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Electrochemical and Surface Characterization of Au Nanoparticle/Multiwalled Carbon Nanotube Composites

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A novel type of gold nanoparticle/multiwalled carbon nanotube (AuNP/MWCNT) composite electrodes is presented. The electrochemical reduction of oxygen on these hybrid electrodes was studied using the rotating disk electrode (RDE) method. The AuNP/MWCNT nanocomposites were prepared by sputter deposition of gold in argon atmosphere on MWCNTs followed by heat-treatment of the catalyst at different temperatures. Au nanoparticles with diameter around 20 nm were dispersed at the tips and on the sidewalls of nanotubes. High resolution scanning electron microscopy (HR-SEM), glancing incidence angle X-ray powder diffraction (GIXRD) and small-angle X-ray scattering (SAXS) techniques were employed to characterize the surface structure and morphology of catalyst materials.

A parametric study of O_2 reduction on AuNP/MWCNT modified GC electrodes, in which the influence of heating treatment of Au nanoparticles on the morphology and electrocatalytic activity toward the ORR in sulphuric acid solution was investigated. It was found that by varying the annealing temperature it is possible to modify essentially the morphology of Au nanoparticles (Fig. 1) and thus, to tailor their electrochemical behavior. Heat-treatment conducted at 300 ° C proved to be beneficial to ensure a larger active area and increased electrocatalytic activity.

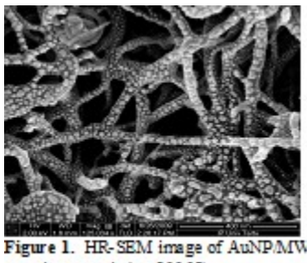

Figure 1. HR-SEM image of AuNP/MWCNT samples annealed at 300 °C.

Figure 2 presents a comparison of the oxygen reduction results obtained with AuNP/MWCNT catalysts heat-treated at different temperatures. The values of half-wave potential ($E_{1/2}$) for O_2 reduction on AuNP/MWCNT modified electrodes were determined to be -75 mV, -60 mV and -85 mV for the composite catalysts annealed at 250, 300 and 400 ° C, respectively. The values of specific activity (SA) were determined at 0.1 V taking into account the real surface area of AuNPs for each electrode. On the basis of recent considerations [1] one might expect that the value of SA is unchanged for the AuNP/MWCNT nanocomposites used in the present work. The average particle size is larger than 5 nm. Apparently, the electrocatalytic activity of the nanogold material toward oxygen reduction might depend on the catalyst support and on the particular arrangement of AuNPs within the structure of the three-dimensional nanocomposite film.

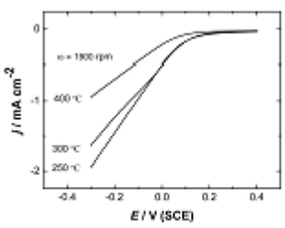

Figure 2. A comparison of RDE voltammetry curves for O_2 reduction on AuNP/MWCNT modified GC electrodes in O_2 saturated 0.5 M H_2SO_4. $v = 10$ mV s^{-1}, $\omega = 1900$ rpm.

The Tafel plots of O_2 reduction for all the AuNP/MWCNT materials studied, were obtained from the kinetic currents. The current densities were normalized to the real area of gold. A high Tafel slope value was observed at low overpotentials (ca -200 mV dec^{-1}). A typical slope value for O_2 reduction on gold in acid solution is -120 mV dec^{-1}, which corresponds to a slow transfer of the first electron to O_2 molecule [2].

It was shown in our earlier work that the AuNP/MWCNT modified GC is more active catalyst for O_2 reduction than bulk gold. The $E_{1/2}$ value for O_2 reduction on AuNP/MWCNTs modified GC electrodes shifted by 50 mV to more positive potentials as compared to that of the bulk Au electrode (3). The oxygen reduction behavior of the AuNP/MWCNTs modified GC electrodes was similar to that observed in our previous studies [3,4]. The successful preparation of AuNP/MWCNT composite by magnetron sputtering opens up the possibility of making an efficient dispersion of nanoparticles for electrocatalyst design.

References

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