Data Article

Data for persulfate activation by UV light to degrade theophylline in a water effluent

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ARTICLE INFO

Article history:
Received 16 August 2019
Received in revised form 9 September 2019
Accepted 26 September 2019
Available online 7 October 2019

Keywords:
Persulfate
UV
254 nm
AOPs
Theophylline
Degradation

ABSTRACT

The aim of this study was to degrade theophylline (TP) drug in a pharmaceutical effluent solution utilizing persulfate (PS). A simulated and a real effluent solution were used, with different conditions tested to optimize the degradation process. HPLC analyses and a modified-HPLC method were used to track concentrations of TP and PS respectively in the treatment process. Experiments were done in triplicates and treated data is presented as graphs. A detailed analyses of this study can be found in the article “Degradation of theophylline in a UV254/PS system: matrix effect and application to a factory effluent” [1] published in Chemical Engineering Journal.

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1. Data

Calibration curve and some properties for TP and presented in Fig. 1. The observed rate constants, obtained for pseudo-first order fitting, for degradation of TP at different [PS]₀ is presented in Table 1. A mini review of effects of chlorides and bicarbonates on the degradation of different organic probes in PS-based AOPs are presented in Table 2 and Table 3 respectively.

TP degradation in UV/PS system in the presence of phosphates (Fig. 2) and chloroform (Fig. 4) is studied in a simulated pharmaceutical effluent with [TP]₀ = 10 mg L⁻¹ and [PS]₀ = 0.25 mM. Comparison between UV and base activation of PS is studied with 10 mM phosphate buffer at pH = 11 used (Fig. 3). Additionally, H₂O₂ and PS oxidants are compared (Fig. 5).

A real effluent solution containing [TP]₀ = 160 mg L⁻¹ is treated with simultaneous and successive additions of PS (Fig. 6a). The UV254/PS system was studied upon spiking the reactor with varying amounts of concentrated effluent solution (Fig. 6b).

The raw data corresponding to all figures and tables can be found in the file “raw data” within this article. These data include concentrations of TP and PS and the standard deviation for their measurement.
2. Experimental design, materials, and methods

2.1. Experimental setup: UV reactors

A bench-scale experimental setup was used simulating the commercial UV water sterilization apparatus. Six units of a generic 1 gallon per minute UV water disinfection element (Fig. 7a) were used in the construction of the experimental setup. The commercial units utilized originally consist of a UV 11-W low-pressure mercury lamp (LPHgL) (Philips TUV 11W G11 T5, Poland), fitted in a quartz tube.
Table 2
Review of the effect of chloride (Cl\(^{-}\)) presence on the degradation of several organic probes in Persulfate activated systems. (+) and (−) represent an increase or a decrease in the degradation rate constant of the oxidation reaction, respectively.

| Family       | Probe         | Structure | PS system used | \([PS]_0\) mM | \([\text{Cl}^-]\) (mM) | \([\text{NaCl}]\) (mg L\(^{-1}\)) | Effect on \(k_{\text{obs}}\) | Main reactive radical species | Reference |
|--------------|---------------|-----------|----------------|----------------|-------------------|-------------------------------|-------------------|-------------------------------|-----------|
| Antibiotic   | Florfenicol   |           | UV/PS          | 1              | 1, 10, 100        | 58.4, 584, 5844               | −                 | No data                        | [3]       |
|              | Sulfamethazine|           | Heat/PS        | 2              | 5, 10             | 292, 584                   | Negligible effect       | \(\text{SO}_4^{2-}\)           | [4]       |
|              | Penicillin G  |           | Heat/PS        | 0.5            | 0.171             | 10                          | −                 | \(\text{SO}_4^{2-}\)           | [5]       |
| Antipyretic  | Acetaminophen |           | Fe\(^{2+}\)/PS | 0.8            | 0.1               | 5.84                        | Negligible effect       | \(\text{SO}_4^{2-}\)           | [6]       |
|              | Antipyrine    |           | Heat/PS        | 1.855          | 18.5              | 1081                        | +                 | \(\text{SO}_4^{2-}\)           | [7]       |
| Beta blocker | Atenolol      |           | Heat/PS        | 0.5            | 1, 10, 50         | 58.4, 292, 584, 2922        | Negligible effect       | \(\text{SO}_4^{2-}\)           | [9]       |
|              | Bisoprolol    |           | Heat/PS        | 1              | 0.0855            | 5                           | +                 | \(\text{SO}_4^{2-}, \text{OH}^*\) | [10]      |
| Hormone                  | UV/PS | Slow-release flow through system | Concentration (µg/L) | Nonsteroidal anti-inflammatory |
|-------------------------|-------|---------------------------------|----------------------|-------------------------------|
| 17β-estradiol           |       |                                 | 0.028, 0.282, 2.82  |                               |
|                         |       |                                 | 28.2, 42.3           |                               |
|                         |       |                                 | 56.4, 564, 846, 1269 |                               |
|                         |       |                                 | 1.6, 16.5, 165       |                               |
|                         |       |                                 | 1648, 2472           |                               |
|                         |       |                                 | 3256, 32960, 49440, 74160 |                |
| Nonsteroidal anti-inflammatory | UV/PS |                                 | 0.028, 0.282, 2.82  |                               |
|                         |       |                                 | 28.2, 42.3           |                               |
|                         |       |                                 | 56.4, 564, 846, 1269 |                               |
|                         |       |                                 | 1.6, 16.5, 165       |                               |
|                         |       |                                 | 1648, 2472           |                               |
|                         |       |                                 | 3256, 32960, 49440, 74160 |                |

**Nonsteroidal anti-inflammatory**

| Diclofenac   | UV/PS | 25, 50, 100, 200 | 1461, 2922, 5844, 11688 | No data                     |

**Ketoprofen**

| UV/PS | 1, 25, 50 | 1461, 2922 | 5844, 11688 | No data                     |

| Heat/PS | 1, 50, 100 | 58.4, 584.4, 5844 | Negligible effect | No data                     |

**Nonsteroidal anti-inflammatory**

| Nonsteroidal anti-inflammatory | UV/PS | 25, 50, 100, 200 | 1461, 2922, 5844, 11688 | No data                     |

**Persistent organic molecules**

| Cyclohexanoic acid | UV/PS | 14.1 | 824 | No data                     |

| 1,1,1-Trichloroethane | Heat/PS | 15 | 1 | 58.4 | Negligible effect | No data |
|                       |       |    | 10, 100 | 584.4, 5844 |                   |       |

**Pesticides**

| 1,1,1-Trichloroethane | Heat/PS | 15 | 1 | 58.4 | Negligible effect | No data |
|                       |       |    | 10, 100 | 584.4, 5844 |                   |       |

| Triclosan | Heat/PS | 0.155 | 0.15, 1, 10 | 8.8, 58.4, 584.4 | Negligible effect | No data |
|           |       | 20, 50 | 1689, 2922 | 1169, 2922 |                   |       |

| Triclosan | Heat/PS | 0.155 | 0.15, 1, 10 | 8.8, 58.4, 584.4 | Negligible effect | No data |
|           |       | 20, 50 | 1689, 2922 | 1169, 2922 |                   |       |

| Diatrizoate | UV/PS | 12 | 1, 10, 100 | 58.4, 584.4, 5844 | Negligible effect | No data |
|            |       |    | 500 | 29220 |                   |       |

**Pesticides**

| Diatrizoate | UV/PS | 12 | 1, 10, 100 | 58.4, 584.4, 5844 | Negligible effect | No data |
|            |       |    | 500 | 29220 |                   |       |
### Table 3

Review of the effect of bicarbonate (HCO₃⁻) presence on the degradation of several organic probes in Persulfate activated systems. (+) and (−) represent an increase or a decrease in the degradation rate constant of the oxidation reaction, respectively.

| Family     | Probe       | Structure | PS system used | [PS]₀ (mM) | [HCO₃⁻] (mM) | Effect on kₜₚₘₛ | Main reactive radical species | Reference |
|------------|-------------|-----------|----------------|------------|--------------|----------------|-------------------------------|-----------|
| Antibiotic | Florfenicol | UV/PS     | 1              | 1, 10, 100 | −            | No data                   | [3]          |
|            | Sulfamethazine | Heat/PS   | 2              | 5, 10, 50  | +            | SO₄⁻                     | [4]          |
|            | Penicillin G  | Heat/PS   | 0.5            | 0.0943     | −            | SO₄⁻                     | [5]          |
| Antipyretic | Acetaminophen | UV/PS     | 0.132          | 2 10, 20, 50, 100 | + | SO₄⁻                     | [7]          |
|            | Antipyrine    | Heat/PS   | 1.855          | 18.55, 92.75, 185.5, 927.5 | − | SO₄⁻                     | [8]          |
| Beta blocker | Atenolol    | Heat/PS   | 0.5            | 1, 5, 10, 50 | −            | SO₄⁻                     | [9]          |
| Hormone    | 17β-estradiol | UV/PS     | Slow-release flow through system | 4.76 11.9, 23.8 | + | SO₄⁻                     | [11]         |
| Category                          | Compound          | Method   | Concentrations | Results | Notes       |
|----------------------------------|-------------------|----------|----------------|---------|-------------|
| Nonsteroidal anti-inflammatory   | Diclofenac        | UV/PS    | 1              | 25, 50, 100 and 200 | +          | No data [12] |
|                                  |                   |          |                |         |             |
|                                  | Ketoprofen        | Heat/PS  | 1              | 1, 50, 100 mM    | –          | No data [13] |
| Persistent organic molecules     | 1,1,1-Trichloroethane | Heat/PS  | 15             | 1, 10, 100       | –          | No data [15] |
| Pesticide                        | Triclosan         | Heat/PS  | 0.155          | 1–50             | –          | $SO_4^{2-}$ [16] |
|                                  | Diatrizoate       | UV/PS    | 12             | 2.5, 5, 10, 20, 40 | +          | $SO_4^{2-}$ [17] |
placed in a 280 mL 316 stainless steel casing that continuously intakes pre-filtered water. The UV lamps and their quartz fittings were utilized from the commercial product, and the stainless-steel casing was replaced by a 400 mL home-made borosilicate cylinder. The borosilicate used in the setup is as inert as the stainless-steel used in the commercial product, however the borosilicate allows easy handling and real-time monitoring of reactors to assure no precipitates were formed. Our developed system (Fig. 7b) utilizes a temperature-controlled water bath (20 °C) by the aid of a PolyScience MM7 chiller. UV-254 nm intensity was 3.2 mW cm⁻² ± 0.16 measured using a radiometer (Model UVC-254 Lutron; Taipei, Taiwan) at a distance of 3 cm, which is similar to the distance between the lamp and the termination part of the solution treated in the designed system. All experiments were done in triplicates, where two different experiments could be done at once.
2.2. Experimental procedure

Stock solution of TP (0.555 mM) was prepared by dissolving 100 mg of dry TP powder in one liter of DI water, and that of Persulfate (PS) (100 mM) was prepared by dissolving 2.38 g of sodium persulfate in 100 mL of DI water. Both solutions were stored in dark at 4°C for a maximum period of two weeks.

For persulfate detection method potassium iodide solution (40 mM) was prepared by dissolving 6.64 g of KI and 5 g of NaHCO3 in 1 L volumetric flask and left to stir overnight. Before every experiment LPHgLs (11 Watts) were stabilized for a period of 60 min. Monthly routine check of the LPHgL’s UV-254 nm intensity was performed using a UVC radiometer (UVC-254 Lutron, Taiwan) to confirm that the power of the lamps is within ±10% the original value; otherwise the lamps were replaced by new ones.

The water chiller was turned on and set at 20°C 60 minutes ahead of all experiments. Temperature of water in reaction medium was measured several times during the experiment to make sure it is within a range of 20 ± 2°C. The order of addition of reagents was as follows: first Theophylline solution from stock was placed in the reactor, then DI was added, followed by additive matrix solutions (if any), finally, PS or H2O2, from stock solutions were added. Samples were withdrawn every 2 min using a

Fig. 4. TP degradation in presence and absence of chloroform used as superoxide radical quencher. Solution is saturated with chloroform at 20 °C. Experimental conditions: [TP]0 = 10 mg L−1 and [PS]0 = 0.25 mM.

Fig. 5. Comparison of oxidation effectivity of H2O2 and PS toward TP. (a) [TP]/[TP]o for three different tested systems and (b) the corresponding pH at initial (t = 0 min) and final time (t = 16 min) with the calculated kobs. Experimental conditions: [TP]0 = 10 mg L−1, [PS]0 = 0.25 mM, [H2O2]0 = 0.25 mM and [PS]0 = [H2O2]0 = 0.125 mM for the case of mixed oxidants.

| System     | pHi | pHf | kobs (min⁻¹)  |
|------------|-----|-----|--------------|
| PS         | 5.7 | 3.4 | 1.73 (± 0.04) x 10¹ |
| PS + H2O2  | 5.2 | 5.5 | 3.6 (± 0.4) x 10¹ |
| H2O2       | 5.4 | 4.4 | 3.5 (± 0.2) x 10¹ |

Stock solution of TP (0.555 mM) was prepared by dissolving 100 mg of dry TP powder in one liter of DI water, and that of Persulfate (PS) (100 mM) was prepared by dissolving 2.38 g of sodium persulfate in 100 mL of DI water. Both solutions were stored in dark at 4 °C for a maximum period of two weeks. For persulfate detection method potassium iodide solution (40 mM) was prepared by dissolving 6.64 g of KI and 5 g of NaHCO3 in 1 L volumetric flask and left to stir overnight. Before every experiment LPHgLs (11 Watts) were stabilized for a period of 60 min. Monthly routine check of the LPHgL’s UV-254 nm intensity was performed using a UVC radiometer (UVC-254 Lutron, Taiwan) to confirm that the power of the lamps is within ±10% the original value; otherwise the lamps were replaced by new ones. The water chiller was turned on and set at 20 °C 60 minutes ahead of all experiments. Temperature of water in reaction medium was measured several times during the experiment to make sure it is within a range of 20 ± 2 °C. The order of addition of reagents was as follows: first Theophylline solution from stock was placed in the reactor, then DI was added, followed by additive matrix solutions (if any), finally, PS or H2O2, from stock solutions were added. Samples were withdrawn every 2 min using a
Fig. 6. Degradation of TP in a real pharmaceutical effluent sample in UV254/PS system. (a) PS is added either successively over 5 cycles ([PS] = 5 mM/cycle) or simultaneously at t = 0 min ([PS] = 25 mM) to TP effluent solution ([TP]₀ = 160 mg L⁻¹). (b) Concentrated effluent solution spiked over 3 cycles of 1 h each starting at [TP]₀ = 160 mg L⁻¹ and [PS]₀ = 25 mM added at t = 0 min. The insets correspond to the average % RSE calculated for every cycle. Error bars are calculated as $\frac{\text{ts}}{\text{TP}}$, where absent bars fall within the symbols.
Fig. 7. (a) Generic 1 gallon per minute UV continuous water disinfection element, and (b) an exploded diagram of the experimental setup.
separate syringe for each reactor and placed in 2 mL HPLC vials after filtration by a 0.45 μm PTFE 13 mm disc filters. Wastewater used in experiments was pre-filtered using a 1 μm ashless glass fiber filter. For the case of real pharmaceutical factory effluent, water samples were collected from washing the reactors used in the production process (Fig. 8).

Acknowledgments

This research was funded in part by the Lebanese National Council for Scientific Research (Award Number 103250), the K Shair CRSL fund (Award Number 103191), and the University Research Board (Award Number 103186) of the American University of Beirut and USAID-Lebanon through The National Academy of Sciences under PEER project 5-18 (Award number 103262). The author is thankful to Joan Younes, Samer Khalil, Simon Al-Ghawy, and Boutros Sawaya for their technical assistance and the personnel of the K. Shair CRSL for their kind help.

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.dib.2019.104614.

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