Nanocatalyst based on thermally expanded phlogopite, Pd(II) and Cu(II) compounds for oxidation of CO and SO_2 with atmospheric oxygen

Tatyana Rakitskaya, Anna Nazar*, Tatyana Kiose

Faculty of Chemistry and Pharmacy, Odesa I.I. Mechnikov National University, 2, Dvoryanska St., 65082, Odesa, Ukraine

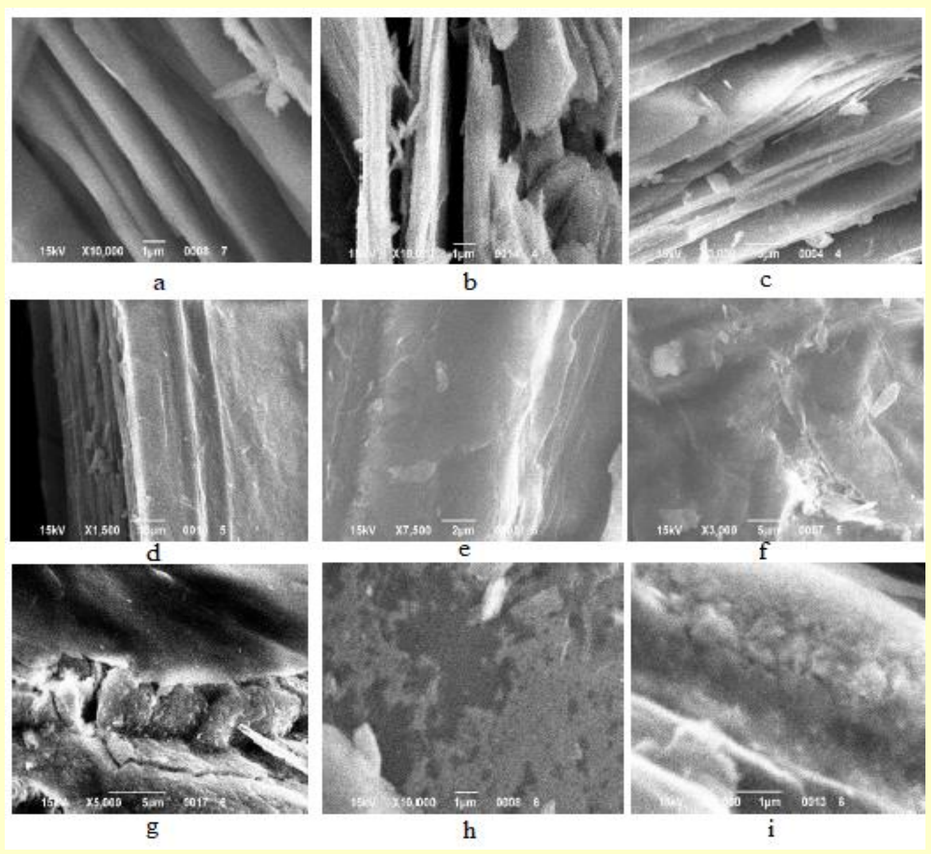
**e-mail: annnzr1401@gmail.com*

Introduction

A common situation is when carbon monoxide and sulfur dioxide are simultaneously present in the waste gases of industrial enterprises. In this regard, research on the polyfunctionality of the Pd(II)-Cu(II)/S (S – different carriers) nanocatalyst and the detection of the influence of SO_2 on its activity are relevant. Natural materials, including polyphase phlogopite concentrate, are effectively studied as carriers ($\bar{S}$) of palladium(II) and copper(II) compounds. The phase composition of the phlogopite concentrate significantly affects the activity of the Pd(II)-Cu(II)/$\bar{S}$ catalyst.

Characterization

SEM characterization



SEM images of samples of natural (a), thermally expanded (b, c) and acid-modified phlogopite (d-i).

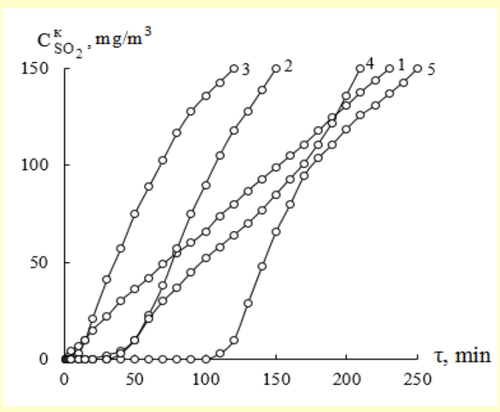
Catalytic properties

Table

Influence of the effective contact time of the gas-air mixture with the catalyst and the pH of the suspension on the degree of CO conversion in the presence of catalysts K_2PdCl_4-$Cu(NO_3)_2$-KBr/$\bar{X}$H-TS-Phl-1.*

Carrier	τ_0, min	τ_{MPC}, min	$Q_{theor}\cdot10^4$, mol SO_2	$t_{1/2}$, s	$k_{1/2}\cdot10^4$, s^{-1}	K_S
TS-Phl	3	15	2.40	6600	1.04	7.5
1H-TS-Phl-1	5	15	1.04	3000	2.30	15
2H-TS-Phl-1	20	50	1.91	3000	2.30	25
3H-TS-Phl-1	30	50	2.80	8400	0.82	12.5
6H-TS-Phl-1	100	120	3.61	9600	0.72	2.7

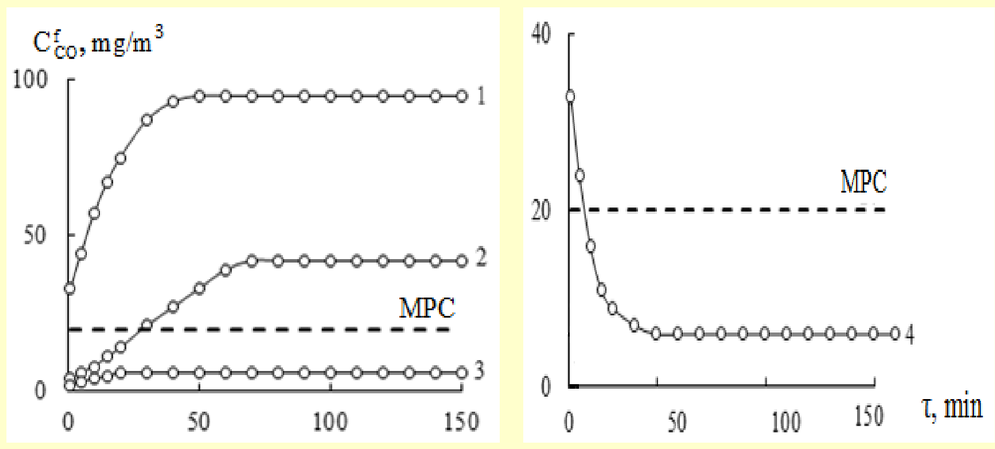
**Conditions: $C_{Pd(II)} = 2.72\cdot10^{-5}$; $C_{Cu(II)} = 5.9\cdot10^{-5}$; $C_{KBr} = 1.02\cdot10^{-4}$ mol/g; $C^{in}_{SO_2} = 150$ mg/m³; U = 4.2 cm/s ; t = 20 °C).*



Dependence of the final concentration SO_2 on time τ in the reaction of oxidation of sulfur dioxide with air oxygen in the presence of the composition Pd(II)-Cu(II)-KBr/$\bar{S}$

$\bar{S}$: 1 –TS-Phl; 2 – 1H-TS-Phl-1; 3 – 2H-TS-Phl-1; 4 – 3H-TS-Phl-1; 5 – 6H-TS-Phl-1.

Catalytic properties



Dependence of the final CO concentration on time τ in the oxidation reaction of carbon monoxide with atmospheric oxygen in the presence of a catalyst K_2PdCl_4-$Cu(NO_3)_2$-KBr/$\bar{S}$.

$\bar{S}$: *1* – 1H-TS-Phl-1; *2* – 2H-TS-Phl-1; *3* – 3H-TS-Phl-1; *4* – 6H-TS-Phl-1.

$C_{Pd(II)} = 2.72\cdot10^{-5}$; $C_{Cu(II)} = 5.9\cdot10^{-5}$; $C_{KBr} = 1.02\cdot10^{-4}$ mol/g; $C^{in}_{CO} = 300$ mg/m³; $U = 4.2$ cm/s ; $t = 20$ °C

Table

Influence of the effective contact time of the gas-air mixture with the catalyst and the pH of the suspension on the degree of CO conversion in the presence of catalysts K_2PdCl_4-$Cu(NO_3)_2$-KBr/$\bar{X}$H-TS-Phl-1.*

Carrier	h, (cm)	τ_{ef}, s	pH_s	$C^f_{CO\ st}$, (mg/m³)	η_{st}, (%)
1H-TS-Phl-1	4.0	0.95	6.23	95	68
2H-TS-Phl -1	4.0		6.09	42	86
3H-TS-Phl-1	4.0		5.89	6	98
6H-TS-Phl-1	6.0	1.43	5.68	6	98

**Conditions: $C_{Pd(II)} = 2.72\cdot10^{-5}$; $C_{Cu(II)} = 5.9\cdot10^{-5}$; $C_{KBr} = 1.02\cdot10^{-4}$ mol/g; $C^{in}_{CO} = 300$ mg/m³; U = 4.2 cm/s ; t = 20 °C).*

Protolytic properties

The condition $\Delta pH_s > 0$ is satisfied for all samples of phlogopite, which indicates the progress of the reaction: $E - OH + HOH \leftrightarrow E - OH_2^+ + OH^-$, which is accompanied by an increase in the pH of the suspension.

Table

Surface acidity characteristics of samples of thermally expanded and modified forms of phlogopite.

№	Sample	pH_0	pH_{st}	ΔpH_s
1	TS-Phl	6.05	6.37	0.32
2	1H-TS-Phl-1	6.03	6.23	0.20
3	2H-TS-Phl-1	5.91	6.09	0.18
4	3H-TS-Phl-1	5.73	5.89	0.16
5	6H-TS-Phl-1	5.44	5.68	0.24