on 5 different carbon nanofibers (CNFs): three different grades and nitrogen and oxygen functionalisations; each one via sol-immobilisation and impregnation techniques. Thorough characterisation has been carried out by XRD, XPS, TEM, SEM-EDX. The catalysts have been evaluated for the formic acid dehydrogenation, which has potential to be a safe and convenient H2 carrier under mild conditions. Those catalysts prepared by sol-immobilisation technique exhibit more activity when compared with catalysts prepared by impregnation due to the higher metal loading and higher Pd0/Pd ratio, smaller particle size (Fig. 1) and lower binding energies, leading to an improved activity due to the weaker interaction between the Pd nanoparticles and the formic acid. The heat treatment on CNFs has an important effect on catalyst activity, increasing with the annealing temperature (Fig. 2). Oxygen functionalities present a higher initial activity that could be addressed to a favoured deprotonation step due to the presence of O- on the surface, leading to an easier dehydrogenation of formic acid. However, deactivation was observed after 30 minutes due to CO evolution. The most active catalyst reached a remarkable TOF of 979 h-1 and high selectivity (>99%) at 30°C. Being this a great value for formic acid dehydrogenation at mild conditions however, further investigation is necessary in order to decrease the CO formation and improve reusability.

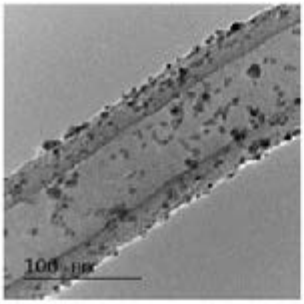

Figure 1. Bright field TEM micrograph of PdSI/CNF-HHT

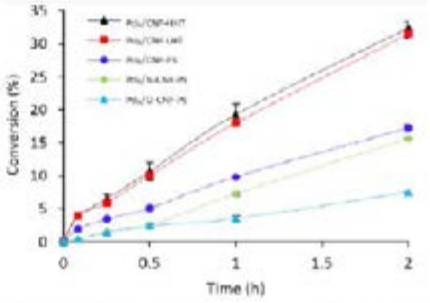

Figure 2. Formic acid dehydrogenation reaction on Pd on different supports.

Biography

Felipe Sánchez is from department of chemical engineering in Málaga. Currently, he joined in Cardiff University for PhD and pursuing 3rd year.

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