



Development of a stagnation point flow system to screen and test TiO₂-based photocatalysts in air purification applications



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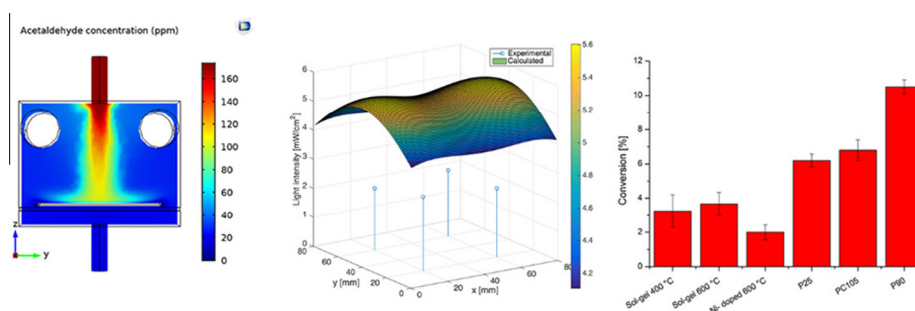
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HIGHLIGHTS

- A new photocatalytic reactor, based on the stagnation point geometry, was designed.
- The reactor geometry minimizes the bulk-surface mass transfer limitations.
- The reactor is carefully designed to homogenize the UV irradiation on the catalyst.
- Metallic plates were pre-treated and used as photocatalyst support.
- Various photocatalysts were screened, confirming the suitability of the system.

GRAPHICAL ABSTRACT



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ABSTRACT

An innovative system suitable for the abatement of VOCs (Volatile Organic Compounds), using photocatalysis under UV light, was designed and built. The design of the reactor is based on the stagnation point flow geometry and the fluid dynamics of the system was carefully investigated in order to avoid mass transfer limitations. The proportions of the elements in the reactor were adjusted in order to homogenize the UV irradiation on the catalyst surface.

The supports used for the coating of the catalysts were aluminum plates in order to accurately reproduce industrial conditions. After each test, the catalytic plate was examined to evaluate the mechanical strength of the bonding between the catalyst powder and the metallic support. The coating proved to be sufficiently stable for tests in the designed set up.

The potential scale-up of the features of the system was considered throughout the design and especially the power of the UV lamps was decided in order to be representative of the industrial cases.

In order to evaluate the suitability of the system for catalysis investigations, various photocatalysts, both synthesized and commercial, were screened. Analyzing the activity results, using acetyl aldehyde as a model VOC, it was possible to evaluate clear differences between the samples and P90 proved to be the most active sample.

All the aspects investigated in this work demonstrate that the design of the reactor is in accordance with the expectations and that the system is suitable for screening and testing of photocatalysts for VOCs removal applications.

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1. Introduction

Many industrial processes involve complex steps of matter transformation, often in presence of solvents, which emit a considerable amount of VOCs (Volatile Organic Compounds) [1,2]. In some cases, the VOCs are aromatic compounds, toxic at very low concentrations [3]. However, toxicity is not the only concern related to VOCs, which may also react with NO, resulting in the formation of tropospheric ozone and smog [4–6]. Odor is another issue related to VOCs emission, since many VOCs have a very pungent odor even at very low concentrations. VOCs emitted from frying industries are typical examples of odorous compounds [7], very often disturbing the surroundings. The attention to this problem has increased in the last few years due to stricter regulations, but also due to a higher awareness of the citizens in the society, resulting in an increased number of complaints to the local authorities [8]. Technologies often used to deal with this problem is thermal or catalytic incineration. Both technologies are rather inefficient from an energy point of view. A catalytic incinerator needs to heat up the whole gas stream between 200 °C and 500 °C to remove a few ppm of VOCs [9]. In case of a thermal incinerator, the situation is even worse, due to the higher temperature, between 700 °C and 1100 °C, needed [9]. Additionally, incinerators requires a large amount of fuel, often fossil [10], resulting in large emissions of fossil-derived CO₂. An attractive solution, to substitute environmentally unfavorable solutions, is the application of methods, based on UV light, inducing oxidation of VOCs at ambient temperatures, thus avoiding the use of fossil fuels.

The working mechanism of UV technology is the combination of the UV irradiation with oxidants to produce hydroxyl radicals, which attacks the organic compounds. An oxidant commonly used is ozone (e.g., [11–17]), easily absorbing the light, emitted by UVC lamps [18], stimulating the formation of hydroxyl radicals in the presence of water. Nevertheless, ozone is a toxic compound and has itself a very pungent odor [19,20]. It is therefore necessary to avoid any ozone slip after the UV step. A photocatalytic step in a process may eliminate the need for ozone addition, since reactive species are produced [21] without involving the use of toxic oxidants.

Previous works [22–24] report on investigations of the potential of applying the photocatalytic technology for VOCs abatement. However, the experimental conditions are, in many cases, not representative of industrial conditions. Often, the power of the UV lamps is too high, considering the gas flow rate treated. Indeed, to be economically feasible for industrial applications, a photocatalytic system needs to reduce the ratio between the UV power and the gas flow rate, as much as possible: realistic values are below 50 W/(m³/h). However, several works [14,25–27] are far from these conditions, having values ranging from 125 to 1600 W/(m³/h). Apart from the UV power, the same works may also have features in the UV reactor difficult to scale-up. In other cases, the UV irradiation is not uniform over the catalyst. One example is the work of Hossain et al. [28], where the photocatalyst is coated on monoliths with the openings placed radially to the UV lamps. This configuration does not allow a homogeneous irradiation of the catalyst surface and it also makes it difficult to model the reactor.

Likewise, the configuration proposed by Mehrvar et al. [29], including a packed bed surrounded by 6 UV lamps, does not allow for a homogenous irradiation. Indeed, the central part of the bed receives a lower irradiation than the outer part. Larson et al. [30] and Preis et al. [31] used a similar configuration, with 6 UV lamps, but, in this case, the packed-bed was substituted by an annular reactor coated by thin films of TiO₂. Two concentric cylinders with a slightly different diameter formed an annular volume for the gas

to flow with both the inner and outer walls coated with the catalyst. However, also in this case, the irradiation on the catalyst is not homogeneous, especially considering the outer catalyst film.

Other research groups [32–34] suggested the use of a fluidized-bed reactor. However, this reactor configuration is very sensitive to the gas phase moisture content. The work of Ciborowski and Włodarski [35] displayed that the addition of moisture increases the rate of charge dissipation, Guardiola et al. [36] expanded this investigation, defining different humidity regimes. They defined a humidity threshold value, RH_T, observing a very thin liquid layer around the particles for moisture contents above RH_T. The mutual cohesion of the particles is strengthened so the fluidization becomes impossible, making this configuration difficult to scale-up [37].

In the work by Mo et al. [38] and Fujimoto et al. [39], a simple annular geometry and a cylindrical shape for the UV reactor, suitable for a quick scale-up, was proposed. The whole inner walls were coated with the catalyst implying an expensive and time-consuming system for screening of catalysts, due to the needs for either careful removal of the previous catalyst or replacement of the reactor.

The use of optical fibers to have both the UV light source and the reaction structure in one single entity was also investigated [40,41]. This is an excellent technique to add controlled doses of the UV light through the fiber. Choi et al. [40] investigated the influence on the VOC conversion in relation to the thickness of the TiO₂ layer and the fiber length. Even though the technique is elegant, the limitation in length of the fiber and their cost makes this solution too difficult and expensive to scale-up to industrial scale.

The aim of this work is to develop a test system for screening and testing of photocatalysts, reproducing industrial conditions. In particular, the ratio between the UV power used and the gas flow rate should be much lower compared to previous studies [14,25–27] and below 50 W/(m³/h), in order to achieve a relevant power consumption of relevance for full-scale applications. The fluid dynamics of the system was carefully investigated and optimized, minimizing the mass transfer limitations. Additionally, the photocatalyst should be homogeneously irradiated. The reactor geometry chosen for assessing the mentioned features is the stagnation point flow geometry. This type of geometry has previously been investigated in reaction modeling [42–47]. In particular, the hydrogen oxidation on platinum could be easily modelled, thanks to low number of intermediates and the predictability of the surface velocity. However, the use of this geometry in photocatalytic applications has not been reported so far, to the best of our knowledge.

Other features of the system should be a quick and easy replacement of the tested catalysts as well as the use of metallic support for the powder catalyst coatings, since these supports may be easily included in a full-scale reactor. The investigation of the mechanical strength and stability of the catalyst coating on the support is another important aspect considered.

Food industry, especially fried potatoes, was chosen as reference for the investigation due to the presence of strong odorants at trace concentrations [7,48]. Acetaldehyde was selected as the model compound, since aldehydes are a type of VOC very often found in the emissions from frying industries and most of them have a very low odor threshold [7,48].

Experimental tests with different photocatalysts were performed in order to evaluate the suitability of the test system for photocatalytic abatement of VOCs with the features mentioned beforehand. The photocatalysts investigated were all TiO₂-based, both synthesized and commercial, with nickel as dopant. Ni was selected due to its ability to reduce the recombination rate of sur-

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