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Modelling of patulin photo-degradation by a UV multi-wavelength emitting lamp



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ABSTRACT

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Keywords: Patulin UV Radiation Photo-reactor Photo-degradation Treatment Juice Apple In this paper, a new mechanism is proposed to describe the degradation of patulin using UV radiation from a midpressure lamp with emission wavelengths between 250 and 740 nm. The mechanism consists of three stages: excitation of the patulin molecule that can desorb the energy to its fundamental state or degrade to photoproducts. It is shown that this mechanism is in line with a first-order kinetic that best describes the process.

Considering the linear spherical emission model and using the Simpson integration method, the spectral radiant power absorbed by the whole solution is evaluated, and also its dependence on patulin concentration and the incident spectral radiant power at different depths.

Previous works studied the degradation of patulin using UV radiation, but using exclusively the germicidal wavelength of 254 nm. Moreover, none of them proposed a reaction mechanism, nor evaluated the spectral radiant power absorbed by the whole solution.

First of all, the absorption spectrum of patulin was obtained. It was concluded that both the absorption by patulin and emission from the lamp coexist in the range between 255 and 350 nm.

The kinetic constants for aqueous solutions of 500 μ g/L of patulin at pH = 4 and pH = 7 and temperatures of 8, 25, 45, and 65 °C were obtained. In order to know the effect of the juice matrix on the photodegradation process, the kinetic constants for clarified 12 °Brix apple juice containing 500 μ g/L of patulin were obtained at the same *pH* and temperature values.

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1. Introduction

Patulin (PAT) is a mycotoxin produced by fruit decay fungi, in particular *Penicillium, Aspergillus* and *Byssochlamys* species, *Penicillium expansum* being the most relevant (Bosco & Mollea, 2012). PAT can be found mainly in apple and apple products, with much less frequent contamination in other food products (Moake, PadillaZakour, & Worobo, 2005). Marín et al. (2011) published a summary of the surveys published after year 2000, showing an average of 47% of positive samples of apple juices and nectars.

As PAT was shown to be mutagenic and to cause neurotoxin, immunotoxic, genotoxic and gastrointestinal effects in rodents (Hopkins, 1993), the maximum tolerance limit in juices was established at 50 μ g/L (European Commission, 2003; FDA-U.S. Food and Drug Administration, 2004; WHO, 1995).

PAT contamination of apple derivatives has to be minimized by removing and trimming decayed and damaged fruit (Codex Alimentarius Commission, 2003; FAO-Food and Agriculture organization of the United Nations, 2003). Nevertheless, a single contaminated apple in a batch of up to 1000 apples can cause a content

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higher than the maximum permitted level (Salomao, Aragao, Churey, & Worobo, 2008). Moreover, fungal growth can occur internally, as a consequence of various kinds of damage, and can result in the occurrence of PAT in externally undamaged fruit (Bosco & Mollea, 2012) and the contamination can also appear or be increased during storage. So, the mould has to be eliminated in fresh fruits and an effective method for degrading PAT without modifying endproduct quality has to be found. As juice processing can only eliminate under half of the initial PAT concentration (Acar, Gokmen, & Taydas, 1998) and it is also highly resistant to pasteurization (Wheeler, Harrison, & Koehler, 1987), many alternative methods have been studied to eliminate this mycotoxin in apple juices. These include filtration (Huebner et al., 2000), adsorption (Kadakal & Nas, 2002), ozone (McKenzie et al., 1997), pressure (Bruna, Voldrich, Marek, & Kamarad, 1999), ionizing irradiation (Zegota, Zegota, & Bachmann, 1988), pulsed light (Funes, Gómez, Resnik, & Alzamora, 2013) and addition of chemical additives such as sulphur compounds (Burroughs, 1977), ascorbic acid (Brackett & Marth, 1979), thiamine hydrochloride, pyridoxine hydrochloride and calcium pantothenate (Yazici & Velioglu, 2002), sodium benzoate (Roland, Beuchat, Worthington, & Hitchcock, 1984), potassium sorbate and sodium propionate (Lennox & McElroy, 1984). Most of these alternative methods provoke great changes to the final juice or are unable to eliminate the total PAT content.

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Ultraviolet irradiation is widely applied in industries to disinfect drinking water, air and surface disinfection and toxin degradation (Falguera, Pagán, Garza, Garvín, & Ibarz, 2011). Dong et al. (2010) reported that UV irradiation at the exclusive germicidal wavelength (254 nm) could lead to a reduction and/or elimination of PAT in apple cider and juice products without unwanted alterations of the final product. Assatarakul, Churey, Manns, and Worobo (2012) later concluded that the reaction followed a first-order kinetic model and Tikekar, Anantheswaran, and LaBorde (2013) concluded the more time needed than for the reduction of pathogens.

All the previous works that studied the UV photodegradation of PAT used a lamp emitting exclusively at the germicidal wavelength of 254 nm and none of them proposed a reaction mechanism, nor evaluated the spectral radiant power absorbed by the whole solution or its dependence on PAT concentration and the incident spectral radiant power at different depths. In all the cases, they studied the effect of UV incident dose at the surface of the reactor, measuring the incident intensity at the surface of the liquid with a radiometer, but without evaluating the absorbed energy, which is the parameter that really influences the reaction. In fact, the incident intensity at the surface is not the incident intensity at any other depth. They concluded that the time needed for PAT reduction is much higher than the time required for pathogen reduction and proposed the combination of UV irradiation with other juice-processing techniques, such as adsorption, filtration or addition of fructose.

So, the main aim of this work is to solve this lack of information, studying the degradation of PAT with a multi-wavelength emitting lamp of much higher power, in order to know if the time needed for germicidal reduction by the 254 nm wavelength is similar to the time needed for PAT reduction at all the wavelengths emitted by the lamp in the range where PAT absorbs energy. To this end, the emitting and absorption spectra must be compared and the absorbed energy and its dependence on the PAT concentration have to be calculated. Finally, a reaction mechanism, related with the absorbed energy, which fits the first-order kinetic, is proposed.

Depending on the variety and ripeness of the apples, the juice has a pH ranging approximately from 3.5 to 4.2, while the washing water pH is close to neutrality. Therefore, aqueous solutions of PAT at pH 4 and pH 7 were irradiated. The photodegradation of PAT added to a reconstituted apple juice from a concentrate was also studied.

2. Material and methods

2.1. Irradiation model

When a solution is irradiated with an electromagnetic radiation, part of the energy is absorbed. The ability to absorb radiation can be known by defining the absorbance as the logarithm of the ratio between the incident (P_{λ}^{0}) and the transmitted (P_{λ}) spectral radiant power at λ wavelength. Taking into account the base of the logarithm, two definitions are possible:

$$A_{10}(\lambda) = \log \frac{P_{\lambda}^{0}}{P_{\lambda}}; A_{e} = \ln \frac{P_{\lambda}^{0}}{P_{\lambda}}.$$
(1)

The spectrophotometers usually measure the absorbance as A_{10} , while the model equations use A_e . The relation between both terms is $A_e = A_{10} \cdot \ln 10$.

Lambert–Beer proposed a linear relation between the absorbance at a fixed wavelength and the concentration of the absorber substance

$$A(\lambda) = \varepsilon_{\lambda} \cdot C_A \cdot D_S \tag{2}$$

where $A(\lambda)$ is the absorbance at λ wavelength, ε_{λ} the molar absorption coefficient at the same wavelength, C_A the concentration of the absorber substance and D_S the path length of light through the solution.

Considering Lambert–Beer's law, the e-base absorbance, a plane photoreactor with a cylindrical lamp (Fig. 1) and a linear spherical emission model (Falguera et al. 2011b), the spectral radiant power reaching (P_{λ}) a specific point (*x*,*y*,*z*) inside the solution at each wavelength, from the spectral radiant power emitted at the same wavelength by a specific point of the lamp, can be obtained as:

$$A_{\lambda} = \ln \frac{P_{\lambda}(S) \cdot 4\pi D_0^2}{P_{\lambda}(x, y, z) \cdot 4\pi D^2} = \varepsilon_{\lambda} C_A D_S$$
(3)

$$P_{\lambda}(x,y,z) = P_{\lambda}(S) \frac{D_0^2}{D^2} \exp(-\varepsilon_{\lambda} C_A D_S) = P_{\lambda}(S) \frac{D_0^2}{D^2} \exp\left(-\varepsilon_{\lambda} C_A \frac{z}{\sin\beta}\right)$$
(4)

where $P_{\lambda}(S)$ is the incident spectral radiant power at λ wavelength at the reactor's surface from the specific point considered in the emitting lamp, being obtained as:

$$P_{\lambda}(S) = \frac{W_{\lambda}(L)/L}{4\pi D_0^2} dy_L$$
(5)

where $W_{\lambda}(L)$ is the total energy emitted by the whole lamp at λ wavelength, L is the lamp length, y_L is the coordinate that defines each specific point of the lamp and D_0 is the radius of the sphere defined by the considered emitting lamp point as the centre of the sphere and the point obtained as the intersection in the reactor's surface of the beam in his way to the considered point (x,y,z).

 D_0 and D_S can be obtained from the following equations:

$$D_0 = \frac{z_0}{\sin\beta} \tag{6}$$

$$\sin\beta = \frac{z_0 + z}{D} \tag{7}$$

$$D^{2} = (x - x_{0})^{2} + (y - y_{L})^{2} + (z + z_{0})^{2}$$
(8)

$$D_S = \frac{z}{\sin\beta}.$$
(9)

The incident spectral radiant energy for a specific depth of the reactor can be obtained integrating Eq. (4) for the whole length of the lamp, and all the *x* and *y* values for the specific *z* value:

$$P_{\lambda}(z) = \frac{W_{\lambda}(L)/L}{4\pi} \int_{x=0}^{x=Ay=By_{L}=y_{0}+L} \int_{y_{L}=y_{0}}^{x=Ay=By_{L}=y_{0}+L} \frac{\exp\left(-\varepsilon_{\lambda}C_{A}\frac{z}{\sin\beta}\right)}{D^{2}} dy_{L} dy dx$$
(10)

where *A* and *B* are the reactor's dimensions on the *x* and *y* axes.

A point (x',y',z') is considered at an infinitesimal distance from (x,y,z). The transmitted spectral radiant power through this infinitesimal path (d_S) is:

$$P_{\lambda}(x', y', z') = P_{\lambda}(x, y, z) \exp(-\varepsilon_{\lambda} C_A d_S)$$
(11)

being

$$d_{\rm S} = \frac{dz}{\sin\beta}.\tag{12}$$

In the case of dilute solutions, where the values of the product $\varepsilon_{\lambda} \cdot C$ are low, this equation can be simplified as:

$$P_{\lambda}(x',y',z') \approx P_{\lambda}(x,y,z)[1-\varepsilon_{\lambda}C_{A}d_{S}].$$
(13)

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