PHOTOCATALYTIC PRODUCTION OF HYDROGEN FROM CARBONIC ACIDS ON TANTALUM-CONTAINING COMPOSITES UNDER VISIBLE LIGHT

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A promising option for solving environmental problems associated with the use of fossil fuels is the development of technologies for producing “green” hydrogen from aqueous-organic systems in the presence of semiconductor photocatalysts when exposed to solar energy.

The high photocatalytic activity of iron-containing composites obtained by autowave combustion of ferroalloys in nitrogen during the generation of hydrogen from carboxylic acids (HCOOH, $H_2C_2O_4$) under UV irradiation has been established [1]. The effectiveness of the catalysts is associated with the presence in the ceramic matrix of wide-gap semiconductors (β-Si_3N_4, β-$Si_3Al_3O_3N_5$) and a phase of metallic iron (α-Fe), the partial dissolution of which in the presence of H_2O_2 creates conditions for the photo-Fenton process.

To shift the photocatalytic activity of silicon nitride-based composites to the visible light region, the ceramic matrix was modified by introducing semiconductor tantalum compounds (TaON, Ta_2O_5) into its composition. When assessing the performance of Ta-containing catalysts in the process of generating hydrogen from carboxylic acids ($H_2C_2O_4$, HCOOH) under UV and visible light, the highest activity was established for the sample synthesized by nitriding ferrosilicoaluminum (FSA) with the addition of 10% wt. metal tantalum, which is due to its better morphological and optical properties. [2]

To establish the role of iron and ceramic matrix semiconductors in the photocatalytic process of H_2 generation, a composite obtained by nitriding a mixture of elemental powders (Si and Al) with the addition of Ta in an amount of 10 wt. % was studied. The absence of iron in the composites eliminates the role of the photo-Fenton process.

The processes of photocatalytic production of hydrogen from aqueous solutions of carboxylic acids (HCOOH, $H_2C_2O_4$) under visible light using tantalum-containing metal-ceramic composites based on silicon nitride and a mixture of silicon and aluminum powders with tantalum additives, in the absence and with the addition of hydrogen peroxide, depending on the concentration of the substrate, have been studied and pH of the suspension.

It has been established that the dependence of the rate of photocatalytic hydrogen production on the concentration of formic acid without hydrogen peroxide obeys the Langmuir–Hinshelwood mechanism. A sharp increase in the rate of the photocatalytic process in the presence of hydrogen peroxide is observed with increasing concentration of formic acid. The highest rate of hydrogen generation from formic acid was recorded on an iron-containing composite synthesized from FSA, without the addition of hydrogen peroxide.

REFERENCES

[1] I.A. Artukh, K.A. Bolgaru, K. A. Dychko , A.V. Bavykina, F. Sastre, L.N. Skvortsova Hydrogen production by Photocatalytic Degradation of Organic Substances Using Iron-Containing Metal-Ceramic Composites Under UV and Visible-Light Irradiation // J. ChemistrySelect. –2021. N 6. –pp. 10025–10032.

[2] Скворцова Л.Н., Болгару К.А., Казанцева К.И., Тихонова И.А., Регер А.А., Дычко К.А Фотокаталитическая деградация фармацевтических поллютантов при облучении УФ и видимым светом с применением железосодержащих металлокерамических композитов // Известия вузов. Физика. –2022. –Т.65, № 11, –С. 145–154.