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Composite photocatalysts prepared with materials from spent Li-ion batteries and g-C_3N_4 for the photoreforming of organics in aqueous solution to obtain H_2

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Every year, millions of tons of spent lithium-ion batteries (s-LIBs) are generated. The recycling methods to recover the precious metals they contain, mainly Ni, Co and Li, are energy-intensive, non-selective and expensive. The development of processes for the direct reuse of s-LIBs is an urgent issue[1]. The current work is an attempt to develop innovative materials, as graphitic carbon nitride (g-C_3N_4) enriched with a metal-based co-catalyst obtained from the mechanical grinding of the mixture of anode and cathode powders contained into the spent s-LIBs, called *black mass* (BM).
The photoreforming technology of aqueous solutions of organics is aimed at addressing energy and environmental concerns by obtaining H_2 from water and simultaneously oxidizing organic molecules to other higher added-value substances. In the last decade, carbonaceous materials such as C_3N_4 have demonstrated good performances as free metal photocatalysts in this type of reactions. Graphitic carbon nitride (g-C_3N_4) is a semiconductor that possesses optimal characteristics to be used as a photocatalyst even under solar irradiation, however it can suffer from recombination of the photogenerated electron-hole pairs with consequent decrease in photocatalytic activity. Its coupling with various materials can improve its performance. In fact, g-C_3N_4 composites with graphene[3] or TiO_2[4] have given excellent results in terms of H_2 productivity by photoreforming.
In this research, BM from s-LIB, containing ca. 60 % w/w of Co, was used either raw or after being oxidized or reduced with H_2 at 600 °C for 6 h. g-C_3N_4 was prepared by calcination of melamine, urea or thiourea. Various sets of composites were prepared by following some different strategies to obtain g-C_3N_4/BM photocatalysts with an amount of BM enough to bring to the composite a content of Co in the range 0.5 to 2 % w/w. Additionally, hydrochar (HC) obtained from the hydrothermal combustion of orange peel waste was also used as a co-catalyst. The obtained materials were tested for H_2 production using aqueous solutions of ethanol, glycerol or triethanolamine irradiated with UV or natural solar light. Photoreactivity was performed in the absence or presence of 1% Pt. The best results were obtained with catalysts containing g-C_3N_4 with a 1% of Co from BM. The solid obtained by grinding g-C_3N_4 and BM in a planetary mill gave a H_2 productivity of 0.9 mmol h^{-1} g^{-1} whereas the composite obtained by grinding melamine and BM, followed by calcination at 520 °C for 2 h gave 1.7 mmol h^{-1} g^{-1}. The best material was obtained by ball-milling melamine and the BM obtained after successive cycles of reduction and oxidation giving a H_2 productivity of 2.2 mmol h^{-1} g^{-1}.

References

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