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Comparison of photocatalytic space-time yields of 12 reactor designs for wastewater treatment

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ABSTRACT

Different designs for photocatalytic wastewater treatment were compared with a new benchmark measure, photocatalytic space-time yield. This benchmark is the ratio of the reactor space-time yield to the standardized lamp power, defined as the volume of water treated for each kW lamp power per volume of reactor per unit of time. This benchmark gives a clearer view of the reactor performance than the current benchmarks such as pseudo first order rate constant or photonic efficiency. Using the benchmark, 14 reactors with 12 different designs were compared. Comparison showed that the photocatalytic membrane reactors scored the highest. It is also shown that with efficient light distribution, the microreactor technology can prove to be the new generation wastewater treatment reactor conditional to effective scale out strategies. The proposed benchmark measure indicates a new direction to the research on photocatalytic wastewater treatment, which is lighting design instead of new geometries.

1. Introduction

The scientific work done on photocatalytic reactors is wealthy and growing exponentially including 13,500 papers, reviews and reference work over the last 38 years. However, photocatalytic reactors are still not implemented in industrial processes [1]. The majority of publications are on wastewater treatment, whereas the number of patents filed for these reactors fall behind the air treatment counterparts [2]. Fig. 1 shows the published papers and filed patents on photocatalytic reactors between 1977 and 2015.

The lack of integration of photocatalytic reactors to industry is partly due to the suboptimal designs available [3]. Several challenges for the design of photocatalytic reactors are inherited from conventional catalytic reactors such as the need to have a large specific surface area and high mass transfer rates. Therefore, various reactor configurations that are applied to conventional catalytic applications have been investigated for photocatalytic applications as well [1,4]. Conventional designs however, are currently being revised using process intensification principles and adapted to photocatalysis. These intensification studies in some cases surpass their usage in photocatalytic water treatment but can also used in photochemical synthesis. [5] The main optimization and intensification works focus on the following criteria [1,4]:

1. Overcoming mass transfer limitations: promoting fast adsorption—desorption, increasing catalyst surface
2. Optimizing photon transfer limitations: optimum lighting strategy and reactor geometry to maximize irradiance
3. Industrial integration: scale-up, catalyst separation, retrofitting to existing systems.

In this work, we perform a comparison of 12 common photocatalytic reactor designs for wastewater treatment. The basis for comparison is a new benchmark measure, being the photocatalytic space-time yield (STY), which essentially reflects the effects of reaction, mass and photon transfer rates and light utilization efficiency on the volumetric specific yield of these reactors.

2. Photocatalytic reactor designs

To overcome the said limitations, a wide variety of designs have been proposed. 10 of the 12 design principles considered in this work are shown in Fig. 2. The other two designs are continuous stirred tank reactor and slurry version of the foam reactor which need no illustration. This work considers 14 reactors based on the 12 design principles. There will also be hybrid designs which combine different design principles in one reactor.

The annular reactor (AR) is the most popular slurry reactor [7] which is essentially a tubular reactor with a single lamp or a lamp array located only at the axis. The AR benchmarked in this work

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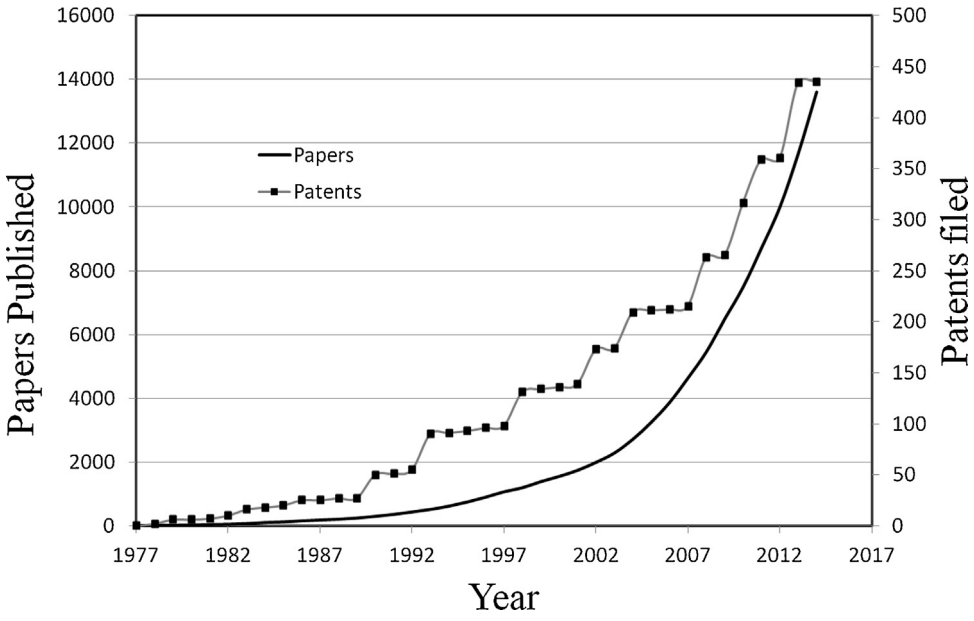

Fig. 1. Cumulative evolution of published papers and filed patents per year with keywords "photocatalytic reactor". Figures on papers are from Science Direct, figures on patents are from Google patent search.

was applied in a phenol degradation study by Chiou et al. [16]. The procedure involves P25 type titania powder and three types of mercury lamps with 20 W low pressure, 100 W and 400 W high pressure powers. The reaction rate constant from [15] considered in the present comparative study corresponds to 0.36 mmol L^{-1} initial concentration, 1 g L^{-1} titania content and 400 W lamp power.

The biggest advantage of AR, and by extension all the slurry reactors (SLR), is the high surface area due to the small catalyst particles. One of the drawbacks of AR is the absence of agitation. Mixing and enhanced illumination of the reaction medium was introduced by more elaborate designs such as multi lamp reactor (MLR) [8], rotating annular reactor (RAR) [7] and spinning disc reactor (SDR) [15]. These designs eliminated concentration gradients and enabled uniform illumination of every catalyst particle.

There are two MLR designs benchmarked in this study, which are both pilot scale reactors to treat polluted river water [17,18]. One of them [18] is a rectangular stirred tank with 36 mercury lamps of 40 W power. The results shown in the benchmark were recorded with 0.4 g L^{-1} titania (P25) content. This MLR is a hybrid design with membrane integration. The membrane is a hollow fibre of 0.4 μm pore size and 1.5 m^2 membrane area. Chemical oxygen demand was monitored to assess the performance. The first order removal rate calculated with batch operation was used in this work.

The second MLR design [17] is a tubular reactor of 11.4 m^3 volume. This reactor is also a membrane integrated design bearing additional catalyst recovery unit consisting of bag filters with nominal pore size of 10 μm. The reactor is equipped with 32 mercury lamps, which can deliver up to 4.24 kWh m^{-3} UV dose when all lamps are lit. The reactor is operated at a constant flowrate of 24 L min^{-1} carrying a P25 type of catalyst of 50 ppm concentration. Even though no first order kinetic data were given for this reactor, various direct STY data were given for various pharmaceutical and pesticide pollutants that are found in the treated river water. For this work, the pollutant with one of the lowest STY (highest energy requirement) was considered (diethyltoluamide—DEET). DEET entered the reactor at 1500 ng L^{-1} and treated to 1 ng L^{-1}, which amounts to more than three orders of magnitude decrease. The electrical current of the lamps is given (70 A three phase) in the reference work's supporting information. The total lamp power of 48 kW was calculated from the Ampere data assuming 400 V line to line voltage.

The RAR is a prototype by Subramanian et al. [7] that was used for phenol removal from synthetic wastewater. This reactor consists of a rotating lamp fixture wall and a static outer wall. This design enables the catalyst particles to undergo a controlled periodic illumination, which gives the catalyst illuminated and dark periods. This way of operation allows (at least in theory) for temporal decoupling of mass transfer (dark phase) and the photocatalytic degradation (illuminated phase) and hence for yield increase. RAR was shown to perform better with agitation. The data used in this work correspond to operation with 8 g L^{-1} catalyst concentration and 120 W of total lamp power.

The SDR is a prototype by Yatmaz et al. [15] which was used for 1-4-chlorophenol removal from wastewater. This design utilizes a spinning disc and pumps the reaction slurry on this disc to create a thin film, which is illuminated evenly by 40 W or 400 W mercury lamps. The data recorded for comparison in the present study correspond to the lower power low-pressure mercury lamps and 10 g L^{-1} P25 titania concentration. This study [14] indicated that the low-pressure lamps are more efficient than the medium pressure lamps, which have a significant portion of visible range light emission. Furthermore, it was indicated that with faster mixing and smaller characteristic thickness of flow, higher catalyst concentrations can be utilized.

The photocatalytic membrane reactor (MEM) design, utilizing a microporous membrane at the reactor outlet to keep the catalyst inside the reactor addresses a drawback of the AR design, which is the catalyst recovery problem. For stirred tank slurry types of membrane reactors, a strategically placed aeration nozzle [18], or membrane placement on the agitator itself, as in Fig. 2e [10], facilitates catalyst redistribution. However, for tubular membrane integrated designs, a catalyst recovery unit apart from the reactor is needed, as in the case of the pilot reactor for river water treatment [17]. The membrane reactor concept was utilized as a hybrid design with the MLR discussed above.

The catalyst recovery problem was also addressed by the immobilized catalyst reactors. Parallel plate reactors (PPL) shifted