

Effect of humidity on the photocatalytic degradation of gaseous hydrocarbons mixture



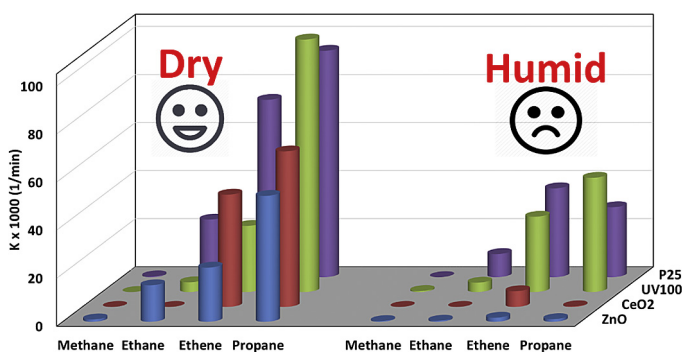
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HIGHLIGHTS

- The photocatalytic degradation of gaseous mixture contains five C1-C3 hydrocarbons was investigated.
- The study was performed over four commercial photocatalysts under dry and humid condition.
- TiO₂(P25) was the most active photocatalyst in the dry conditions, then ZnO.
- Hombikat TiO₂(UV100) was the most resistant photocatalyst against humidity deactivation.
- CeO₂ and ZnO were the most negatively influenced catalysts by humidity.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of humidity on the photocatalytic performance of four semiconductors was investigated in the degradation of gaseous mixture contains five light C1-C3 hydrocarbons. The photocatalytic study was performed with/without 40000 ppm of water vapour at 40 °C in order to simulate the real operating conditions of the hot climate areas with high humidity level. In dry environment, TiO₂(P25) and ZnO exhibited the highest photocatalytic activity. However, in the presence of water vapour, the investigated photocatalysts were deactivated with different extend. ZnO and CeO₂ were the most negatively influenced materials by humidity, while, TiO₂(UV100) was the most resistant photocatalyst against the deactivation.

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1. Introduction

As a result of increasing industrial activities, huge amounts of gases, liquids, and solids waste are realising every day in air, water streams, and soil which affect negatively on the environment [1,2].

Volatile short chain hydrocarbons (C1-C3) are releasing in air as a result of combustion processes such as of natural gas combustion in gas power plants and fuels combustion in vehicles [3,4]. Short chain hydrocarbons consider serious air pollutants because they have a potential contribution in the global warming [5] and create several problems on human health [6].

Photocatalysis is an interesting alternative for the conventional methods which are used in indoor and outdoor air purification such

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as air filters [7,8]. Air decontamination by photocatalysis has several advantages such as it can be carried out under ambient conditions (i.e. room temperature and atmospheric pressure), it can utilize solar radiation and oxygen or water vapour from air as an oxidant, and more importantly, it is able to mineralize a wide range of organic and inorganic contaminants into harmless products such as CO₂ and H₂O [9]. Hence, the addition of photocatalysts to construction materials (i.e. paints [10], concretes [11], gypsum [12], and cements [13]) is a point of interest for academia and industry.

The use of photocatalytic paints to purify air has been investigated. Tang and co-workers [10] investigated the use of TiO₂ nanoparticles supported on Latex paints in the degradation of ammonia. Capucci and co-workers [14] reported the use of TiO₂ Pigmentary to reduce NO_x level around the coated area. In another study, Maggos and co-workers reported the NO_x depollution in a closed parking area and he observed 20% reduction in NO and NO₂ levels [15]. Águia and co-workers studied the addition of several types of TiO₂ to vinyl exterior paint [16,17], the photocatalytic activity of the prepared paints was evaluated in the NO_x oxidation. Moreover, the disinfection of air by using photocatalytic paints was also investigated [18], for example, the paints contain ZnO exhibited high potential in microorganism inactivation [19]. Hence, the photocatalytic paints show promising results in the field of air purification, either in the photocatalytic degradation of contaminants or in disinfection. However, air purification by photocatalytic paints is facing several challenges [20,21] such as catalyst deactivation [22], change the paint colour [23], formation of undesirable toxic by-products [24,25], and paint chalking [26]. So, further investigations are required to improve the photocatalysts performance of the paints.

Humidity is an important deactivation factor of photocatalytic paints [27]. Many countries are suffering from high humidity level such as Saudi Arabia, Egypt, Thailand, Mexico, and some cities in USA. In those countries, temperature can easily reach 40 °C, if the humidity reaches 53%, this means 41'000 ppm of water vapour molecules are present in the air. Therefore, in order to improve the performance of the photocatalytic paints, the effect of humidity on the photocatalytic process should be carefully investigated. In the current study, the photocatalytic oxidation of five short chain hydrocarbons present in a mixture with equal amounts, was investigated over four different commercially available photocatalysts (TiO₂(P25), TiO₂(UV100), ZnO, and CeO₂) under dry and humid conditions. The study was carried out in order to establish the most efficient catalyst(s) which might be dedicated to paints industry [20] to be used in indoor and outdoor coatings at high humid areas.

2. Materials and methods

Hombikat TiO₂ (UV100) was obtained from Sachtleben[®], Germany. TiO₂ (P25) was obtained from Evonic[®], Germany. CeO₂ and ZnO were purchased from Sigma Aldrich. All the obtained materials were used directly without any further treatment/modification. A standard gas mixture of 1 vol% of CH₄, C₂H₄, C₂H₆, C₃H₆, and C₃H₈ in Helium was used as reactants.

XRD was performed by using Shimadzu 6000 DX instrument diffractometer equipped with a graphite monochromator using CuK_α radiation ($\lambda = 0.1541$ nm). The diffuse reflectance spectra were converted into Kubelka-Munk function F(R) by using equation $F(R) = (1 - R)^2/2R$. Moreover, the bandgap of the prepared composites was calculated from equation $E = h \times C/\lambda$ where h is Planck's constant (6.626×10^{-34} J/s), C is the speed of light (3.0×10^8 m/s), and λ is the cutoff wavelength (nanometers). Nitrogen adsorption/desorption isotherms were recorded on a QuantaChrome NOVA 2000e instrument. DR UV–Vis spectra were collected at ambient conditions on a CaryWin 300 spectrometer in the wavelength range

of 200–800 nm by using BaSO₄ as a reference material. Scanning electron microscopy (SEM) Jeol Model 6360 LVSEM, USA, was used to observe the pore structure of the synthesized sorbent materials.

The photocatalytic oxidation of the hydrocarbons mixture was performed in a multi-cell home-made photocatalytic set-up as described earlier [28]. The set-up consists of 12 identical cylindrical reactors, the inner volume of each is 50 mL and the radius of the base is 1.5 cm which gives a total base area of 7 cm². In a real reaction, 150 mg of the photocatalyst was spread in the bottom of the reactor to make a uniform film with a thickness of 1–2 mm. The reactors were evacuated down to 3 mbar and then a He stream containing 25 ppm of each component was introduced into the reactors over the catalyst films. Evacuation/filling cycle was also repeated at least three times before lamp ignition and starting the oxidation experiment. The effect of water was studied by introducing 4 vol% of water vapour (i.e. 40'000 ppm) with the gas feed into the reactors. The applied light source is a 120W high-pressure mercury lamp with a spectrum ranging from 280 to 650 nm. All the reactors were operated in batch mode and illuminated for 135 min. The concentration of hydrocarbons was monitored by a compact gas chromatograph equipped with TCD and FID detectors with an accuracy of ± 0.5 ppm. Molsieve 5A (5 m) and a capillary Porabond Q column (10 m) were connected to the TCD detector while Porabond Q column (10 m) was coupled to an FID detector. The photoactivity profiles were fitted assuming first order kinetics: $\ln(C/C_0) = -kt$ where C is the concentration of the hydrocarbons at time t , C_0 is the initial concentration, and k is the observed rate constant.

The activity change due to humidity was calculated from the following equation:

$$\text{Activity change \%} = \frac{C_d - C_h}{C_d} \times 100$$

where C_d is the converted hydrocarbons in dry air (ppm), and C_h is the converted hydrocarbons in humid air (ppm).

All the experiments were carried out at least for three times, the results presented here is the average of the three experiments with a standard deviation does not exceed than 3%.

3. Results

Blank experiments were carried out to confirm the synergy between light and the applied catalysts in the oxidation of the gas mixture. In the first experiment, the gas mixture was introduced into the reactor cells as described in the experimental section but without catalyst. No change in the concentration of the gases was observed either in dark or under light illumination for 135 min. This is an indication for the non-leaking property for the used reactors and the high stability of the investigated gases against photolysis, respectively. In the second control reaction, the gas mixture was introduced into the reactor cells which contained the different catalysts. However, the reactions were carried out in the dark with/without water vapour. Again, no change in the gases concentration was observed as an indication for the stability of the gas against catalytic decomposition at room temperature in the absence/presence of water vapour.

The change in the gases concentration was obtained only in the presence of light illumination and the catalytic materials, as an indication for the true photocatalytic reaction of the investigated gases. In the following section, the observed concentration change of the mixture components (C1–C3 hydrocarbons) is presented for each gas individually in three cases: i) dark-dry, ii) light-dry, and iii) light-wet conditions.

Fig. 1 shows the concentration changes of C1–C3 mixture over

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