PHOTOCATALYTIC SYSTEMS FOR HYDROGEN PRODUCTION FROM Ta$_2$O$_5$ NANOTUBES SENSIBILIZED WITH CdSe

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Introdução

Hydrogen is considered an ideal fuel for the future, and its use represents a visionary strategy because it is abundant, clean, flexible, and secure. After the work of Honda and Fujishima in 1972[1], the H$_2$ production reactions has emerged as a viable option for cleaner fuels productions due to smaller cost and to the absence of carbon compounds derived. Among the various technologies the water splitting method has been considered a form of clean energy and low cost using solar energy. Recently studies show the tantalum oxide (Ta$_2$O$_5$), as a promising material for the hydrogen photogeneration [2]. However, its activity is limited to the ultraviolet (UV) region, which accounts for only 4-5% of the solar radiation. In order to get better photocatalytic activity, semiconductors, such as CdS and CdSe, have been used as sensitizer into Ta$_2$O$_5$. In this work, the Ta$_2$O$_5$ was used in the form of nanotubes (NTs), synthesized by an anodization with sulfuric acid (H$_2$SO$_4$) using tantalum and copper sheets as electrodes for 10 minutes in thermostatic bath, and submitted for an annealing at 800ºC for 1 hour. The CdSe colloidal solution was prepared using distinct steps. The first step consisted of reducing selenium to selenite through high-frequency ultrasonic radiation for 5 min. In the second step the L-cysteine (1.2 mmol) was added to the mixture CdCl$_2$ (0.4mmol) dissolved in 200mL water, after the pH was adjusted to 9.0. Finally the selenite solution was added in the mixture L-cysteine/CdCl$_2$. After manual stirring, a clear, yellow suspension of CdSe QDs was obtained. Then impregnates in Ta$_2$O$_5$ nanotubes shows the evolution of H$_2$ photogeneration with the irradiation time. Since, nanoparticles and amorphous Ta$_2$O$_5$ nanotubes generate the lowest H$_2$ production, because they have a larger number of defects that act as electron traps increasing the recombination and decreasing the electronic transport. The nanotubes submitted for an annealing at 800ºC exhibit an improvement in the generation of H$_2$, this is due to crystalline form which has fewer defects, and consequently increases electronic mobility. Thus when the nanotubes are sensitized with CdSe the production increases by 8.5 times in regards to amorphous nanotube. It was observed that nanotubes when sensitized with CdSe show an improvement in catalytic activity.

Resultados e Discussão

Different temperatures and potential were tested to obtain Ta$_2$O$_5$ nanotubes and it was found that the temperature has a significant effect on the crystallinity and the average length of the nanotubes and consequently the length of the grass, which is defined as oxide barrier layer formed on the surface of nanotubes not solubilized. This occurs because the increase in temperature causes better solubilization of Ta$_2$O$_5$ the electrolyte solution, favoring the solubilization process of the oxide barrier layer. The best condition of obtaining the nanotubes was 35ºC and 50V. The analysis of TEM together with EDS analysis identified a nanotubular morphology and the presence of Cd and Se elements in this array Ta$_2$O$_5$. The photocatalytic activity of the synthesized samples was determined by the photo-generation of hydrogen by water splitting in a solution containing 0.1 M and 0.1 M S$_2$SO$_4$, used as sacrificial agents. The figure 1 shows the evolution of H$_2$ photogeneration with the irradiation time. Since, nanoparticles and amorphous Ta$_2$O$_5$ nanotubes generate the lowest H$_2$ production, because they have a larger number of defects that act as electron traps increasing the recombination and decreasing the electronic transport. The nanotubes submitted for an annealing at 800ºC exhibit an improvement in the generation of H$_2$, this is due to crystalline form which has fewer defects, and consequently increases electronic mobility. Thus when the nanotubes are sensitized with CdSe the production increases by 8.5 times in regards to amorphous nanotube. It was observed that nanotubes when sensitized with CdSe show an improvement in catalytic activity.

Conclusões

In NT synthesis can be concluded that the temperature has a great influence on the length of the nanotubes and the tension influences the shape of NT. The nanotubes was evaluated with CdSe and a significant improvement was observed for catalytic activity.

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1 A. Fujishima, K. Honda, Nature 238 (1972), 37-38