Bimetallic Nanocatalysts Based Green Process for Production of Hydrogen Peroxide

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Abstrak

[ABSTRAK
Tujuan dari penelitian ini adalah untuk mengembangkan proses ?green? baru untuk produksi H2O2 melalui rute sintesis langsung, di mana selama reaksi hidrogen dan oksigen saling kontak satu sama lain. Sebuah pendekatan elektrokimia dengan rotating ring disk electrode (RRDE) telah dieksplorasi dan dikembangkan secara sistematis yang bertujuan untuk mengukur H2O2 yang diproduksi. Dua metode yang berbeda - co-reduction and successive reduction dengan menggunakan microwave diadopsi untuk mempersiapkan bimetal nanocatalysts Pd-Au/C. Hubungan antara struktur nanocatalysts dan aktivitas katalitik dalam proses sintesis langsung diselidiki. Bimetal Pd-Au/C yang telah disintesa, dikarakterisasi dengan ICP-AES, XRD, SEM, TEM, dan XAS untuk pemahaman yang lebih baik dalam aktivitas katalitik sintesis H2O2 secara langsung.

Pendekatan dalam elektrokimia untuk mengukur H2O2 yang dihasilkan dari sintesis langsung telah berhasil dilakukan dengan sistem reaksi 2, dimana katalis tersebar secara homogen dalam larutan. Kurva kalibrasi variasi konsentrasi H2O2 dibuat dalam parameter 0,891 V (vs Ag/AgCl) dan dengan scan rate 50 mV/s. CR Pd3%-Au2%/C yang disintesa oleh co-reduction merupakan optimal loading dengan produktivitas H2O2 65,8 mol.kgcat-1h-1. Produktivitas ini lebih tinggi dari sample katalis lainnya, seperti monometallic Pd0%-Au5%/C & Pd5%-Au0%/C dan bimetal SR Pd-Au/C yang disintesis dengan successive reduction. Produktivitas yang lebih tinggi atau lebih rendah dari satu sampel ke yang lain dijelaskan oleh parameter-parameter seperti ukuran partikel, struktur bimetal Pd-Au/C, bidang kristal yang selektif, dan peran paladium dan emas. Ukuran partikel yang lebih kecil cenderung memiliki Pd yang lebih banyak, sedangkan yang lebih besar cenderung memiliki Au yang lebih banyak. Ukuran partikel yang lebih kecil memiliki daerah permukaan yang lebih tinggi, sehingga produktivitas meningkat. Namun, jika ukuran partikel terlalu kecil, permukaan yang aktif atau bidang kristal yang selektif mungkin sedikit muncul (seperti dapat dilihat dalam SR Pd-Au/C), sehingga produktivitas menurun.
vii

Dari analisis XAS, CR Pd-Au/C memiliki struktur Au lebih banyak di core dan Pd

lebih banyak di shell. Struktur SR Pd-Au/C di beberapa bagian dari katalis adalah Au lebih banyak di core dan Pd lebih banyak di shell, sementara pada bagian lain, Pd lebih banyak di core dan Au lebih banyak di shell. Nilai Q pada SR PdAu (0,638) lebih tinggi daripada CR PdAu (0,605), yang menunjukkan bahwa keberadaan atom Au di shell SR PdAu lebih dari itu CR PdAu. Perbedaan dalam struktur adalah salah satu alasan mengapa produktivitas H2O2 CR PdAu lebih tinggi dari SR PdAu. Peran Pd adalah untuk memberikan luas permukaan untuk oksidasi selektif dari hidrogen dan peran Au adalah untuk menyediakan situs aktif untuk reaksi dekomposisi dan hidrogenasi H2O2.
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ABSTRACT
The purpose of this study is to develop a new green process for production of H2O2 through the direct synthesis route, of which the hydrogen and oxygen contacts each other during the reaction. An electrochemical approach with the rotating ring disk electrode (RRDE) had been systematically explored and developed accordingly to measure the produced H2O2. Two different methods ? co-reduction and successive reduction prepared in the microwave were adopted to prepare bimetallic Pd-Au/C nanocatalysts. The relationship between the structure of prepared nanocatalysts and their catalytic activity in the direct synthesis process were investigated. As synthesized bimetallic Pd-Au/C were characterized by ICP-AES, XRD, SEM, TEM, and XAS for better understanding in the catalytic activity of direct synthesis of H2O2.

The approach in the electrochemical to measure H2O2 produced from the direct synthesis has been successfully done with the reaction system 2, where the catalyst is dispersed homogenously in the solution. The calibration curve of the different concentration of H2O2 is made in the parameter of 0.891 V (vs Ag/AgCl) and with the scan rate 50 mV/s. The optimum loading of samples prepared by co reduction was observed in CR Pd3%-Au2%/C with the productivity of H2O2 is 65.8 mol.kgcat
-1h-1. This productivity is higher than the other prepared catalysts, such as monometallic Pd0%-Au5% & Pd5%-Au0% and bimetallic SR Pd-Au/C that is prepared by successive reduction. The higher or the lower productivity of one sample to another is explained by the parameter of the particle size, the structure of the bimetallic Pd-Au/C, the selective crystalline plane, and the role of palladium and gold. The smaller the particle size tends to Pd rich, while the larger one tends to Au rich. The smaller particle size yielded in the high surface area, thus the productivity increases. However, if the particle size is too small, the active site or selective crystalline plane may be slightly appeared (as can be seen in SR Pd-Au/C), thus the productivity decreases.

From XAS analysis, the structure CR Pd-Au/C is Au rich in core and Pd rich in

shell. The structure of SR PdAu at some part of catalyst is Au rich in core and Pd rich in shell, while at the other part, the structure is Pd in core and Au in shell.
ix
The Q value of SR PdAu (0.638) is higher than that of CR PdAu (0.605), which indicates that the existence of Au atoms in the shell of SR PdAu is more than that of CR PdAu. The difference in their structure is one reason why the H2O2 productivity of CR PdAu is higher than SR PdAu. The role of Pd is to provide the surface area for the selective oxidation of hydrogen and the role of Au is to provide inactive site for the reaction of decomposition and hydrogenation of H2O2., The purpose of this study is to develop a new green process for production of H2O2 through the direct synthesis route, of which the hydrogen and oxygen contacts each other during the reaction. An electrochemical approach with the rotating ring disk electrode (RRDE) had been systematically explored and developed accordingly to measure the produced H2O2. Two different methods – co-reduction and successive reduction prepared in the microwave were adopted to prepare bimetallic Pd-Au/C nanocatalysts. The relationship between the structure of prepared nanocatalysts and their catalytic activity in the direct synthesis process were investigated. As synthesized bimetallic Pd-Au/C were characterized by ICP-AES, XRD, SEM, TEM, and XAS for better understanding in the catalytic activity of direct synthesis of H2O2.
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