

ORIGINAL PAPER

Cost-effectiveness analysis to assess commercial TiO₂ photocatalysts for acetaldehyde degradation in air

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In the commercialisation of photocatalytic air purifiers, the performance as well as the cost of the catalytic material plays an important role. Where most comparative studies only regard the photocatalytic activity as a decisive parameter, in this study both activity and cost are taken into account. Using a cost-effectiveness analysis, six different commercially available TiO₂-based catalysts are evaluated in terms of their activities in photocatalytic degradation of acetaldehyde as a model reaction for indoor air purification.

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Introduction

In recent years, poor Indoor Air Quality (IAQ) has become a major topic. There are three general methods for addressing the problem: *a*) controlling the source, *b*) increasing (natural) ventilation and *c*) pollutant abatement (Zhang et al., 2003; Birnie et al., 2006; Bennett, 2009). Current advances in building management significantly impede IAQ improvement by the first two methods (Bennett, 2009), hence the option of pollution abatement is receiving more attention. Several technologies are available and already operational to this end, such as filter systems (Batterman et al., 2005; Bekö et al., 2008; Zhang et al., 2011), ozone generators (Kwong et al., 2008; Zhang et al., 2011) and plasma catalytic systems (Van Durme et al., 2007; Van Wesenbeeck et al., 2013).

Besides the above techniques, photocatalysis could be a “green” and viable technological alternative for

indoor air purification (Mo et al., 2009; Yu et al., 2011). A good photocatalyst should meet the following criteria: be chemically and biologically inert, stable, efficient in catalysing the reaction and relatively inexpensive (Carp et al., 2004; Fujishima & Zhang, 2006). Taking all of the above into consideration, TiO₂ is frequently the photocatalyst of choice (Carp et al., 2004). Of all TiO₂-based materials available, the most frequently used is Aeroxide[®] P25; this has become established as a reference in comparative research studies (Ohtani et al., 2010; Saha et al., 2012). These comparisons are almost always based on degradation efficiency (Doudrick et al., 2012; Verbruggen et al., 2012). While this is a crucial aspect, the associated costs will ultimately determine the possible use in real-life applications.

This study investigated the relation between photocatalytic activity and the associated catalyst cost using a cost-effectiveness analysis (CEA). CEA can be

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an alternative decision tool in assessing the suitability of different photocatalysts. Six different, commercially available photocatalysts were compared in terms of degradation of acetaldehyde. Like formaldehyde, acetaldehyde is an important indoor air pollutant and known as one of the possible causes of sick building syndrome, resulting in headaches, irritation and fatigue (Xu & Shiraiishi, 1999). Hence it is a relevant contaminant for gas phase photocatalytic remediation studies (Sopyan et al., 1996; Sopyan, 2007; Bianchi et al., 2014).

Theoretical

This research sought to compare the photocatalytic activities of different catalysts for use in indoor air purification. Considering this industrially relevant issue, the photocatalytic activity of different commercially available TiO₂ catalysts is assessed, taking their price into account. A traditional cost-benefit analysis is not feasible in this case, since it is difficult to express the benefits (i.e. the photocatalytic activity) in monetary terms. Cost-effectiveness analysis (CEA) provides an alternative method in which the benefits are expressed as a quantifiable measure (Boardman et al., 2006). CEA is often used to evaluate projects dealing with environmental (Hansen et al., 1998; Compernelle et al., 2012) or health issues (Black, 1990; Briggs & Fenn, 1998; Löthgren & Zethraeus, 2000). In the present experiments, the effectiveness is represented by the turnover frequency (TOF):

$$\text{TOF} = \frac{\text{Moles of acetaldehyde degraded}}{\text{Moles of catalyst} \times \text{Time}} \quad (1)$$

The TOF is deliberately expressed per moles of catalyst (and not per unit area) since this is directly linked to the mass of the catalyst which, in turn, is readily linked to its cost. With regard to costs, only the material cost is taken into account. The costs involved in the coating procedure are negligible and comparable for all samples.

Two types of ratio are used to quantify cost-effectiveness. The first is the average cost-effectiveness ratio (ACER), representing the cost associated with a certain effect (Eq. 2). The alternative with the lowest ACER is preferred for an obvious reason.

$$\text{ACER} = \frac{\text{Cost of catalyst in reactor}}{\text{TOF}} \quad (2)$$

The second ratio is the incremental cost-effectiveness ratio (ICER), corresponding to the additional cost associated with the effect increase (Eq. 3). P25 was always taken as the reference.

$$\text{ICER} = \frac{\Delta \text{Cost}}{\Delta \text{Effect}} = \frac{\text{Cost}_{\text{alternative}} - \text{Cost}_{\text{P25}}}{\text{TOF}_{\text{alternative}} - \text{TOF}_{\text{P25}}} \quad (3)$$

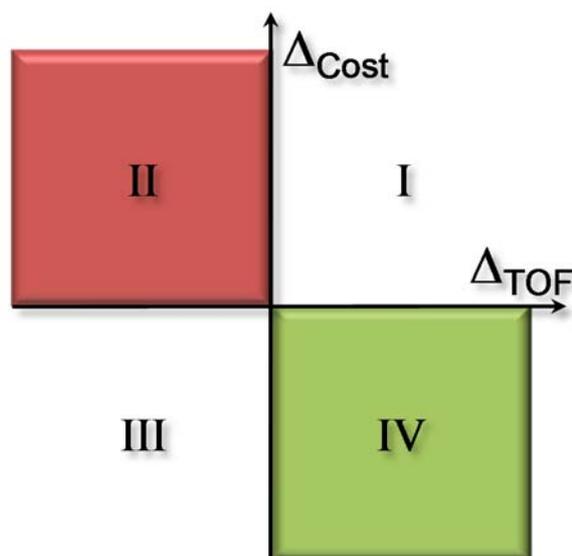


Fig. 1. Cost-effectiveness (CE) plane used in this work.

The concept of ICER is better understood when plotting the incremental costs and effects in a CE plane (Fig. 1). In this plane, the reference (P25) is located at the origin. Every single point in the plane represents an alternative catalyst. Alternatives located in quadrant IV are always preferred as they combine higher activity with lower cost when compared to the reference. In contrast, alternatives located in quadrant II should be discarded (less active at higher cost). The suitability of catalysts in quadrants I (more active but also more expensive) or III (less active but also less expensive) for a certain application depends on the technological and economic specifications of the project.

Experimental

All products were used as received without further purification. Prices in the Belgian market were obtained from the manufacturer's websites or through personal communication with the company. Six TiO₂-based catalysts were selected with quite diverse properties and obtained from several sellers: Aeroxide[®] P25 (Acros Organics, Belgium), Aeroxide[®] PF2, Aerolyst[®] 7710, Aeroxide[®] P90 (Evonik, Germany), PC500 (Cristal Global, France), Hombikat (Sigma-Aldrich, Germany).

The catalyst-coating procedure, photocatalytic test protocol and procedure are discussed in detail elsewhere (Jammaer et al., 2011; Verbruggen et al., 2011, 2012; Tytgat et al., 2012). In summary, the catalysts were immobilised on 108 g of 2 mm glass beads (Assistent, Belgium, no. 1401/2) by suspension coating. Only the Aerolyst[®] sample was first ground in an agate mortar prior to suspending in ethanol. The coated beads were dried at 338 K overnight. The photocatalytic reactor consisted of a glass cylinder with

Table 1. Product specifications according to product sheets or manufacture data

Catalyst	Pricing ^a /(€ kg ⁻¹)	BET area/(m ² g ⁻¹)	Contains rutile	Other remarks
P25	120	50 ± 15	Yes	–
PF2	78	60 ± 15	Yes	2 mass % Fe(III) oxide
Aerolyst	500	50 ± 10	Yes	pelletised
P90	25.5	90 ± 20	Yes	–
PC500	25	350	No	15–20 mass % not crystalline
Hombikat	277	50 ± 15	Yes	–

a) Price excl. VAT. With a view to industrial applications, the lowest price for the largest available quantity was used (Retrieved on 2013).

the lamp (Philips Cleo, Germany, 25 W, 205 μW cm⁻² at λ_{max} = 365 nm) placed longitudinally at the centre. The coated beads were loaded in the annular space between the lamp and reactor wall (3 mm wide). A continuous air-flow (Air Liquide Alphagaz, Belgium) spiked with 175 μL L⁻¹ of acetaldehyde (Air Liquide, Germany, 1 vol. % in N₂) was directed through the reactor at a total flow-rate of 2 L min⁻¹. These conditions correspond to a reactor void fraction of 47 %, a mean gas residence time of 1 s and a gas contact time of 57 g h mol⁻¹. The variation in pollutant and reaction product concentrations in the reactor outlet was monitored in time and on-line using FTIR spectroscopy (Nicolet TM 380, Thermo Fisher Scientific, USA), with MacrosBasic software installed. The FTIR apparatus was equipped with ZnSe windows and a 2 m heated gas cell for higher resolution (detection limit acetaldehyde ca 4.5 μL L⁻¹). The FTIR data were converted to concentrations using calibration curves constructed with a calibrated organic vapour sensor (DrägerPolytron, Germany). The contaminated air-flow passed through the reactor in darkness for 20 min to establish adsorption equilibrium, after which the lamp was switched on and photocatalytic degradation took place. The difference in the stable reactor levels of acetaldehyde in the darkness and under UV exposure corresponded to the amount of pollutant that was steadily and continuously degraded photocatalytically which can, in turn, be used to calculate the TOF (Eq. 1).

Results and discussion

This study sought to illustrate how CEA can be used as an alternative decision tool for evaluating the suitability of different photocatalysts; however, a detailed description of the structure-activity relation of all catalysts is not given here. For completeness, Table 1 summarises the specifications of the materials used, as stated on the product sheets. Four out of six catalysts are characterised by a specific surface area of approximately 50 m² g⁻¹, including the P25 reference catalyst. P90 exhibits a larger surface area of 90 m² g⁻¹ and that of PC500 is substantially higher, 350 m² g⁻¹. The high specific surface area of PC500 is due largely to the presence of very small particles

(primary particle diameter of ca 10 nm) and micro-porosity. Apart from PC500, all the photocatalysts consist of anatase and a fraction of rutile. It has previously been reported that a modest fraction of rutile contributes to higher photocatalytic activities (Su et al., 2011), probably due to improved charge separation and structural effects. PF2 is a P25-based material enriched with iron oxide to afford higher thermal stability in certain applications.

Fig. 2a shows a typical acetaldehyde concentration time profile in a photocatalytic experiment. In the first 20 min of the measurement, the polluted air-flow is directed through the reactor in darkness and a stable concentration level is established. Then (at minute 20) the UV lamp is switched on for one hour. The steep decline in the acetaldehyde concentration is attributed to photocatalytic degradation. CO₂ and H₂O were detected in the outlet gas stream as mineralisation products. No intermediate gaseous species could be observed in detectable amounts. Blank experiments have proved that photolysis and heat effects do not contribute to the acetaldehyde decrease (Verbruggen et al., 2012). After one hour of illumination, the lamp is switched off again (at minute 80) and the acetaldehyde level in the continuous gas stream is quickly restored to its original level in darkness. Fig. 2b shows the total acetaldehyde conversion, calculated as the difference in stable levels in darkness and under UV, as a function of the total P25 content in the reactor. With ca 100 mg P25 in the reactor, the acetaldehyde conversion is almost complete and saturates at higher reactor loadings. This is not an appropriate starting point for comparative tests, as it is preferable to operate at sub-optimal values. Accordingly, 50 mg of photocatalyst was selected as a suitable sample amount. Throughout the remainder of this study, the amount of test sample for all catalysts was kept close to 50 mg, albeit some fluctuations in catalyst-loading were observed because the coating procedure cannot be perfectly controlled. The actual measured coated amounts of catalyst loaded into the reactor are given in Table 2, together with the associated costs and TOFs for steady-state degradation of the acetaldehyde in air.

CEA was used to interpret the results. Fig. 3a plots the ACER (Eq. 2) of all the materials tested. It is clear

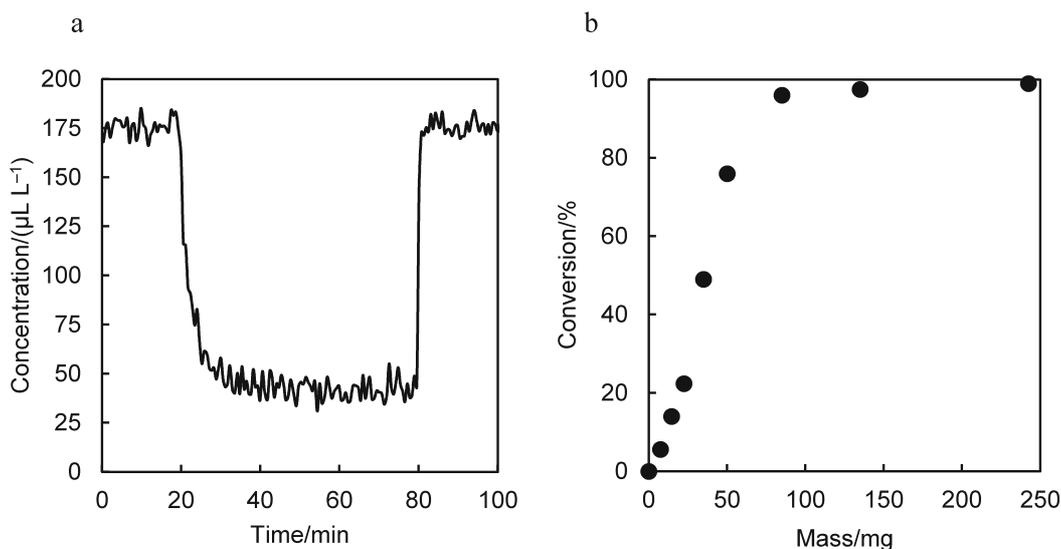


Fig. 2. Time profile of acetaldehyde concentration during photocatalytic test (50.1 mg of P25 in reactor) (a). Acetaldehyde conversion as function of P25 content in reactor (b).

Table 2. Experimental parameters and results

Catalyst	Catalyst mass reactor/mg	Cost/€	TOF/h ⁻¹
P25	50	6.00	0.54
PF2	61	4.76	0.02
Aerolyst	49	24.50	0.37
P90	65	1.66	0.53
PC500	50	1.25	0.70
Hombikat	59	16.35	0.46

that PF2 and Aerolyst should not be selected as the ACERs are much larger than those of the other catalysts. In the case of Aerolyst, this is largely due to the higher cost (Table 1), whereas for PF2 the high ACER arises mainly from poor activity (Table 2). CEA analysis based on the ICER is better understood from the CE plane in Fig. 3b. As explained in the theoretical section, catalysts located in quadrant II of the CE plane are not of interest. As with the ACER, the CE plane indicates that Aerolyst is no suitable alternative to P25. Interestingly, Hombikat is also located in this quadrant, despite its photocatalytic activity (TOF) not being much lower than that of P25 (Table 2). This clearly illustrates the usefulness of CEA: without taking the catalyst pricing into account, Hombikat would probably not have been discarded as an alternative to P25. Concerning P90 and PF2, both located in quadrant III, no conclusive statements can be made without knowing the technological or financial specifications of the final application. However, it is clear that P90 might still be of interest since its activity is only slightly lower than that of P25 but it is also significantly less expensive. By contrast, PF2 is probably of no particular interest, in view of the large activity loss associated with only a minor cost gain. This was already clear from the ACER.

The most important result from this CEA concerns the PC500 catalyst. It is located in the preferred quadrant IV with higher activity and lower cost than the reference. In a previous study, P25 and PC500 were compared on the basis of their property-activity relation (Verbruggen et al., 2012). The main conclusion was that PC500 outperforms P25 due to its high surface area; this appears to be the crucial parameter in gas phase applications where diffusion barriers are low. It is worth noting that CEA leads to the same conclusion, despite being a totally different approach.

Apart from affording a direct comparison of alternatives, CEA can also be used to make insightful market evaluations. For instance, it is possible to determine to what extent the PC500 pricing may increase and still be a viable alternative to P25. Increasing the PC500 cost until the ACER reaches the value of P25 can simulate this issue. The result of this exercise is a PC500 price increase from 25 € kg⁻¹ to 155.1 € kg⁻¹ so that each unit of TOF is equally expensive for both P25 and PC500. Alternatively, the price of P25 should decrease drastically from 120 € kg⁻¹ to 19.3 € kg⁻¹ to reach the cost-effectiveness of PC500. For comparison, the pricing of Evonik's Aerioxide P25 is 38 € kg⁻¹ for bulk quantities, which is double the allowed cost. The CEA thus points to the conclusion that, based on

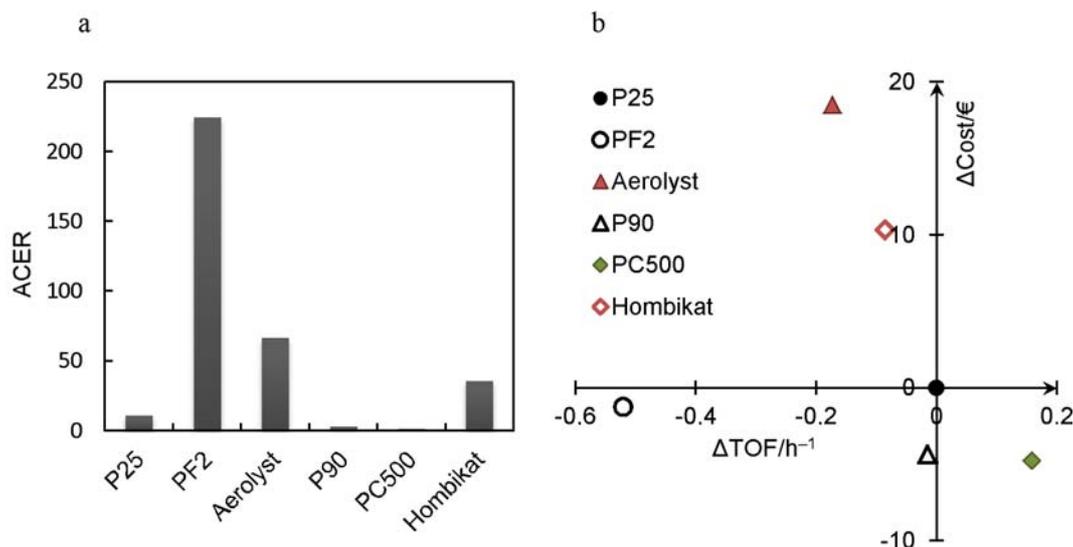


Fig. 3. ACER obtained for six different TiO₂ photocatalysts (a). CE plane constructed with P25 as reference catalyst (located at origin) (b).

the current market and activity data, PC500 is a cost-effective photocatalyst for acetaldehyde degradation. This is confirmed by the low ACER value, as well as its position in quadrant IV, the preferred quadrant of the CE plane.

Conclusions

This study shows that CEA can be used as an alternative evaluation tool to compare different TiO₂-based photocatalysts towards acetaldehyde degradation in air. As a case study, six commercially available catalysts were analysed and compared against P25 as reference. From CEA, it was concluded that PF2 and Aerolyst were of minimal interest, considering their high average cost-effectiveness ratio (ACER). Hombikat is also of little interest from the perspective of CEA, due to its unfavourable position in quadrant II of the CE plane, corresponding to lower TOF and higher cost than the P25 reference. The suitability of P90 for acetaldehyde degradation will depend on the economic and technological project parameters. The low ACER value is favourable, but its position in quadrant III of the CE plane indicates that not just cost but also activity is lower in comparison with P25. PC500 is found to be the most cost-efficient material for acetaldehyde degradation in comparison with P25. This is concluded from the low ACER, as well as the position in quadrant IV, the preferred of the CE plane that is indicative of high turnovers at reduced costs.

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