

Supporting Information

Preparation and performance of Pd_{Multi}@HCS catalyst with Pd nanoparticles partially embedded in the inner wall of hollow carbon sphere for the direct synthesis of hydrogen peroxide from hydrogen and oxygen

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Typical synthetic steps for Pd_{Multi}@HCS catalysts

(1) Preparation of SiO₂ nanospheres by Stöber method

SiO₂ nanosphere as a hard template were synthesized by Stöber method. 4.5ml TEOS was quickly added to a mixed solution of 62ml absolute ethanol, 25ml deionized water and 1.5ml ammonium hydroxide, and the reaction was stirred at 25°C for 2 hours. The products were washed with absolute ethanol and centrifuged one time, and then washed with deionized water and centrifuged three times, which denoted as SiO₂.

(2) SiO₂ nanospheres modified by DMDAAC

The SiO₂ nanospheres were ultrasonically dispersed in 100ml deionized water, then 1.5ml DMDAAC was added and stirred at 35°C for 16h. The products were washed with deionized water and centrifuged three times, which denoted as SiO₂-DMDAAC.

(3) Preparation of Pd_{Multi}/SiO₂-DMDAAC

The SiO₂-DMDAAC nanospheres were ultrasonically dispersed in 100ml deionized water, and 1ml H₂PdCl₄ (0.075M) was added and stirred at 35°C for 20h, then 500μl freshly prepared NaBH₄ (10wt%) was added into the solution slowly, and continued stirring at 35°C for 3h. The products were washed with deionized water and centrifuged for three times, and denoted as Pd_{Multi}/SiO₂-DMDAAC.

(4) Preparation of Pd_{Multi}/SiO₂-DMDAAC@RF, Pd_{Multi}/SiO₂@C and Pd_{Multi}@HCS

Firstly, Pd_{Multi}/SiO₂-DMDAAC nanospheres were ultrasonically dispersed in a mixed solution of 70ml deionized water and 30ml absolute ethanol, and stirred at 35°C for 10min. And 2.0g CTAB, 0.20g resorcinol and 0.2ml ammonium hydroxide were added successively, and stirred at 35°C for 1h. Then 0.286ml of formaldehyde solution was quickly added (resorcinol/formaldehyde (R/F) ratio was controlled to be 0.7), and the self-assembly polymerization reaction was carried out for 16h. The products were washed with absolute ethanol and centrifuged for three times to obtain Pd_{Multi}@SiO₂-DMDAAC@RF nanocomposites, which was dried overnight in an oven at 80°C. The dried products were carbonized at 600°C under N₂ atmosphere for 2h to obtain Pd_{Multi}@SiO₂@C nanocomposites. Then Pd_{Multi}@SiO₂@C

nanocomposites were dispersed in a mixed solution of 35ml deionized water and 5ml hydrofluoric acid and etched at room temperature for 24h to remove silicon. The products were washed with deionized water and centrifuged for three times and dried at 80°C overnight. Finally, the dried products were oxidized for 2h at 250°C in air and reduced for 2.5h at 250°C under H₂ atmosphere. The obtained Pd catalyst with Pd nanoparticles partially embedded in the inner wall of hollow carbon spheres was denoted as Pd_{Multi}@HCS.

The above typical synthesis methods were used for the preparation of Pd_{Multi}@HCS catalysts with different CTAB mass, but the CTAB mass were 0g, 1.0g, 1.5g, 2.0g and 4.0g, respectively.