Highly Efficient RGO-supported Pd catalyst for Low Temperature Toluene Oxidation

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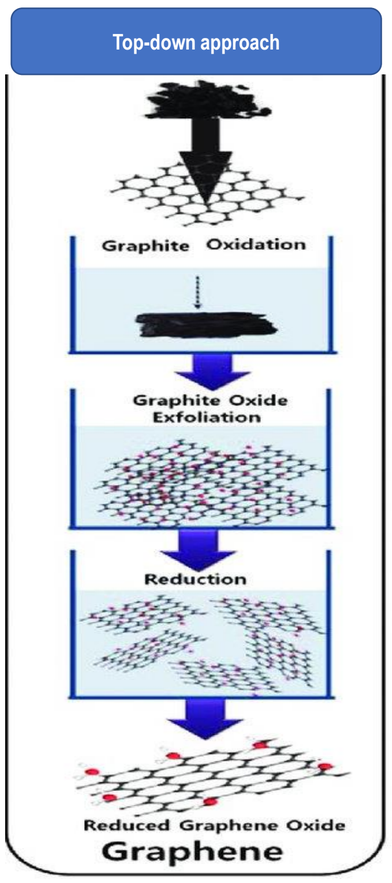
Introduction

Oxygen-containing volatile organic compounds are among the main air pollutants, which are precursors to the formation of ozone in the ground layer of the atmosphere and, along with fine dust particles, are the main cause of the formation of photochemical smog. Among the technologies applied for their elimination, the catalytic oxidation to CO_2 and H_2O has been considered the most beneficial and efficient. Supported noble metals are considered as promising catalysts for total oxidation at low temperatures. The use of Pd in these reactions is due to its thermal stability, low cost and high activity. Inorganic supports for catalysts, widely studied in oxidation of VOCs, However, the use of inorganic supports contribute to deactivation in presence of water vapors. The application of *carbonaceous hydrophobic supports* will sustain the complete oxidation of VOCs at lower temperatures in wet media. The hypothesis is that the hydrophobic surface of the support hinders the adsorption of the water vapor thus preserving the catalyst from deactivation and simultaneously facilitates the adsorption of organic molecules on the catalyst surface. *The aim of this work is the preparation and characterization of Pd catalysts supported on reduced graphene oxide (RGO) with high low-temperature activity and water resistance for the oxidation of toluene.*

Preparation

- The reduced graphene oxide (RGO) was prepared from graphene oxide (GO) obtained by modified Hummers' method.
- After reduction of GO with ascorbic acid, the produced RGO was dried and Pd was loaded on it by incipient wetness method.
- The resulting material was applied on the surface of mullite particles in a form of:
  - fine powder with alumina-hydrogel as a binder (Pd/RGO-surf)
  - mixture paste of alumina-hydrogel and RGO (Pd/RGO-bulk).



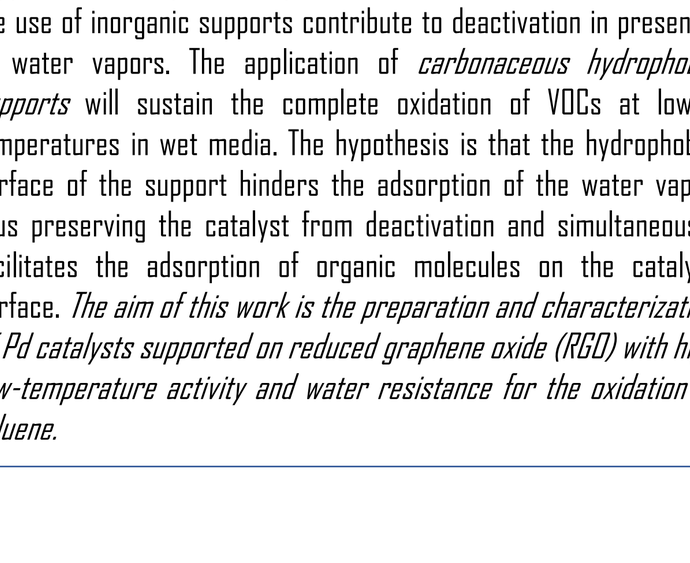
Experimental

The **Pd/RGO** catalyst was characterized by

- powder XRD,
- SEM-EDS,
- XPS
- N_2-physisorption.

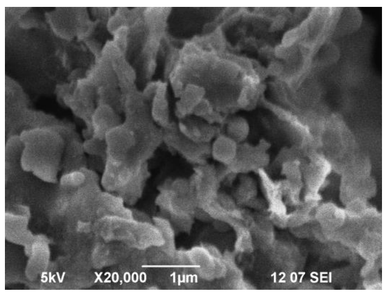
The tests on complete catalytic oxidation of toluene were carried out in a fixed bed reactor with GHSV of 30 000 h^{-1} and inlet concentrations of the reagents:
75- 450 ppm toluene; 1- 16 vol. % O_2, 0– 1.8 vol. % water vapor in nitrogen.

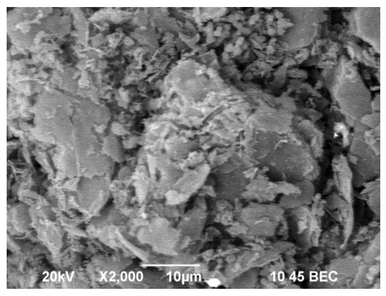
XRD

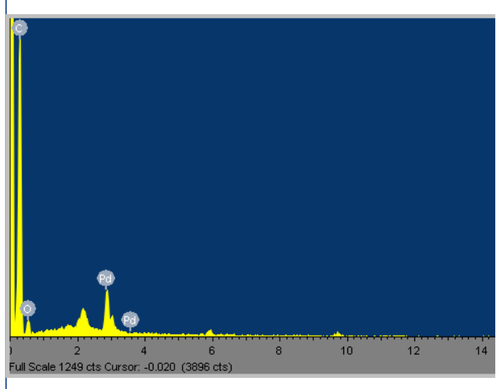


The analyses of XRD patterns reveal the successful loading of Pd on the RGO support forming mainly PdO with traces of metallic Pd.

SEM

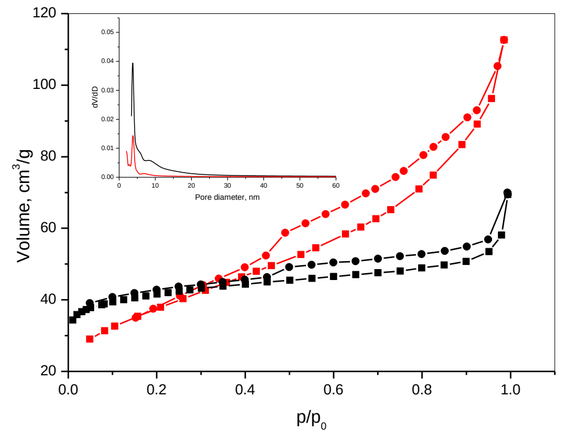






SEM images confirm the homogeneous distribution of Pd particles on the surface of RGO. .

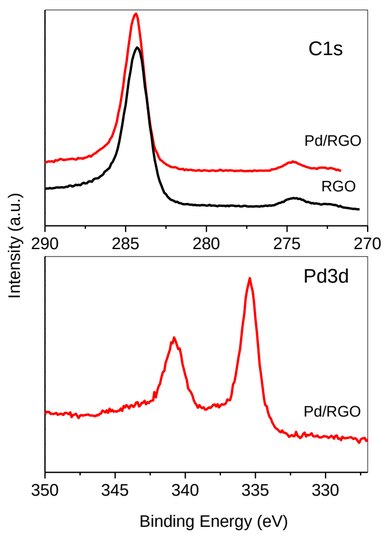
N_2 adsorption



Texture parameters

Sample	S, m^2/g	V, cm^3/g	Dav, nm	Smi, m^2/g	Sext, m^2/g	Vmi, cm^3/g
RGO	158	0.11	2.7	123	35	0.05
Pd/RGO	134	0.17	5.2	28	106	0.012

XPS

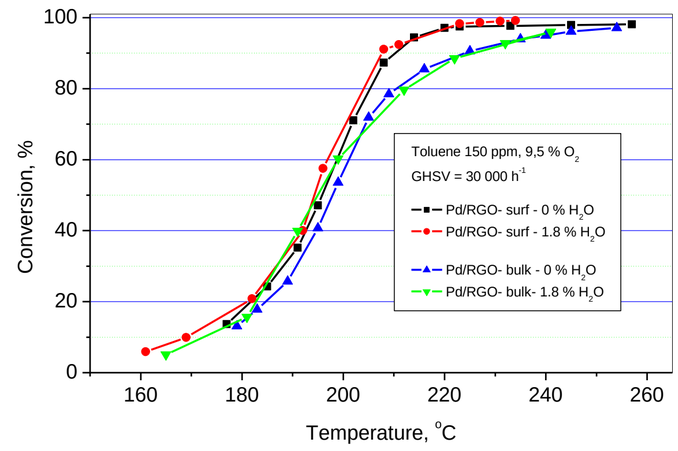


The analysis of XPS spectra confirms that the oxidation state of Pd on the surface of the RGO support is mainly Pd^{2+} with traces of Pd^0.

Kinetics parameters

Model PWL	Catalyst	k_o	E_a	m (TOL)	n (O_2)	p (H_2O)
	Pd/RGOs	$5.07.10^{10}$	117.2	-0.2	0.7	0.0
	Pd/RGOb	$5.05.10^{10}$	117.0	-0.2	0.6	0.0

Catalytic studies



- **High activity at low temperatures**
- **No deactivation in water vapor presence**

Conclusions

A highly active catalyst for toluene combustion has been successfully prepared using composite active phase of Pd-nanoparticles with RGO loaded on mullite support. Due to the specific interaction between the composite's constituents the new formulation of the active phase permits an effective operation of the catalyst in low temperature toluene oxidation. The results obtained lead to further perspective for application of the currently developed approach for preparation of large scale monolithic catalytic systems for air pollution control.

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