



Modelling of succinic acid heterogeneous catalytic ozonation on metallic foam



Aude Audirac^{a,b}, Florence Pontlevoy^b, Nathalie Karpel Vel Leitner^{a,*}

^a University of Poitiers, Institute of Chemistry of Materials and Natural Resources, UMR CNRS 7285, Department of Water, Geochemistry and Health, ENSIP, 1 rue Marcel Doré, 86073 Poitiers Cedex 9, France

^b Technavox, ENSIP, Plate-forme Eaux, Bâtiment B16, 7 rue Marcel Doré, 86073 Poitiers Cedex, France

HIGHLIGHTS

- A metallic foam was used in a catalytic ozonation device.
- The concentration of the model molecule decreased linearly with ozonation time.
- Modelization of the concentration pattern was consistent with the experimental results.
- Above a given concentration of compound, the rate reached a constant value.
- Results suggest catalytic ozonation proceeds through two successive reaction steps.

ARTICLE INFO

Article history:

Received 19 February 2015

Received in revised form 4 May 2015

Accepted 8 May 2015

Available online 15 May 2015

Keywords:

Ozone

Solid foam catalyst

Modelling

ABSTRACT

This work aims at envisaging the use of a solid catalyst in the catalytic ozonation process to avoid the separation step needed after the treatment for catalyst recovery when powdered material is involved. This work devoted to the removal of succinic acid (SA), a low molecular weight refractory organic acid whose heterogeneous catalytic ozonation was previously studied using powder form catalysts. Heterogeneous catalytic ozonation of succinic acid was carried out in a reactor fitted with a recirculation loop. This study investigated if the location of the solid catalyst, i.e. inside the reactor in the zone of ozone transfer or in a cartridge placed on the recirculation loop, had an influence on the amount of SA removed or on the elimination rate expression. No influence of the catalyst location was detected. The rates of SA removal by catalytic ozonation were found to depend on both the catalyst concentration and the initial SA concentration for weak SA initial concentrations, but only on the catalyst concentration for greater initial SA concentrations. The pattern of SA concentration from modelization closely matched the experimental results. This implementation of the catalytic ozonation process with solid catalyst is of interest for the application at industrial scale. The 1.8 ratio between the rates of SA and DOC removal showed that mineralization to CO₂ did not occur simultaneously with SA removal.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Heterogeneous catalytic ozonation is viewed as a promising way to treat waters containing refractory compounds such as dyes, some aromatic structures, low molecular weight substances, etc. [1–4]. There is growing interest in the choice and preparation of appropriate catalysts, as indicated by the numerous studies devoted to this subject [5–8]. However, only a few articles have assessed catalytic ozonation at semi-industrial or industrial scales. Most laboratory studies are performed on powder form catalysts poured in stirred vessels [9–11], and few of them optimize the

operating conditions from a process standpoint [12]. Several patented processes based on heterogeneous catalytic ozonation, with a powder catalyst operating in slurry-type reactor [13–15] or with pellets operating in a trickle-bed-type reactor, have also been reported [16–19]. However, no published data are available regarding the design and optimization of the operating conditions.

Using a powder catalyst implies a separation step to recover the catalyst. This step requires time, penalizing the whole process and being prohibitive to its use in industrial field. Furthermore, industrial effluents often contain suspended solids which could disturb the separation process whatever the technology is used. Using a solid catalyst, fixed in the system makes the separation step unnecessary and would allow the treatment of large flowrate.

* Corresponding author. Tel.: +33 (0)5 49 45 39 16.

E-mail address: Nathalie.karpel@univ-poitiers.fr (N. Karpel Vel Leitner).

This study was devoted to the optimization of operating conditions in a heterogeneous catalytic ozonation system through a study of the catalyst location in the process. The model molecule was succinic acid (SA), a low molecular weight refractory organic acid whose heterogeneous catalytic ozonation was previously studied by Ernst et al. [20], Karpel Vel Leitner et al. [21] and Delanoë et al. [22] using powder form catalysts. It was found that the presence of 0.2 g L^{-1} of a Ru/CeCO₂ catalyst enabled 100% removal of succinic acid (2 mM) within 90 min ozonation [22]. Total mineralization of a 1.25 mmol L^{-1} solution was achieved in about 1 h with 50 g L^{-1} of Al₂O₃ [20].

In the present study, the catalyst location and kinetic parameters were studied, while focusing particularly on the influence of succinic acid and catalyst concentrations. The catalyst used in this study was supported on a metallic porous structure.

2. Materials and methods

2.1. Material

The reactor containing 17 L of aqueous solution was continuously fed with gaseous ozone through a fine bubble diffuser. The device included a recirculation loop for the solution, as shown in Fig. 1. Ozone was produced from oxygen by a CFS-1A ozone generator (Ozonia). The volumetric flow rate was $0.16 \text{ Nm}^3 \text{ h}^{-1}$, with an ozone concentration of 130 g Nm^{-3} . Various recirculation flow rates between 80 and 160 L h^{-1} were tested. When the system was operated in continuous mode, the solution flow rate was 10 L h^{-1} .

The ozone gas concentration was monitored online using a BMT-694-BT ozone analyser (BMT Messtechnik). The catalyst was located either directly inside the reactor or in the recirculation loop and the system was operated in continuous or semi-continuous mode. When the catalyst was located directly inside the reactor, it was placed in a stainless steel basket submerged in the liquid phase. The catalyst was placed in a cartridge when it was located in the recirculation loop.

The catalyst was stuck on a disk shaped metallic foam (40 mm of diameter). The foam pore diameter was 2.3 mm, and the disk thickness was 5 mm. The mass of active material on the solid was around 400 mg per disk. One or three disks were used for the experiments, corresponding to 20.3 and 64.7 mg of catalyst per litre of solution in the reactor, respectively.

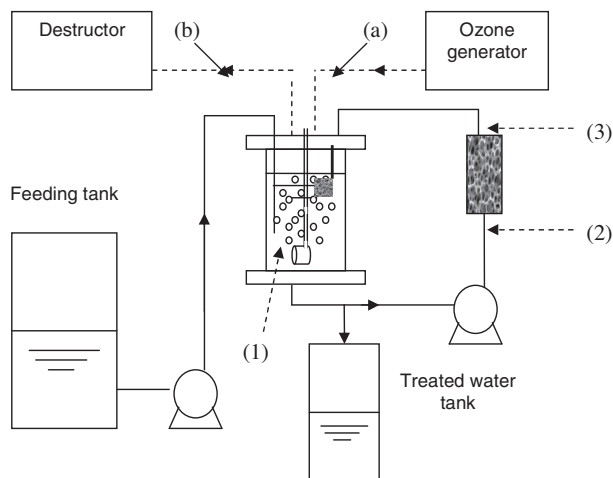


Fig. 1. Experimental reactor system with the catalyst located directly in the reactor, or in the recirculation loop. Samples can be drawn from three sampling points (1–3) and in the treated water tank.

2.2. Methods

All experiments were performed in demineralised water ($\text{DOC} = 0.1 \pm 0.05 \text{ mg C L}^{-1}$, $\text{pH} = 5.7$, conductivity = $1.6 \mu\text{S cm}$). Succinic acid of over 99% purity was supplied by Sigma–Aldrich®. Its concentration was measured by HPLC for different ozonation times, using an ion exchange resin-packed column composed of sulfonated di-vinylbenzene polystyrene (SPELCOGEL C-610) and a UV-detector set at 210 nm. The mobile phase was ultrapure water acidified with 1‰ (v/v) of phosphoric acid.

To assess the degree of mineralization of the succinic acid, DOC was measured using a TOC-Vcsh analyzer (Shimadzu Corporation).

Dissolved ozone was measured by the Indigo Trisulfonate method [23].

3. Results and discussion

3.1. Preliminary validation experiments

3.1.1. Mixing efficiency

Several experiments were performed to examine if: (i) the concentration was similar at a given time in different points of the reactor and in the recirculation loop, and (ii) the time needed to achieve a perfect mixing was sufficiently short to be considered negligible.

The mixing efficiency was assessed under semi-continuous conditions using Acid Red 94 (Rose Bengal) dye (Aldrich®) as tracer. For these experiments, gaseous ozone was replaced by oxygen. At t_0 , the initial time, 10 mL of Acid Red solution (5.2 g L^{-1}) were poured in the reactor containing 17 L of pure water. The recirculation flow rate was set at 120 L h^{-1} . Three different sampling points were chosen (Fig. 1): within the reactor (1), before (2) and after the catalyst-loaded cartridge (3) containing the catalyst. The absorbance of the samples withdrawn at different times was measured at 549 nm. The results are shown in Fig. 2.

These results were clearly repeatable: tests 1 and 2 exhibited the same time-course pattern. The dye concentration variations were the same in samples from the reactor and from the recirculation loop (sampling points: 1 and 2, respectively, in Fig. 1).

It appeared that after 40 s approximately 90% of the final dye concentration was reached inside the reactor and in the recirculation loop and perfect mixing was obtained within only 1 min. Consequently, injecting fine bubbles of ozone gas ($0.16 \text{ Nm}^3 \text{ h}^{-1}$) with a recirculation flow rate of 120 L h^{-1} could provide sufficient

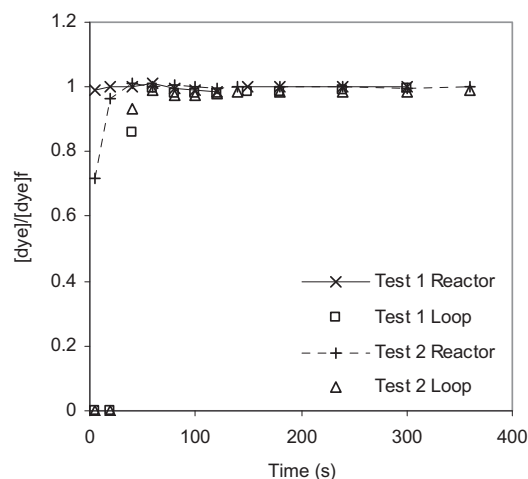


Fig. 2. Time-course variations in the dye concentration in the reactor and in the recirculation loop (gas flow rate: $0.16 \text{ Nm}^3 \text{ h}^{-1}$, recirculation flow rate: 120 L h^{-1}).

Download English Version:

<https://daneshyari.com/en/article/146140>

Download Persian Version:

<https://daneshyari.com/article/146140>

[Daneshyari.com](https://daneshyari.com)