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Highly dispersed Pd catalysts prepared by a sonochemical method for the direct synthesis of hydrogen peroxide

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ABSTRACT

Pd catalysts prepared by a sonochemical method that is known to be effective in metal dispersion are applied to direct H_2O_2 synthesis. To determine the effect of the preparation method on the catalytic performance, Pd catalysts were supported on TiO_2 and SiO_2 by a sonochemical method and general incipient wetness method (IWI), and supports were compared. The sonochemical preparation method achieved the highest Pd dispersion value of 16.6% in the TiO_2 supported catalyst, which is attributed to a synergistic effect between the sonochemical method and TiO_2 support. A near-homogeneous and small size nucleation is often reported in sonochemical methods, and oxygen vacancies in TiO_2 are reported to be strong adsorption sites for Pd nucleation. However, SiO_2 support has not been reported to have obvious oxygen vacancies, and sonochemically prepared Pd/SiO_2 showed the lowest Pd dispersion in addition to a large Pd particle size. In the direct synthesis of H_2O_2, a remarkably high H_2 conversion of 22% was achieved in sonochemical Pd/TiO_2, which led to the highest H_2O_2 yield of 16% and H_2O_2 production rate of 729.0 mmol/g_{Pd} •$_h$.

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1. Introduction

Hydrogen peroxide is a valuable and eco-friendly oxidant compared to other oxidants (*e.g.*, HNO_3, N_2O, and NaClO) [1–3]. A major advantage of hydrogen peroxide is that the byproducts of hydrogen peroxide (O_2 and H_2O) following oxidation are 'greener' than those of other oxidants (*e.g.*, NO_x, N_2O and NaCl) [4,5]. Also hydrogen peroxide is the most efficient oxidant; the content of active oxygen in hydrogen peroxide is 47% by weight, which is the highest value among oxidants, with the exception of molecular oxygen [5,6]. The global demand of hydrogen peroxide has increased due to its widespread use as a bleaching agent of pulp or paper and as a raw material for various syntheses that produce chemicals in the presence of proper catalysts (*e.g.*, titanium silicalite-1) [7,8]. Hydrogen peroxide can be used in conjunction with a titanium silicalite-1 catalyst for ammoximation, hydroxylation, oxidation and epoxidation processes [8–12].

As the demand for hydrogen peroxide has increased, the number of plants that manufacture hydrogen peroxide has also increased [5]. Most hydrogen peroxide is produced by an anthraquinone oxidation process (AO process), but it has several drawbacks in environmental and economic aspects [5,13]. The AO process uses highly toxic chemicals, including anthraquinones (*e.g.*, 5,6,7,8-tetrahydroanthrahydroquinone and anthrahydroquinone) and organic solvents (*e.g.*, naphthalene and octanol), and is economically viable only at a large scale plant due to its complicated setup [13]. The transportation of hydrogen peroxide from manufacturing sites to the point of use requires an additional cost and is accompanied by the danger of explosion [13].

Therefore, alternative hydrogen peroxide production processes that use greener materials with an on-site route have been studied. The direct synthesis of hydrogen peroxide from H_2 and O_2 has received attention as a potential candidate [14–16]. The direct synthesis of hydrogen peroxide uses greener solvents (*e.g.*, water, ethanol and methanol), and the byproducts of the process, H_2O and O_2, are eco-friendly. In addition, it can be economically operated at a much smaller scale than the AO process; therefore, on-site installation is feasible and the cost of hydrogen peroxide transportation is obviated. Also, on-spite produced hydrogen peroxide

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