EasyChair Preprint

№ 2172

Mineralization of Phenol Using Au/Ceo2-ZnO Photocatalysts

Claudia Martínez Gómez, Gloria Alicia del Ángel, Israel Rangel-Vázquez, Ramón Zarraga Nuñez, Emmanuel Castillo Toxqui and Gustavo Rangel Porras

December 16, 2019

MINERALIZATION OF PHENOL USING Au/CeO_2-ZnO PHOTOCATALYSTS

Claudia. M. Gómez [1*], G. del Ángel [2], I. Rangel-Vázquez [2], R. Zarraga Núñez [1], E. Castillo Toxqui[3], G. Rangel Porras [1]

[1]*Departamento de Química, División de Ciencias Naturales y Exactas, Campus Guanajuato de la Universidad de Guanajuato Noria Alta S/N, Col. Noria Alta, Guanajuato, Gto. C.P. 36050, México*

[2]*Departamento de Química, Área de Catálisis, Universidad Autónoma Metropolitana-Iztapalapa Av. San Rafael Atlixco N° 1865, México 09340 D.F. México.*

3Departamento de Ciencias básicas Instituto Tecnológico Superior de Guanajuato, Carretera Guanajuato a Puentecillas Km 10.5, Puentecillas, 36262 Guanajuato, Gto; México.

**claudia.martinez@ugto.mx*

Resumen

Los materiales fotocatalíticos de Au soportado al 1 % en CeO_2-ZnO se prepararon por el método de impregnación por vía húmeda variando concentraciones de zinc (0.5, 1.5 y 2.5 % en peso); el metal soportado fue reducido a alta temperatura en una atmósfera reductora con flujo de hidrógeno gas. Los materiales (ACZ) fueron caracterizados con las técnicas analíticas de Difracción de Rayos X, Microscopia Electrónica de Barrido con detector EDS, Espectroscopía UV-Vis de Reflectancia Difusa y sus propiedades texturales se determinaron por fisisorción de N_2. Se probó la actividad fotocatalítica de los materiales en la degradación del ácido 2,4-diclorofenoxiacético (2,4-D) y azul de metileno (AM), dos contaminantes orgánicos que se encuentran presentes en aguas residuales. Los resultados se siguieron mediante espectroscopía UV-vis y por la determinación de COT, demostrando que fueron efectivos degradando más del 85 al 95 % de la concentración inicial de los contaminantes a partir de 240 min en el caso del 2,4-D y 180 min en el caso del AM. En ambos casos el fotocatalizador más activo fue el ACZ1.5, debido a sus propiedades fisicoquímicas. La impregnación de Au en los soportes de CeO_2-ZnO, mejoró notablemente la actividad de los soportes solos.

Palabras clave: **Au/CeO_2-ZnO, fotodegradación, propiedades fisicoquímicas, acid 2,4-D, azul de metileno**

Abstract

Photocatalytic materials of 1% supported Au in CeO2-ZnO were prepared by the wet impregnation method varying the zinc concentrations (0.5, 1.5 and 2.5% by weight); The supported metal was reduced at high temperature in a reducing atmosphere with hydrogen gas flow. The materials (ACZ) were characterized with the analytical techniques of X-ray Diffraction, Scanning Electron Microscopy with EDS detector, UV-Vis Diffuse Reflectance Spectroscopy and their textural properties were determined with N2 fisisorption. The photocatalytic activity of the materials was tested in the degradation of 2,4-dichlorophenoxyacetic acid (2,4-D) and methylene blue (AM), two organic pollutants that are present in wastewater. The degradation results were followed by UV-vis spectroscopy and by the determination of total organic carbon, demonstrating that they were effective by degrading more than 85 to 95% of the initial concentration of the contaminants from 240 min in the case of 2, 4-D and 180 min in the case of AM. In both cases the most active photocatalyst was ACZ1.5, due to its physicochemical properties. The impregnation of Au in the CeO2-ZnO supports markedly improved the activity of the supports.

Keywords: **Au/CeO_2-ZnO, Photodegradatión, Physicochemical properties, acid 2,4-D, methylene blue**

Por medio del presente confirmo que estoy de acuerdo en someter el trabajo para su posible publicación en Topics in catalysis en caso de ser seleccionado.

1. Introductión

The problem of water derives from the increase of the population, from its demand for both food and material goods, from the scarcity and contamination of water systems due to the discharge of contaminated effluents from industry, has yielded alarming levels of elements pollutants, including dyes, in recent decades [1].

As a result, the search for new water treatment methods has been promoted, such as the Advanced Oxidation Processes (POA), which ensure the complete elimination of such compounds. The main advantage of POAs is that organic pollutants are oxidized to CO_2, involving highly reactive hydroxyl radicals formed in the médium [1].

Photodegradation catalytic is one of the most promisisng methods for the mineralization of recalcitrans compounds, being TiO_2 the most used. However owever, this material has performance deficiencies and the main one is the short time of recombination of the electron-hole pair.

CeO_2 is a chemically stable oxide (cerium being the most abundant element of rare earths) that, according to its physical and chemical properties, has been shown to have great advantages for photocatalysis, this is mainly attributed to its great ability to oxygen storage and its high oxide-reducing capacity [1]; in addition to that it has presented activity in the visible region[2].

On the other hand, ZnO is a broadband semiconductor widely studied as a potential alternative in photocatalytic applications, especially due to its morphological versatility, high photosensitivity and chemical stability, as well as being non-toxic and relatively inexpensive [3].

The impregnation of metals has increased photoactivity and one of the most used is Au, which is capable of reducing molecules such as oxygen to generate reactive species and; The gaps that remain in the valence band of ZnO are involved in oxidation reactions to generate hydroxyl radicals.

Due to this antecedents this work is focused to the study of mineralization of acid 2,4-D and methylene blue, with material of Au/CeO_2-ZnO

2. Experimental

The materials were sintetized by wet impregnation of Au in supports of CeO_2-ZnO, after this materials were dryed, then they received an oxidation treatment and finally a reduction in hydrogen flow at 400 ° C.

The materials were characterizated by by nitrogen fisisorcion, x-ray diffraction, uv-vis spectroscopy and scanning electron microscopy.

The reaction of photodegradation was follow by UV-vis spectroscopy and total organic carbon (TOC).

3.Results

In the Fig. 1 for CeO_2, the peaks observed in angle 2θ at 28.6º, 33.0º, 47.5º, 56.4º, 59.1º, 69.4º, 76.7º and 79.2º, correspond to the diffraction planes (111), (200) , (220), (311), (222), (400), (331) and (420) respectively. The ZnO with planes (100), (002), (101), (103), (112), in 2θ equal to 31.74 °, 34.36 °, 36.20 °, 62.93 ° and 67.97 °, indicate that it is a structure hexagonal crystalline (zincite).

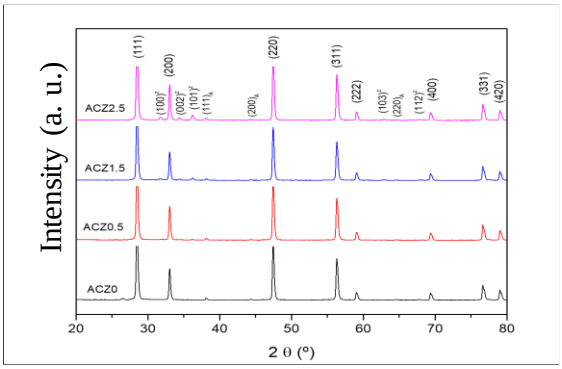

Figura 1. XRD of the materials ACZ

In the table 1 the physical properties of the different materials are observed, in this it is observed that the properties of the ACZ1.5 material do not follow a trend compared to the others, this could provide different photocatalytic properties in the reactio

Table 1. Physicochemical properties

Catalyst	Surface área m^2/g	Cristal size (nm)	Pore volume (cm^3/g)
ACZ0	5.0	7.92	0.0121
ACZ0.5	5.1	8.16	0.0139
ACZ1.5	5.2	8.71	0.0140
ACZ2.5	4.7	7.68	0.0129

Photocatalytic degradation of 2,4-D acid was evaluated for 4 hours under ultraviolet irradiation, degrading almost entirely and obtaining very

favorable results from 240 minutes in the case of 2,4-D and from 180 minutes for blue methylene (AM). Therefore, it was decided to shorten the reaction times to better appreciate the differences in reaction kinetics between the different materials.

Table 2. 2,4-D acid degradation kinetics

Catalyst	K_{app} (min^{-1})	$t_{1/2}$ (min)
ACZ0	0.0090	77.01
ACZ0.5	0.0096	72.20
ACZ1.5	0.0105	66.01
ACZ2.5	0.0100	69.31

Table 3. Methylene blue degradation kinetics

Catalysts	K_{app} (min^{-1})	$t_{1/2}$ (min)
ACZ0	0.0216	32.09
ACZ0.5	0.0201	34.48
ACZ1.5	0.0234	29.62
ACZ2.5	0.0225	30.80

In the table 2 and 3 it can been see that the material with the best activity is the ACZ1.5, due to their physicochemical properties.

4. Conclusions

The XRD shows the characteristic peaks of the materials.

There is no change in the specific area and this was not determined in the activity of the materials.

The best material was the ACZ1.5

5. Acknowledgment: To DAIP for the support

6. References

[1] Claudia M. Gómez, E. Ramos, G. del Ángel, I. Rangel, JCTB, 91, (2016), 2211-2222

[2] E. Gaya, U.I., Abdullah, A.H., J. Photochem. Photobiol. Photochem. Rev. 9, (2008) 1-12

[3] E. Sherly, J. Judith, L. Kennedy, Journal of Molecular Structure 1099 (2015) 114-125