Data Article

Data on treatment of nafcillin and ampicillin antibiotics in water by sonochemistry

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Article info
Article history:
Received 30 January 2020
Received in revised form 22 February 2020
Accepted 24 February 2020
Available online 2 March 2020

Keywords:
β-Lactam antibiotics
Combination of processes
Matrix effect
Sonochemistry
Water treatment

Abstract
Ampicillin and nafcillin antibiotics were treated by high frequency ultrasound (at 375 kHz and 24.4 W). Degradations followed pseudo-first order kinetics, which constants were k: 0.0323 min⁻¹ for AMP and k: 0.0550 min⁻¹ for NAF. Accumulation of sonogenerated hydrogen peroxide and inhibition degree of sonochemical removal (IDS) in presence of a radical scavenger were also established. Afterwards, ultrasound was combined with UVC light (sono-photolysis), with ferrous ion (sono-Fenton), and with ferrous ion plus UVC light (sono-photo-Fenton) to degrade the antibiotics. Furthermore, treatment of the pollutants in a complex matrix and removal of antimicrobial activity (AA) were considered. The antibiotics evolution was followed by HPLC-DAD technique and the accumulation of sonogenerated H₂O₂ was measured by an iodometry-spectrophotometry methodology (77.6 and 57.3 μmol L⁻¹ of H₂O₂ after 30 min of sonication were accumulated in presence of AMP and NAF, respectively). IDS was analyzed through treatment of the antibiotics in presence of 2-propanol (87.1% for AMP and 56% for NAF) and considering the hydrophobic character of pollutants (i.e., Log P values). Antimicrobial activity evolution was assessed by the Kirby-Bauer method using Staphylococcus aureus as indicator microorganism (sono-photo-Fenton process removed 100% of AA after 60 and 20 min for AMP and NAF, respectively). Finally, for degradations in the complex matrix, a...
simulated effluent of municipal wastewater treatment plant was utilized (sono-photo-Fenton led to degradations higher than 90% at 60 min of treatment for both antibiotics). The data from the present work can be valuable for people researching on treatment of wastewaters containing antibiotics, application of advanced oxidation technologies and combination of sonochemical process with photochemical systems.

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1. Data description

To determine the chemical structure effect of antibiotics, the individual elimination by sonochemistry (at 375 kHz of frequency and 24.4 W of actual power) of ampicillin (AMP) and nafcillin (NAF) was initially carried out in distilled water. The pollutants degradation followed a pseudo first-order
kinetics; thus, the corresponding degradation constants (k) were calculated, which are shown in Fig. 1A [1].

It is well-known that the sonochemical process produces hydroxyl radical (HO·) through water cleavage (Eq. (1)). Hydroxyl radicals can attack organic pollutants (Eq. (2)) such as antibiotics, or combine themselves to form hydrogen peroxide (H2O2, Eq. (3)) [1]. Indeed, the accumulation of H2O2 during process is an indicator of sonochemical activity [2]. Fig. 1B shows the H2O2 evolution during the sonochemical treatment of NAF or AMP.

\[
\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{HO}^-
\]  

(1)

\[
\text{HO}^- + \text{organic pollutants} \rightarrow \text{degradation products}
\]  

(2)

\[
2 \text{HO}^- \rightarrow \text{H}_2\text{O}_2
\]  

(3)

![Fig. 1. Individual treatment of ampicillin (AMP) and nafcillin (NAF) by sonochemistry. A. Pseudo-first order degradation constants (k). B. Hydrogen peroxide accumulation during degradation of the antibiotics by sonochemistry. Experimental conditions: [AMP]: [NAF]: 30 μM, power: 24.4 W, frequency: 375 kHz, initial pH: 6.5, volume: 250 mL, temperature: 20 ± 1 °C.](image-url)
To test the closeness of the antibiotics to cavitation bubbles, the compounds were treated in
presence of 2-propanol (100 times more concentrated than the pollutants) [3]. Then, inhibition degree
of sonochemical degradation (IDS) was calculated according to Eq. (4) (based on the pseudo-first order
constants for treatment in absence and presence of the scavenger). Table 1 presents the IDS values,
which was 87 and 56% for AMP and NAF, respectively. Additionally, Table 1 contains the Log P values for
both antibiotics (this parameter is related to the hydrophobic nature of organic pollutants, [4]).

\[
\text{IDS} = \left( \frac{k_{\text{in}} \text{ distilled water} - k_{\text{in}} \text{ 2-propanol presence}}{k_{\text{in}} \text{ distilled water}} \right) \times 100
\]

A strategy to increase the degradation kinetics is the combination of ultrasound with other
advanced oxidation processes [6]. Thus, in this work, ultrasound was combined with UVC light radi-
ation (US/UVC, sono-photoysis) to promote extra formation of radicals through a homolysis sono-
generated hydrogen peroxide (Eq. (5)). Also, it was evaluated the addition of UVC plus ferrous ions to
the sonochemical system (US/UVC/Fe(II), sono-photo-Fenton), with the purpose of increasing the
amount of radical species by interaction of sonogenerated H$_2$O$_2$ with iron (generating a photo-Fenton
process, (Eqs. (6)–(8))) [7]. Moreover, control experiments (i.e., the individual degrading action of Fe (II)
or UVC light) were also taken into account to determine the contribution of degradations due to iron
(Eqs. (9)–(10)) and the photolysis of antibiotics (Eq. (11)). The data are given in terms of the pseudo-
first order degradation constants for each process in Fig. 2.

\[
\begin{align*}
\text{H}_2\text{O}_2 + \text{UVC} & \rightarrow 2\text{HO}^- \quad (5) \\
\text{Fe}^{2+} + \text{H}_2\text{O}_2 & \rightarrow \text{Fe}^{3+} + \text{HO}^- + \text{OH}^- \quad (6) \\
\text{Fe}^{3+} + \text{H}_2\text{O} + \text{hv} & \rightarrow \text{Fe}^{2+} + \text{HO}^- + \text{H}^+ \quad (7) \\
\text{Fe}^{3+} + \text{H}_2\text{O}_2 + \text{hv} & \rightarrow \text{Fe}^{2+} + \text{HO}_2^- + \text{H}^+ \quad (8) \\
\text{Fe}^{2+} + \text{O}_2 & \rightarrow \text{Fe}^{3+} + \text{O}_2^- \quad (9) \\
\text{O}_2 + \text{organic pollutants} & \rightarrow \text{degradation products} \quad (10) \\
\text{UVC} + \text{organic pollutants} & \rightarrow \text{photodegradation products} \quad (11)
\end{align*}
\]

One of the most important parameters to consider during degradation of antibiotics is the evo-
lution of antimicrobial activity (AA), due to in some cases, despite of antibiotic removal the activity can
persist [8,9]. Thus, for AMP and NAF treatment by ultrasound (US), sono-photoysis (US/UVC) and sono-
photo-Fenton (US/UVC/Fe(II)) processes, the evolution of AA was determined. Fig. 3 presents the data
of the antimicrobial activity for each system.

The individual elimination of the antibiotics in a complex matrix by sono-photo-Fenton system
(which showed the best performance in Figs. 2–3) was applied to a simulated effluent of wastewater
treatment plant (WWTP, composition in Table 2). Fig. 4 compares the antibiotics removal in distilled
water (DW) and in the complex matrix (WWTP) by the sono-photo-Fenton process.

### Table 1

| Antibiotic       | IDS (%) | Log P $^a$ |
|------------------|---------|------------|
| Ampicillin       | 87.1    | 1.35       |
| Nafcillin        | 56.0    | 3.30       |

$^a$ Log P values were taken from PubChem [5].

Fig. 2 presents the data of the evolution of antimicrobial activity for each system.
Finally, it is presented the Table 3, which contains data from our research and previous works about degradation of AMP and NAF by others advanced oxidation processes (AOP).

2. Experimental design, materials, and methods

2.1. Reagents

Ampicillin trihydrate was provided by Syntopharma. Sodium nafcillin was purchased from Sigma. Sodium chloride, potassium chloride, acetonitrile, urea, nutrient agar, magnesium sulfate heptahydrate and sodium sulfate were purchased from Merck. Dipotassium hydrogen phosphate, sodium bicarbonate, calcium sulfate dihydrate and ferrous sulfate heptahydrate were provided by Panreac. Formic

![Fig. 2. Degradation rate constants (k) for the processes combination. A. Case of AMP. B. Case of NAF. US: sonochemistry, UVC: photolysis by UV 254 nm, US/UVC: sono-photolysis, US/UVC/Fe(II): sono-photo-Fenton and Fe (II): action of iron (II) alone. Experimental conditions: [AMP]: [NAF]: 30 $\mu$M, [Fe$^{2+}$]: 90 $\mu$M, UVC lamp: 4 W, actual ultrasound power: 24.4 W, frequency: 375 kHz, initial pH: 6.5, volume: 250 mL, temperature: 20 $\pm$ 1°C.](image-url)
Fig. 3. Elimination of antimicrobial activity (AA) against S. aureus by the different systems. A. Data for AMP. B. Data for NAF. US: sonochemistry, US/UVC: sono-photolysis and US/UVC/Fe(II): sono-photo-Fenton. Experimental conditions: [AMP]: [NAF]: 30 μM, [Fe²⁺]: 90 μM, UVC lamp: 4 W, actual ultrasound power: 24.4 W, frequency: 375 kHz, initial pH: 6.5, volume: 250 mL, temperature: 20 ± 1 °C.

Table 2
Composition of simulated effluent of wastewater treatment plant (WWTP, [10]).

| Compound       | Concentration (mg/L) | Concentration (μM) |
|----------------|----------------------|--------------------|
| NaCl           | 7                    | 119                |
| KCl            | 4                    | 54                 |
| CaCl₂ · 2H₂O   | 4                    | 27                 |
| NaHCO₃         | 96                   | 1.142              |
| CaSO₄·2H₂O     | 60                   | 348                |
| MgSO₄·7H₂O     | 125                  | 507                |
| K₂HPO₄         | 28                   | 161                |
| Urea           | 6                    | 99.9               |
| Peptone        | 32                   | –                  |
| Meat extract   | 22                   | –                  |

-Not applicable.
acid from Carlo Erba was used. Peptone and meat extract were purchased from Oxoid. All chemicals were used as received. The solutions of antibiotics were prepared using distilled water.

2.2. Reaction systems

A Meinhardt ultrasound reactor was used for sonochemical process operated at 375 kHz and 24.4 W. For the combined system, the ultrasound reactor was complemented by an UVC-lamp (4 W) with main emission at 254 nm (OSRAM G4T5/OF) placed on a quartz sleeve (which was submerged in the aqueous sample). In all cases, the reactor temperature was controlled using a Huber Minichiller.

Fig. 4. Comparison of antibiotics degradation in distilled water (DW) and in synthetic municipal wastewater treatment plant effluent (WWTP) by sono-photo-Fenton treatment. A. Case of AMP. B. Case of NAF. Experimental conditions: [AMP]: [NAF]: 30 μM, [Fe^{2+}]: 90 μM, UVC lamp: 4 W, actual ultrasound power: 24.4 W, frequency: 375 kHz, initial pH: 6.5, volume: 250 mL, temperature: 20 ± 1 °C.
Table 3
Data on AMP and NAF degradation by diverse AOP.

| Antibiotic [reference] | AOP                                      | Experimental conditions                                                                 | Pseudo-first order constant (k) | Other relevant data                                                                 |
|------------------------|------------------------------------------|-----------------------------------------------------------------------------------------|--------------------------------|-------------------------------------------------------------------------------------|
| AMP [11]               | Electrochemical oxidation                | [AMP]: 50 mg L\(^{-1}\) BDD anode/GDE cathode Current density: 5 mA cm\(^{-2}\) [Na\(_2\)SO\(_4\)]: 0.05 M pH: 2.8 Volume: 250 mL | 0.549 min\(^{-1}\) (9.15 x 10\(^{-3}\) s\(^{-1}\)) | • 32% of mineralization after 120 min of treatment.                                   |
| AMP [11]               | Electro-Fenton                           | [AMP]: 50 mg L\(^{-1}\) BDD anode/GDE cathode Current density: 5 mA cm\(^{-2}\) [Na\(_2\)SO\(_4\)]: 0.05 M pH: 2.8 [Fe\(^{2+}\)]: 1 mg L\(^{-1}\) Volume: 250 mL | 0.606 min\(^{-1}\) (1.07 x 10\(^{-2}\) s\(^{-1}\)) | • 43% of mineralization after 120 min of treatment.                                   |
| AMP [11]               | Photo-Electro-Fenton                     | [AMP]: 50 mg L\(^{-1}\) BDD anode/GDE cathode Current density: 5 mA cm\(^{-2}\) [Na\(_2\)SO\(_4\)]: 0.05 M pH: 2.8 [Fe\(^{2+}\)]: 1 mg L\(^{-1}\) UVA light: 5.0 W m\(^{-2}\) Volume: 250 mL | 1.086 min\(^{-1}\) (1.81 x 10\(^{-3}\) s\(^{-1}\)) | • 63% of mineralization and Complete AA removal after 120 min of treatment.          |
| AMP [11]               | Non-thermal plasma                       | [AMP]: 20 mM (6.99 g L\(^{-1}\)) Plasma was generated using a nanosecond-pulsed power supply with alternating polarity and a floating electrode-dielectric barrier discharge. Samples were treated under atmospheric conditions with no gas flow. The treatment of all samples was at 11.2 kV and 690 Hz Not reported | 0.015 min\(^{-1}\) | • Complete AMP degradation was achieved after 5 min of treatment.                    |
| AMP [12]               | ZnO photocatalysis                       | [AMP]: 105 mg L\(^{-1}\) [ZnO]: 0.5 g L\(^{-1}\) UVA light: 6 W pH: 11.0 Volume: 500 mL | 0.1065 min\(^{-1}\) | • 9.7% of mineralization after 180 min of treatment.                                 |
| AMP [in this work]     | Sono-photo-Fenton                        | [AMP]: 30 μM (10.5 mg L\(^{-1}\)) [Fe\(^{2+}\)]: 90 μM (5.0 mg L\(^{-1}\)) UVC light: 4 W Ultrasound power: 24.4 W Frequency: 375 kHz initial pH: 6.5 Volume: 250 mL | 0.604 min\(^{-1}\) (1.00 x 10\(^{-3}\) s\(^{-1}\)) | • Complete AA removal after 60 min of treatment.                                       |
| NAF [14]               | Electrochemical oxidation                | [NAF]: 50 mg L\(^{-1}\) BDD anode/GDE cathode Current density: 5 mA cm\(^{-2}\) [Na\(_2\)SO\(_4\)]: 0.05 M pH: 2.8 Volume: 250 mL | 0.604 min\(^{-1}\) (1.00 x 10\(^{-3}\) s\(^{-1}\)) | • -50% of AA removal after 90 min of treatment.                                        |
| NAF [14]               | Electro-Fenton                           | [NAF]: 50 mg L\(^{-1}\) BDD anode/GDE cathode Current density: 5 mA cm\(^{-2}\) [Na\(_2\)SO\(_4\)]: 0.05 M pH: 2.8 [Fe\(^{2+}\)]: 1 mg L\(^{-1}\) Volume: 250 mL | 0.873 min\(^{-1}\) (1.46 x 10\(^{-3}\) s\(^{-1}\)) | • -60% of AA removal after 90 min of treatment.                                        |
2.3. Analyses

Antibiotics degradation was followed using UHPLC Thermoscientific Dionex UltiMate 3000 instrument equipped with an AcclaimTM 120 RP C18 column (5 μm, 4.6x150 mm) and a diode array detector, through the methods utilized by Vidal et al. [11,14]. Accumulation of hydrogen peroxide was determined by an iodometry-spectrophotometry methodology according to Serna-Galvis et al. [2]. The antimicrobial activity (AA) was determined by measurement of the inhibition zone in the agar diffusion test [15].

Acknowledgments

Authors from GIRAB thank Universidad de Antioquia UdeA for the support provided to their research group through “PROGRAMA DE SOSTENIBILIDAD” and the financing from Minciencias (before called COLCIENCIAS) through the project No. 111577757323. Y. Ávila-Torres thanks Universidad Santiago de Cali for support through the project DGI No. 63661. E. A. Serna-Galvis thanks Minciencias (before called COLCIENCIAS) for his PhD fellowship during July 2015—June 2019 (Convocation 647 de 2014).

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.2020.105361.

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