Degradation of sucralose present in Splenda® sweetener by TiO$_2$ photocatalysis assisted with photo-Fenton

Degradación de Sucralosa presente en el edulcorante Splenda® por fotocatálisis con TiO$_2$ asistida por foto-Fenton

A. Sandoval-González[1,*], A. Álvarez-Gallegos[2], J.A. Hernández[2], S. Silva-Martínez[2*]

[1]*Cátedras-CONACYT-Centro de Investigación y Desarrollo Tecnológico en Electroquímica S.C., Parque Tecnológico Querétaro, Sanfandila, 76703, Pedro Escobedo, Querétaro, México.*
[2]*Centro de Investigación en Ingeniería y Ciencias Aplicadas, Universidad Autónoma del Estado de Morelos, Av. Universidad 1001, Col. Chamilpa, Cuernavaca, Morelos C.P. 62209, México.*

Abstract

The pollutant legislation and its ecotoxicological dates recognize several water pollutants. However, there are others that have not yet been recognized as emerging organic pollutants because the damage that could be caused to the environment, especially in the water, is still unknown, like Splenda® sweetener. For example, it is interesting to pay attention on Splenda® because it is increasingly consumed worldwide today. Splenda® is one of the emerging pollutants that has not yet been formally declared as a pollutant, but there are some findings that show Splenda® as a water pollutant, specifically the sucralose molecule, due to persistence in the environment. In this work, the mineralization of Splenda®, in different initial concentrations (72 mg L^{-1}, 144 mg L^{-1} and 288 mg L^{-1}), was carried out by TiO$_2$ photocatalysis assisted with photo-Fenton. Splenda® mineralization is less than 58% by these processes when used separately; however, Splenda® was mineralized 62.8% (72 mg L^{-1}) 83.7% (144 mg L^{-1}) and 58.8% (288 mg L^{-1}) by TiO$_2$ photocatalysis assisted with photo-Fenton at 2 h, 5 h and 5 h, respectively. In all cases, the mineralization followed a pseudo-first-order reaction.

Keywords: Water, emerging contaminant, photo-Fenton, TiO$_2$ photocatalysis, sucralose- Splenda®.

Resumen

La legislación actual del agua reconoce a algunos compuestos orgánicos como contaminantes de la misma. No obstante, hay otros compuestos que no han sido reconocidos por la legislación, como los contaminantes emergentes, debido a que aún no se conoce el daño que pueden causar en el agua y el ambiente. Uno de ellos es la Splenda®, edulcorante artificial que se consume cada vez en mayor cantidad a nivel mundial. Existe información que demuestra que el Splenda® es un contaminante emergente del agua, por los iones cloruro presentes en la molécula de la sucralosa y por su persistencia en el agua. En este trabajo, se investigó la mineralización del Splenda® a diferentes concentraciones (72 mg L^{-1}, 144 mg L^{-1} y 288 mg L^{-1}) mediante fotocatálisis TiO$_2$ asistida con foto-Fenton. Cuando estos procesos actúan por separado la mineralización de Splenda® es inferior al 58%. Sin embrago, cuando se usan en conjunto, se mineraliza 62.8% (72 mg L^{-1}), 83.7% (144 mg L^{-1}) y 58.8% (288 mg L^{-1}), a las 2 h, 5 h y 5h, respectivamente. En todos los casos la mineralización siguió una reacción de pseudo-primer orden.

Palabras clave: Agua, contaminantes emergentes, foto-fenton, fotocatálisis TiO$_2$, Sucralosa- Splenda®.

1 Introduction

Splenda® is a brand of artificial sweetener that is increasingly consumed worldwide. Today, global pollution of water, air and soil is increasing due to anthropogenic activities to meet human needs.

There are different types of organic compounds that contaminate the environment such as pesticides, steroids, hormones, personal care products, industrial additives, artificial sweeteners, oils, medicines, alcohols, food, amongst some others. There are other compounds, some of these are known as emerging organic contaminants, present in drinking water (Fawell & Ong, 2012).

Emerging organic contaminants are compounds yet to be recognized for the contaminant legislation, meaning that the incidence, risk, and ecotoxicological dates are not available, and it is difficult to know their potential harm in humans, animals and plants (Hurtado *et al*., 2016; Carraro *et al*., 2018). Emerging organic contaminants do not need to stay long time in the environment to produce negative effects because they are constantly introduced into water bodies (mainly in wastewater), and their concentration is increased little by little, for this reason it is necessary to focus on their degradation or transformation to none contaminant molecules (Naidu *et al*., 2016).

Furthermore, the marketing bombards the human mind forcing them to use many products that could be dangerous for their body and for the environment, like Splenda® sweetener, where the main sweetener is sucralose. Sucralose is extremely persistent in the environment with a half-life in water of up to several years, depending on the pH and temperature (Lubick, 2008). According to Kylin, there is also the possibility that sucralose interferes with algae photosynthesis that may affect CO_2 absorption (Lubick 2008). In most cases, Splenda® is discarded in water bodies without molecule modification and because it has not been considered as contaminant, there are not studies that show the human damage after ingestion. Discarding wastewaters with Splenda® content into the environment may produce accumulation for several decades and could provoke changes in the aquatic life, plants and animals (Loos *et al*., 2009).

Splenda® is composed by sucralose, dextrose and maltodextrin. The sucralose (1,6-dichloro-1,6-dideoxy-*β*-D-fructofuranosyl-4-chloro-4-deoxy-*α*-D-galactopyranoside) is the principal sweetener of Splenda®. Sucralose is modified sucrose by removing hydroxyls and replacing them with Cl^- atoms. Chlorine atoms give sucralose stability under different conditions of pH and temperature for long periods of time (Grotz *et al*., 2012). Although, sucralose is claimed that does not interact with the human body system when consumed, only 15% is absorbed the rest is disposed in the wastewater by the urine and feces principally (Baird *et al*., 2000; Magnuson *et al*., 2017), persisting for a long time (Sharma *et al*., 2019). Dextrose and maltodextrin are not considered as contaminant molecules.

In spite of the fact that the Splenda® is regularized by Food Drugs Administration (FDA) as safe product (Sucralose-FDA, 1998; Grotz & Munro, 2009; Carocho *et al*., 2015), and sucralose is claimed to be very stable (Barndt & Jackson, 1990; Binns,

2003) even at high temperatures, there have been reported contrary information; for instance, Bannach *et al*., (2009), and De Oliveira *et al*., (2015) observed that sucralose produced chloropropanols and other related chlorinated compounds when baked after 100°C, such compounds are considered dangerous for human health. Similar observations were reported by others (Gallus *et al*., 2007; Wiklund *et al*., 2012; Dong *et al*., 2013). Hutchinson (1996) studied the stability of sucralose in hot buffered solutions (pH 3, 7 and 11), such solutions were heated between 100-180°C for 1 h, and found that the stability of sucralose decreased at 100°C and 140°C with pH increase, while no sucralose remained at 180°C at all pH levels. Hutchinson *et al*., (1999) indicated that the stability studies of sucralose demonstrate that in aqueous systems, sucralose will break down into 4-chlorogalactose and 1,6-dichlorofructose. In addition, Abou-Donia *et al*., (2008) and Schiffman *et al*., (2012), administered sucralose (from Splenda®) on male Sprague-Dawley rats over 12 weeks, using dosages based on doses approved by the FDA and the European Union. These authors found evidences that a 12-week administration of Splenda® exerted numerous adverse effects, including (1) reduction in beneficial fecal microflora, (2) increased fecal pH, and (3) enhanced expression levels of P-glycoprotein and two cytochrome metabolic enzymes, which are known to limit the bioavailability of orally administered drugs (Bannach *et al*., 2009; De Oliveira *et al*., 2015). Similarly, Grotz *et al*., (2012), found that sucralose at pH 4.4 shows high resistance to hydrolysis and can persist for up to five years, allowing bioconcentration in different organs and tissues of aquatic animals (Saucedo-Vence *et al*., 2017; Suez *et al*., 2014; Gardner *et al*., 2012). Other studies have shown that the presence of sucralose in water causes apoptosis, stress and even DNA modification in *Cyprinus carpio* (Saucedo-Vence *et al*., 2017; Heredia-García *et al*., 2019). On the other hand, Wood *et al*., (2000) and John *et al*., (2000) investigated the fate of sucralose when a dog and a rabbit were exposed to different doses; it was found that between 18 to 48% of sucralose was adsorbed by the body of both animals, while 22% was excreted in urine and 55% in feces, without finding sucralose unchanged. The presence of sucralose, between 49 to 2400 ng L^{-1} sucralose, has been reported in wastewater treated by conventional processes, confirming that sucralose cannot be eliminated after conventional treatment (Doummar & Aoun, 2018; Luo *et al*., 2019; Oppenheimer *et al*., 2012). Although Splenda contains

a low concentration of sucralose, it is not only present in Splenda® sweetener individual packets but also it is used in thousands of food, beverage and pharmaceutical products, which leads to an increase in its concentration in wastewater (Schiffman and Rother, 2013).

Under these premises, clearly Splenda® is an emerging water contaminant. Therefore, there is necessary to focus on alternatives for its molecule oxidation. Some of the methods used for the mineralization of contaminants in water are the advanced oxidation processes (AOP). AOP are physicochemical processes that can produced changes in the structure of chemical contaminants that cannot be observed with biological processes (Contreras-Bustos *et al.*, 2020). There have been shown that sucralose is poorly removed (Tran *et al.*, 2014) or is not eliminated during biological treatment processes even after prolonged time under different experimental conditions (Buerge *et al.*, 2009; Scheurer *et al.*, 2010; Inyang *et al.*, 2016). AOP involve the formation of a strong oxidant such as hydroxyl radicals, HO•, that reacts with organic contaminants in water (Muruganandham *et al.*, 2014). It is important to mention that the AOP do not produce changes of phase or concentration of the contaminant, thus the contaminant is completely destroyed (mineralized) without generation of contaminant mud. Amongst the AOP that produce HO• radicals are ozone (Kohli 2015), hydrogen peroxide (Wang *et al.*, 2016), Fenton reaction (Papoutsakis *et al.*, 2016), ultraviolet irradiation and photocatalysis with titanium dioxide (Benitez *et al.*, 2013; Eskandarian *et al.*, 2016; Gines-Palestino, *et al.*, 2018; Morones-Esquivel *et al.*, 2020), among others. It was reported that sucralose can be degraded by photo-Fenton and TiO_2 photocatalysis with the released of chloride ions (Calza *et al.*,

2013; Chen *et al.*, 2016); photo-Fenton process mainly promoted the formation of oxidized products and at a minor extent the cleavage of glycoside bond. Conversely, TiO_2 mediated transformation preceded through four parallel pathways: hydroxylation of the molecule, oxidation of the alcohol function, dichlorination and the cleavage of glycosidic bonds (Calza *et al.*, 2013). Regarding to the degradation of glucose, a study on the glucose photocatalytic conversion in aqueous suspension reported the formation of gluconic acid, traces of glucaric acid, glyceraldehyde arabinose, erythrose and formic acid as oxidation products when bare TiO_2 was used (Bellardita *et al.*, 2015). Also, according to Da Vià and coworkers, it is possible the selective conversion of glucose by TiO_2 under natural sunlight after 7 h exposure, proving the possibility of using TiO_2 as a photocatalyst to upgrade biomass derivatives selectively by tailoring the reaction conditions (Da Vià *et al.*, 2017). According to the literature, the studies were focused on the degradation of sucralose molecule or glucose molecule (Splenda® components) separately since such analytical reagents are used as model contaminants. Clearly, the oxidation of artificial sweeteners, such as Splenda®, represent an environmental challenge since their oxidation is much more complex than any of their analytical components separately. The aim of this work is to study the mineralization of Splenda® by TiO_2 photocatalysis assisted with photo-Fenton process. The pH of the solution and the load of TiO_2 are important influencing factors. The final samples treated by Fenton and photocatalysis were analyzed by high performance liquid chromatography. Splenda® functional groups were analyzed before and after treatment by the Fourier transform infrared spectroscopy.

Table 1. Molecular structures of the organic components of Splenda®.

Sucralose ($C_{12}H_{19}Cl_3O_8$)   Dextrose ($C_6H_{12}O_6$)   Maltodextrin ($C_6H_{10}O_5$)
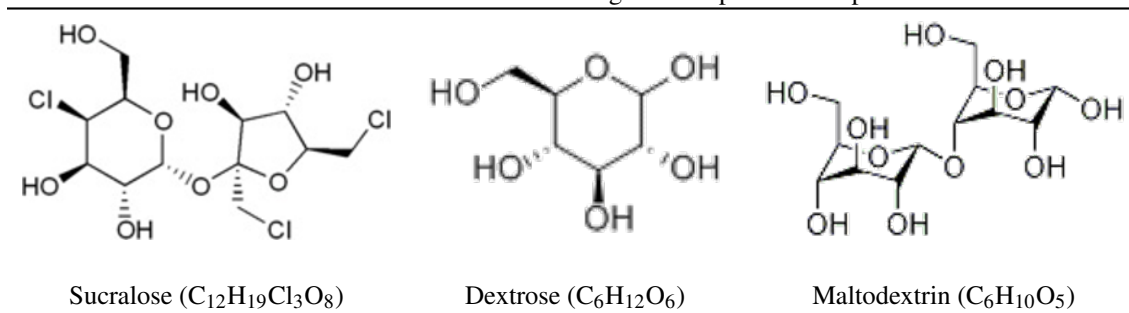
2 Materials and methods

2.1 Chemical reagents

Splenda® sweetener-pack was purchased in a supermarket (with a content of 50 individual packets with a serving size of 1g per packet). Table 1 shows the molecular structure of Splenda® components (Abou-Donia *et al.*, 2008). Titanium dioxide (TiO$_2$ 99%, Sigma-Aldrich), hydrogen peroxide (H$_2$O$_2$ 30%, Meyer), Total Organic Carbon (TOC, 0-20 mg L^{-1} range, HACH), sulfuric acid (H$_2$SO$_4$ 98%, J.T. Baker) and ferrous sulfate heptahydrate (FeSO$_4$·7H$_2$O 99%, Sigma-Aldrich) were analytical reagent grade and used as received. All aqueous solutions were prepared with distilled water.

2.2 Procedure

In typical experiments using a Pyrex glass, Splenda® content was dissolved in distilled water at different concentrations (72, 144 and 288 mg L^{-1}) at room temperature. Splenda® degradation was investigated using a slurry reactor and TiO$_2$ as photocatalyst (100-300 mg L^{-1}) at three pH values (2, 7 and 10) under UV light (UV lamp, shortwave filter, 1250 μW cm^{-2}, 254 nm). Also, the degradation of Splenda® was observed by photolysis using the same lamp UV irradiation. Samples were withdrawn at different intervals of time to observe the mineralization degree by measuring the TOC.

The degradation of Splenda® by Fenton process was carried out at pH 2 with the stoichiometry of hydrogen peroxide and 2.0x10^{-3} mol L^{-1} ferrous sulphate heptahydrate as catalyst. Previous studies (Silva & Lizama, 2009; Alvarez-Gallegos & Pletcher, 1999; Pignatello *et al.*, 2006) have shown that the concentration of 1.0 mmol L^{-1} of ferrous ions is enough to catalyze the hydrogen peroxide in the Fenton reaction to produce hydroxyl radicals in mildly acidic solutions (2 < pH < 3). Additions of H$_2$O$_2$ and Fe^{2+} were carried out in the experiments that lasted more than two hours to maintain the Fenton reaction. Finally, the degradation of Splenda® was also observed by the combination of TiO$_2$ photocatalysis with photo-Fenton (the same lamp was used for the photocatalysis with the specifications described above) process at pH 2.

2.3 Analytical methods

TOC was analyzed at time intervals using standard method and standard reagents (0-20 mg L^{-1} range, HACH) using previously filtered samples. Splenda® functional groups were analyzed before and after treatment by Fourier transform infrared spectroscopy (FTIR) using the Frontier Perkin Elmer Spectrometer equipment, data was acquired by the Perkin Elmer Spectrum software, using an ATR technique. The analysis was recorded in liquid phase, within range of 4000-600 cm^{-1}, and the spectra baseline corrected. Glucose and sucralose in aqueous solution was measured by the liquid chromatography (HPLC) method in an Agilent Technologies 1200 equipment, with a 1260 RID refractive index detector, with a RezexTM ROA-Organic Acid H$^+$ (8%) LC Column 150x7.8 (Phenomenex), with a 4.0 mg L^{-1} of linearity limit. Dextrose and sucralose were identified by the retention time of 6.12 and 7.91 min, respectively. The concentration was measured by the peak area.

3 Results and discussion

3.1 Oxidation of Splenda® by direct photolysis and TiO$_2$ photocatalysis

Splenda® degradation is studied by photolysis (only UV radiation) and photocatalysis with a fixed amount of TiO$_2$ (200 mg L^{-1}) at pH 2, 7 and 10, at room temperature. Negligible photochemical degradation was recorded at all pH levels, while Splenda® mineralization increased as pH increased with TiO$_2$ photocatalysis at 5 h reaction (as depicted in figure 1). This is attributed to changes in the catalyst surface charge affecting the adsorption/desorption processes due to changes in the basicity/acidity of the solution; in addition to the separation of electron-hole pairs generated on the catalyst surface under UV illumination where holes react with water molecules or with hydroxide ions, generating highly reactive OH$^\bullet$ radicals which in turn attack the Splenda® components that oxidize them (Reutergardh & Langphasuk, 1997). Based on the pH behavior, neutral pH was chosen to observe the variation of TiO$_2$ amount during Splenda® photocatalysis (figure 2). Figure 2, shows that the rate of Splenda® mineralization increases until a catalyst concentration of 200 mg L^{-1} is reached and then reduced again. The increase is associated to extra surface area at

higher catalyst load, which increases the absorption of photons, and also to the increased adsorption of Splenda® molecules on the catalyst surface. Conversely, the decrease in Splenda® mineralization after 200 mg L^{-1} TiO_2 may be explained to the shielding effect produced by the high amount of TiO_2 particles due to turbidity and an enhancement of UV light reflectance (Bizani *et al.*, 2006). These results clearly show poor Splenda® oxidation by photocatalysis; thus, the use of only one advanced oxidation process is not enough to solve the Splenda® contamination in wastewater. Based on these results, the Fenton process was used to observe if higher Splenda® oxidation could be attained than by photocatalysis.
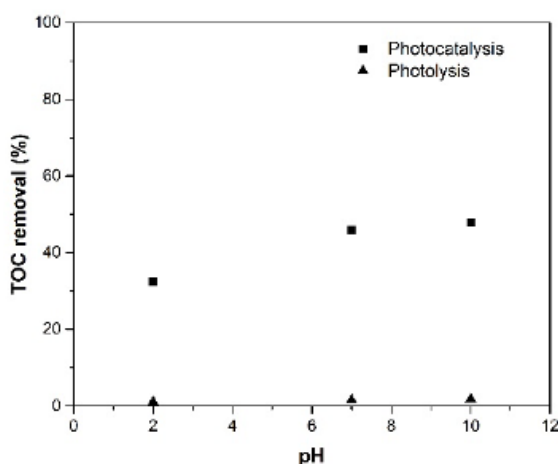
Fig. 1. TOC removal from 72 mg L^{-1} of Splenda® solution as function of pH at 5 h reaction: (■) 200 mg L^{-1} TiO_2 photocatalysis, and (▲) photolysis.
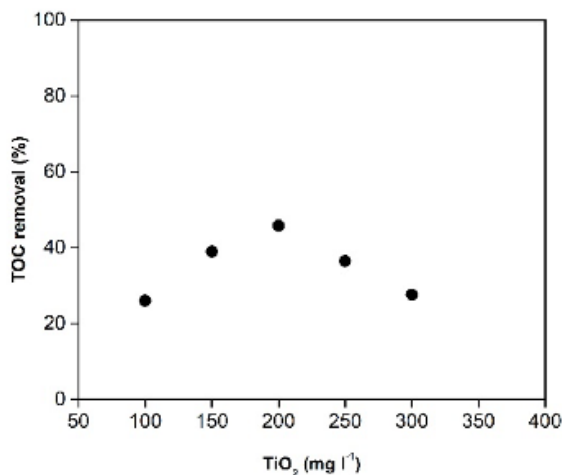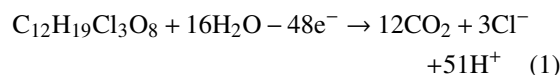

Fig. 2. TOC removal from 72 mg L^{-1} of Splenda® solution as function of catalysis load at neutral pH at 5 h reaction.
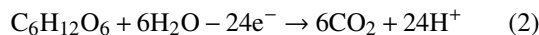
3.2 Splenda® oxidation by Fenton Process

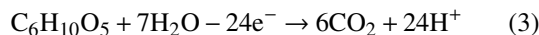The complete chemical oxidation of Splenda® involves 96 e^- according to the reactions (1) to (3):
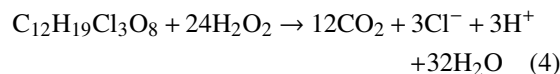
Sucralose:

$$C_{12}H_{19}Cl_3O_8 + 16H_2O - 48e^- \rightarrow 12CO_2 + 3Cl^-$$
$$+51H^+ \quad (1)$$

Dextrose:

$$C_6H_{12}O_6 + 6H_2O - 24e^- \rightarrow 6CO_2 + 24H^+ \quad (2)$$

Maltodextrin:

$$C_6H_{10}O_5 + 7H_2O - 24e^- \rightarrow 6CO_2 + 24H^+ \quad (3)$$
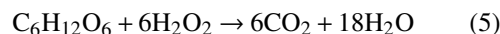
Thus, the stoichiometric conversion of 1 mol of sucralose, 1 mol of dextrose and 1 mol of maltodextrin (components of Splenda®) to CO_2 requires 24 mol, 12 mol and 12 mol of H_2O_2, respectively:
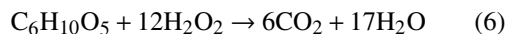
Sucralose:

$$C_{12}H_{19}Cl_3O_8 + 24H_2O_2 \rightarrow 12CO_2 + 3Cl^- + 3H^+$$
$$+32H_2O \quad (4)$$

Dextrose:

$$C_6H_{12}O_6 + 6H_2O_2 \rightarrow 6CO_2 + 18H_2O \quad (5)$$

Maltodextrin:

$$C_6H_{10}O_5 + 12H_2O_2 \rightarrow 6CO_2 + 17H_2O \quad (6)$$

Therefore, based on the reactions (4) to (6) and the content of the Splenda® components (1.2% sucralose, 95.8% dextrose and 3.0% maltodextrin) the total amount of H_2O_2 required to mineralize completely the Splenda® artificial sweetener is obtained. The performance of the Fenton process on Splenda® mineralization is depicted in figure 3. As can be seen, the initial TOC content of Splenda® decreases almost linearly during 5 h of reaction. Splenda® mineralization is attributed to the highly reactive OH• radicals produced by the ferrous ions which enhanced the oxidizing power of the hydrogen peroxide (Pignatello *et al.*, 2006). Negligible Splenda® oxidation was observed by hydrogen peroxide in the absence of iron ions. In addition, a similar Splenda® mineralization was obtained when a 50% more than the stoichiometry of the H_2O_2 concentration was used in the Fenton process; such results are in agreement with the stoichiometry calculated from reactions (4 - 6).
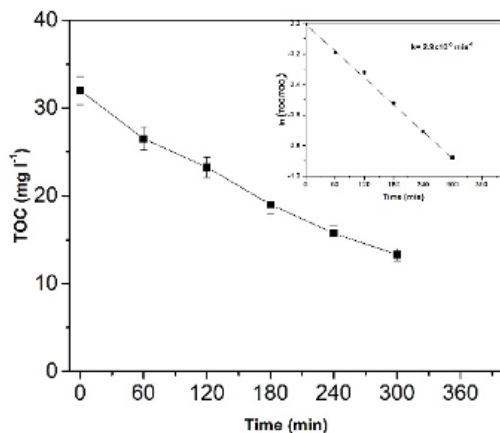
Fig. 3. Mineralization of 72 mg L^{-1} Splenda$^®$ at pH 2 by Fenton process (0.1 mol L^{-1} hydrogen peroxide and 2.0×10^{-3} mol L^{-1} ferrous sulphate heptahydrate).
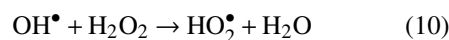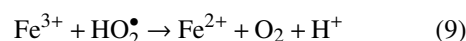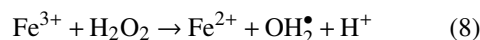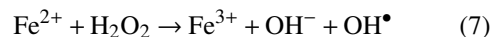
Nevertheless, Splenda$^®$ mineralization decreased when the stoichiometry of H_2O_2 was doubled which is attributed to the competing reaction between the H_2O_2 and the hydroxyl radicals (Pignatello *et al.*, 2006). The inset of figure 3 depicts a linear behavior of Splenda$^®$ mineralization by the Fenton process with a rate constant of 2.9×10^{-3} min^{-1} ($R^2 = 0.9978$) indicating a pseudo-first order reaction. These results clearly show that higher Splenda$^®$ mineralization is obtained by Fenton process (58%) than by photocatalysis (33%) at the same pH value (pH 2) and at 5 h reaction. Thus, the use of Fenton process or TiO$_2$ photocatalysis is not possible to achieve complete mineralization of Splenda$^®$. These results may be attributed to the fact that sucralose is an organochlorine compound and according to the literature it has been found that so far the Fenton process has not been effective in mineralizing this type of molecule (Chen *et al.*, 2016; Bigda, 1995), in addition to the persistence of sucralose in the medium carrier, as mentioned in the introduction.

## 3.3 Splenda$^®$ oxidation by the combination of TiO$_2$ photocatalysis and Fenton process

Based on the results obtained during Splenda$^®$ degradation by photocatalysis and Fenton processes used separately, a set of experiments were carried out to observed the combined effects of both processes used simultaneously, as shown in figure 5. Figure 5a reports the mineralization of Splenda$^®$ as function

of its initial concentration by the combination of the Fenton (photo) process and TiO$_2$ photocatalysis at pH 2. It is observed that the mineralization time (2 h) is greatly reduced in the oxidation reaction of 72 mg L^{-1} of Splenda$^®$ with a mineralization yield of 63.1%. Clearly, these results show that the combination of these two processes produced better results than those obtained by each process used separately. Such behavior is attributed to the higher amount of $^•$OH radicals produced by photo-Fenton and photocatalysis, as depicted in the schematic representation of the mechanism for the mineralization of Splenda$^®$, observed in figure 4.

It is generally accepted that the active oxidation in dark and in light of Fenton systems is the hydroxyl radical, OH$^•$, in which its origin in the dark is the free radical chain mechanism (Pignatello *et al.*, 1999; Walling 1975), as depicted in reactions (7-10).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + OH^• \qquad (7)$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + OH_2^• + H^+ \qquad (8)$$

$$Fe^{3+} + HO_2^• \rightarrow Fe^{2+} + O_2 + H^+ \qquad (9)$$

$$OH^• + H_2O_2 \rightarrow HO_2^• + H_2O \qquad (10)$$

In light, the rate of OH$^•$ formation is increased by photo reactions of H_2O_2 ($\lambda <\sim 320$ nm) (Barb *et al.*, 1951), reaction (11), and/or photo reducing Fe(III) product in the near UV region in the presence of H_2O_2 (Huston & Pignatello, 1999), reaction (12), that produce OH$^•$ directly or regenerate Fe(II) to feed into reaction (7), as shown in the photo-Fenton mechanism (Figure 4).

$$H_2O_2 \xrightarrow{h\nu} 2OH^• \qquad (11)$$

$$Fe^{3+}(OH^-) \xrightarrow{h\nu} Fe^{2+} + OH^• \qquad (12)$$
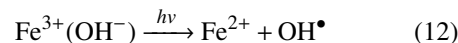
In addition, TiO$_2$ photocatalyst upon illumination with UV light ($\lambda < 380$ nm) produces excited high-energy states of electron, e_{CB}^-, (in the conduction band, CB) and hole, h_{VB}^+, (in the valence band, VB) pairs, reaction (13), capable of initiating chemical reactions (Gaya & Abdullah, 2008), as shown in the mechanism of TiO$_2$ photocatalysis (Figure 4).

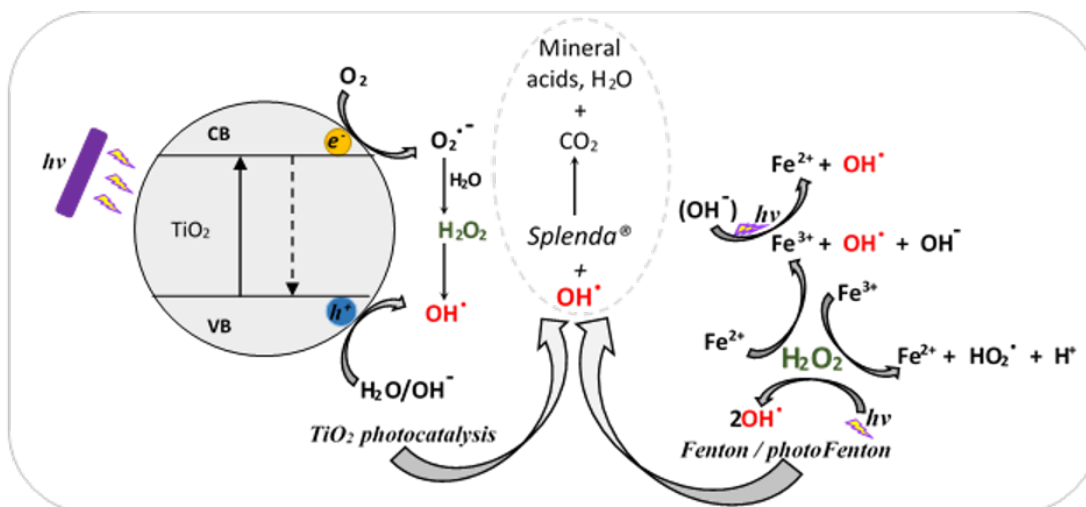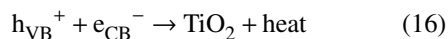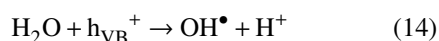$$TiO_2 + h\nu \rightarrow e_{CB}^- + h_{VB}^+ \qquad (13)$$
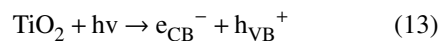
Fig. 4. Schematic representation of the mechanism for the mineralization of Splenda® by combining the process of TiO$_2$ photocatalysis with photo-Fenton.

The holes are responsible for the degradation of organic compounds and can react with water (reaction (14) or hydroxyl ions (reaction (15) to produce hydroxyl radicals; though, the recombination of the (e_{CB}^-/h_{VB}^+) pair (reaction (16) and the reduction of OH$^\bullet$ radicals (reaction (17)) produce large inefficiencies in the photocatalytic process (Choi *et al*., 1994). Thus, combining the photocatalysis with the photo-Fenton reaction (Figure 4), such inefficiencies may not affect the degradation process in great extent. Hence, a higher effective accumulation of holes and OH$^\bullet$ from reactions above is feasible, leading to much higher overall efficiency than photocatalysis or photo-Fenton separately.

$$H_2O + h_{VB}^+ \rightarrow OH^\bullet + H^+ \tag{14}$$

$$OH^- + h_{VB}^+ \rightarrow OH^\bullet \tag{15}$$

$$h_{VB}^+ + e_{CB}^- \rightarrow TiO_2 + heat \tag{16}$$

$$OH^\bullet + e_{CB}^- \rightarrow OH^- \tag{17}$$

The oxidation of organic pollutants (R-H) can take place by a direct reaction with holes (reaction (18)) or indirectly via hydroxyl radicals (reaction (19)) or free organic radicals (reaction (20)) (Fujishima *et al*., 2000):
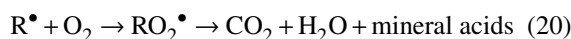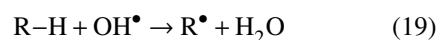
$$R{-}H + h_{VB}^+ \rightarrow R^{+\bullet} + degradation\ products \tag{18}$$

$$R{-}H + OH^\bullet \rightarrow R^\bullet + H_2O \tag{19}$$

$$R^\bullet + O_2 \rightarrow RO_2^\bullet \rightarrow CO_2 + H_2O + mineral\ acids \tag{20}$$

Figure 5a also reports that 83.5% mineralization of Splenda® is obtained at ~5.0 h when Splenda® concentration is double (144 mg L^{-1}). Nevertheless, an increase in its concentration to 288 mg L^{-1}, only 58.98% of mineralization is obtained at 5 h; such percentage is still much higher than those acquired by the same processes used separately even a much lower initial Splenda® concentration. Figure 5b shows the linear behavior of all Splenda® concentrations tested during the combined oxidation processes indicating that the mineralization is a pseudo-first order reaction, as depicted in table 2. Table 2 summarizes the values of the pseudo-first-order reaction rate constant (k), the correlation coefficient (R^2), and the mineralization percentage of Splenda® at different initial concentrations. The oxidation rate constant decreases as the concentration of Splenda® increases due to a decrease of the reaction rate, which is attributed to a decrease in the number of photons and TiO$_2$ particles available, since the number of organic molecules of Splenda® adsorbed onto the catalyst may block catalyst activation. This behavior is in agreement with the observations of other authors (Zhu *et al*., 2012; Neren and Yilmaz, 2008).

Table 2. Parameters obtained from Splenda$^{®}$ oxidation by the combined Fenton (photo) and TiO$_2$ photocatalysis advanced oxidation processes.

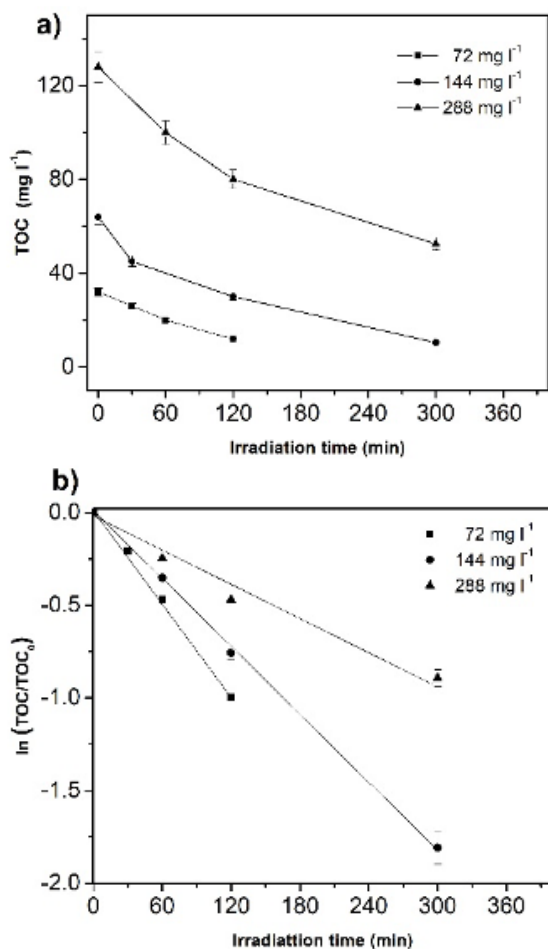| Splenda$^{®}$ (mg L^{-1})	k (min^{-1})	R^2	Mineralization (%)
72	0.0084	0.9977	63.1 ± 5
144	0.0060	0.9981	83.8 ± 5
288	0.0029	0.9821	59 ± 5

Fig. 5. TOC decrease as a function of the initial concentrations of Splenda$^{®}$ during the combined oxidation processes, Fenton (stoichiometry amount of hydrogen peroxide and 2.0x10^{-3} mol L^{-1} ferrous sulphate heptahydrate) and PC (pH 2 and 200 mg L^{-1} TiO$_2$): a) TOC decay with time, and b) irradiation time vs ln(TOC/TOC$_0$).

These results show that complete Splenda$^{®}$ mineralization can only be acquired by the combination of TiO$_2$ photocatalysis and photo-Fenton process. It is clear that greater mineralization is obtained by combining these two processes, but a longer reaction time is required as concentrations of Splenda$^{®}$ increase to obtain complete mineralization. Based on the degradation yields of Splenda$^{®}$ obtained separately by Fenton and PC, the Fenton process assisted with UV light (photo-Fenton) improved the photocatalytic degradation process.

These advanced oxidation processes were used because some studies have shown that the presence of light in the Fenton process increases the mineralization effectiveness of the polluting organic matter. Also, in the case of photocatalysis, the light photoexcites the semiconductor catalyst in the presence of oxygen causing the oxidation of Splenda$^{®}$ (Glaze *et al.*, 1993). In these two processes when sucralose is fully mineralized, chloride ions are released to the medium, when complete mineralization of sucralose does not occur, compounds that are harmful to humans, such as chloromethane (CH$_3$Cl) can be formed (Chen *et al.*, 2016).

3.3.1 FT-IR spectra

Splenda$^{®}$ functional groups are analyzed before and after treatment by Fourier transform infrared spectroscopy (FT-IR). FT-IR spectra of Splenda$^{®}$ solutions without and with treatment by the combined processes are shown in figure 6. The FT-IR spectra of the emerging organic pollutant are labeled as concentration of untreated Splenda$^{®}$ solutions (mg L^{-1}): a) 72, b) 144, and c) 288, and concentration of treated Splenda$^{®}$ solutions (mg L^{-1}): a') 72, b') 144, and c') 288. Splenda$^{®}$ untreated sample was used as reference, along with the spectra of the Splenda$^{®}$ oxidation at different concentrations. All of them show broad and intense bands around 3566-3000 cm^{-1} corresponding to the O-H stretching vibrations of the water molecules and hydrogen-bonded hydroxyl groups (Max & Chapados 2009). The spectra a), b), and c), for Splenda$^{®}$ untreated solutions, show peaks from 3725 to 2947 cm^{-1}, 1632, 1623, 1640 cm^{-1} corresponding to the OH stretching modes (Smrčková *et al.*, 2013; Santiago-Adame *et al.*, 2015), at 2920, 2917 and 2848 cm^{-1} is found CH stretching modes, at 1700 (1721, 1730, 1739) (Smrčková *et al.*,

2013; Loret *et al.*, 2005; Tabani *et al.*, 2015) and near 1000 cm^{-1} (1000 cm^{-1} slightly moved, should be at 1002, 996 and 994 cm^{-1}) corresponding to the CO stretching modes (Paulino *et al.*, 2011), at 1435 cm^{-1} (slightly moved, should be at 1430 cm^{-1}) C-C stretching modes, at 1480 cm^{-1} CH$_2$ deformation and at 1361 cm^{-1} CH rocking modes. The spectra, for the Splenda® oxidation at different concentration: a') 72 mg L^{-1}, b') 144 mg L^{-1} and c') 288 mg L^{-1}, show very well defined O-H stretching vibration from 3600 to 3000 cm^{-1}, due to the presence of water in spectra a') and b'). The OH modes are not well defined in spectrum c'), it might be attributed to an incomplete oxidation of Splenda®. Around of 1619, 1617 and 1614 cm^{-1} (for a', b', and c' spectra) it is observed a signal that could be attributed to the CO or C-C vibrations, but in the spectrum c' it is not observed easily. Finally, in all three spectra at 1458 cm^{-1} and 1200-700 cm^{-1} there are peaks that may correspond to the CO, COH stretching and CH bending vibrations. In addition, it is observed that when the Splenda® concentration is high; the FT-IR signals are not well defined.

3.3.2 HPLC

According to HPLC analysis, complete mineralization of sucralose, component of Splenda®, was achieved in all the concentrations of Splenda® samples treated with photocatalysis assisted by photo-Fenton. Nevertheless, the mineralization of dextrose was 61.2% (72 mg L^{-1} initial Splenda® content), 82.9% (144 mg L^{-1} initial Splenda® content) and 57.0% (288 mg L^{-1} initial Splenda® content) at 2 h, 5 h and 5 h treatment, respectively.

Conclusions

In summary, the Splenda® might be an emerging contaminant of water that is not very easy to reach complete mineralization by the use of process of Fenton or photocatalysis separately. It is necessary to use the combination of these two advanced oxidation processes to increase the mineralization as demonstrated in this study. Splenda® mineralization is not complete by using either Fenton or the photocatalytic process since it is only removed less than 58% of TOC, approximately by any of these processes used separately. In addition, when the two processes are used simultaneously under certain conditions, sucralose was successfully mineralized, but Splenda® in different initial concentrations (72 mg L^{-1}, 144 mg L^{-1} and 288 mg L^{-1}) showed mineralization yields of 63.1%, 83.8% and 59% at 2 h, 5 h and 5 h, at its corresponding initial concentration, respectively. According to the mechanism of reaction for photo-Fenton and TiO$_2$ photocatalysis, Splenda® oxidation is found to be a pseudo-first-order reaction. Therefore, this study shows that sucralose can be mineralized with TiO$_2$ photocatalysis assisted by photo-Fenton, in agreement with the findings of other workers that studied the oxidation of sucralose as model pollutant. However, the rest of the components of Splenda® (considered as non-polluting molecules) are yet to be fully mineralized.

Acknowledgements

References

Abou-Donia, M. B., El-Masry E. M., Abdel-Rahman A. A., McLendon R. E., & Schiffman S. (2008). Splenda alters gut microflora and increases intestinal p-glycoprotein and cytochrome P-450 in male rats. *Journal of Toxicology and Environmental Health Part A 71*, 1415-1429. https://doi.org/10.1080/1528739080 2328630

Alvarez-Gallegos, A., & Pletcher, D. (1999). The removal of low level organics via hydrogen peroxide formed in a reticulated vitreous carbon cathode cell. Part 2: The removal of phenols and related compounds from aqueous effluents. *Electrochimica Acta 44*, 2483-2492. https://doi.org/10.1016/S0013-4686(98)003 71-5

Baird, I. M., Shephard, N. W., Merritt, R. J., & Hildick-Smith, G. (2000). Repeated dose study of sucralose tolerance in human subjects. *Food Chemical Toxicology l 38*, S123-S129. https://doi.org/10.1016/S0278-6915(00)000 35-1

Bannach, G., Almeida, R. R., Lacerda, L. G., Schnitzler, E., & Ionashiro, M. (2009).

Thermal stability and thermal decomposition of sucralose. *Eclética Química 34*, 21-22. https://doi.org/10.1590/S0100-46702009000400002

Barb, W.G., Baxendale, J. H., George, P., Hargrave, K. R. (1949). Reactions of ferrous and ferric ions with hydrogen peroxide. *Nature 163*, 692-694. https://doi.org/10.1038/163692a0

Barndt, R. L., & Jackson, G. (1990). Stability of sucralose in baked goods. *Food Technology 44*, 62-67. https://www.cabdirect.org/cabdirect/abstract/19900396651

Bellardita M., García-López, E. I., Marcì, G., Megn, B., Pomilla, F. R., Palmisano, L. (2015). Photocatalytic conversion of glucose in aqueous suspensions of heteropolyacid-TiO$_2$. *The Royal Society Chemistry Advances 5*, 59037-59047. https://doi.org/10.1039/C5RA09894G

Benitez, F. J., Acero, J. L., Real, F. J., Roldan, G., Rodriguez, E. (2013). Photolysis of model emerging contaminants in ultra-pure water: Kinetics, by-products formation and degradation pathways. *Water Research 47*, 870-880. https://doi.org/10.1016/j.watres.2012.11.016

Bigda, R. J. (1995). Consider Fenton's chemistry for wastewater treatment. *Chemical Engineering and Processing: Process Intensification 91*, 62-66. https://www.semanticscholar.org/paper/Consider-Fenton's-chemistry-for-wastewater-Bigda/ddb7c0d78f4bfe1a0a3e4b4284722ba3e2586a79

Binns, N. M. (2003). Sucralose-all sweetness and light. *British Nutrition Foundation Nutrition Bulletin 28*, 53-58. https://doi.org/10.1046/j.1467-3010.2003.00307.x

Bizani, E., Fytianos, K., Poulios, I., Tsiridis, V. (2006). Photocatalytic decolourization and degradation of dye solutions and wastewaters in the presence of titanium dioxide. *Journal of Hazardous Materials 136*, 85-94. https://doi.org/10.1016/j.jhazmat.2005.11.017

Buerge, I. J., Buser, H. R., Kahle, M., Müller, M. D., Poiger, T. (2009). Ubiquitous occurrence of the artificial sweetener acesulfame in the aquatic environment: an ideal chemical marker of domestic wastewater in groundwater. *Environmental Science & Technology 43*, 4381-4385. https://doi.org/10.1021/es900126x

Calza, P., Sakkas, V. A., Medana, C., Vlachou, A. D., Dal Bello, F., Albanis, T. A. (2013). Chemometric assessment and investigation of mechanism involved in photo-Fenton and TiO$_2$ photocatalytic degradation of the artificial sweetener sucralose in aqueous media. *Applied Catalysis B 129*, 71-79. https://doi.org/10.1016/j.apcatb.2012.08.043

Carocho, M., Morales, P., Ferreira, I. C. F. R. (2015). Natural food additives: Quo vadis? *Trends Food Science Technology 45*, 284-295. https://doi.org/10.1016/j.tifs.2015.06.007

Carraro, P. M., Benzaquén T. B., Alfan, O. M., Oliva, M. I., Eimer, G. A. (2018). Advanced oxidation technologies to remove endocrine disruptors in water effluents based on zinc supported mesoporous catalysts. *Revista Mexicana de Ingeniería Química 17*, 1135-1146. https://doi.org/10.24275/uam/izt/dcbi/revmexingquim/2018v17n3/Carraro

Chen, S., Zhang, H., Li, S. (2016). The Tenth International Conference on Waste Management and Technology (ICWMT). Investigation of mechanism involved in TiO$_2$ and Photo-Fenton photocatalytic degradation of emerging contaminant sucralose in aqueous media. *Procedia Environmental Science 3*, 753-757. https://doi.org/10.1016/j.proenv.2016.02.064

Choi, W., Termin, A., Hoffmann, M. R. (1994). The role of metal ion dopants in quantum-sized TiO$_2$: correlation between photoreactivity and charge carrier recombination dynamics. *The Journal of Physical Chemistry 98*, 13669-13679. http://pubs.acs.org/doi/abs/10.1021/j100102a038

Contreras-Bustos, R., Cárdenas-Mijangos, J., Dector-Espinoza, A., Rodríguez-García, A., Montoya-Herrera, L., Jiménez-Becerril, J. (2020). Treatment of wastewater fromthe petrochemical industryby chemical Fenton process. *Revista Mexicana de Ingeniería Química 19*, 523-532. https://doi.org/10.24275/rmiq/IA637

Da Già, L., Recchi, C., Gonzalez-Yañez, E. O., Davies, T. E., Lopez-Sanchez, J. A. (2017). Visible light selective photocatalytic conversion of glucose by TiO$_2$. *Applied Catalysis B: Environmental 202*, 281-288. https://doi.org/10.1016/j.apcatb.2016.08.035

De Oliveira, D. N., de Menezes, M., Catharino, R. R. (2015). Thermal degradation of sucralose: a combination of analytical methods to determine stability and chlorinated byproducts. *Scientific Reports 5*, 9598. https://doi.org/10.1038/srep09598

Dong, S., Liu, G., Zhang, B., Gao, L., Zheng, M. (2013). Formation of polychlorinated naphthalenes during the heating of cooking oil in the presence of high amounts of sucralose. *Food control 32*, 1-5. https://doi.org/10.1016/j.foodcont.2012.11.001

Doummar, J., Aoun, M. (2018). Assessment of the origin and transport of four selected emerging micropollutants sucralose, Acelsulfame-K, gemfibrozil, and iohexol in a karst spring during a multi-event spring response. *Journal of Contaminant Hydrology 215*, 11-20. https://doi.org/10.1016/j.jconhyd.2018.06.003

Eskandarian, M. R., Choi, H., Fazli, M., Rasoulifard, M. H. (2016). Effect of UV-LED wavelengths on direct photolytic and TiO$_2$ photocatalytic degradation of emerging contaminants in water. *Chemical Engineering Journal 300*, 414-422. https://doi.org/10.1016/j.cej.2016.05.049

Fawell, J., Ong, C. N. (2012). Emerging contaminants and the implications for drinking water. *International Journal of Water Resources Development 28*, 247-263. https://doi.org/10.1080/07900627.2012.672394

Fujishima, A., Rao, T. N., Tryk, D. A. (2000). Titanium dioxide photocatalysis. *Journal of Photochemistry and Photobiology C: Photochemistry Reviews 1*, 1-21

Gallus, S., Scotti, L., Negri, E., Talamini, R., Franceschi, S., Montella, M., Giacosa, A., Maso, L. D., La Vecchia, C. (2007). Artificial sweeteners and cancer risk in a network of case-control studies. *Annals of Oncology 18*, 40-44.

https://doi.org/10.1093/annonc/mdl346

Gardner, C., Wylie-Rosett, J., Gidding, S. S., Steffen, L. M., Johnson, R. K., Reader, D., Lichtenstein, A. H. (2012). Nonnutritive Sweeteners: Current use and health perspectives. *Diabetes Care 35*, 1798-1808. https://doi.org/10.2337/dc12-9002

Gaya, U. I., Abdullah, A. H. (2008). Heterogeneous photocatalytic degradation of organic contaminants over titanium dioxide: A review of fundamentals, progress and problems. *Journal of Photochemistry and Photobiology C: Photochemistry Reviews 9*, 1-12. https://doi.org/10.1016/j.jphotochemrev.2007.12.003

Gines-Palestino, R.S., Oropeza- De la Rosa, E., Montalvo-Romero, C., Cantú-Lozano D. (2018). Rheokinetic and effectiveness during the phenol removal in mescal vinasses with a rotary disk photocatalytic reactor (RDPR). *Revista Mexicana de Ingeniería Química 19*, 639-652. https://doi.org/10.24275/rmiq/Cat673

Glaze, W. H., Kenneke, J. F., Ferry, J. L. (1993). Chlorinated by products from the TiO$_2$ - mediated photodegradation of trichloroethylene and tetrachloroethylene in water. *Environmental Science & Technology 27*, 177-184. https://doi.org/10.1021/es00038a021

Grotz, V. L., Molinary, S., Peterson, R. C. (2012). *Sucralose in Alternative Sweeteners* (ed. L. O'Brien Nabors). CRC Press Taylor and Francis Group, Boca Raton.

Grotz, V. L., Munro, I. C. (2009). An overview of the safety of sucralose. *Regulatory Toxicology and Pharmacology 55*, 1-5. https://doi.org/10.1016/j.yrtph.2009.05.011

Heredia-García, G., Gómez-Oliván, L. M., Orozco-Hernández, J. M., Luja-Mondragón, M., Islas-Flores, H., SanJuan-Reyes, N., Galar-Martínez, Ma, García-Medina, S., Dublán-García, O. (2019). Alterations to DNA, apoptosis and oxidative damage induced by sucralose in blood cells of *Cyprinus carpio*. *Science of the Total Environment 692*, 411-421. https://doi.org/10.1016/j.scitotenv.2019.07.165

Hurtado, C., Domínguez, C., Pérez-Babace, L., Cañameras, N., Comas, J., Bayona, J. M. (2016). Estimate of uptake and translocation of emerging organic contaminants from irrigation water concentration in lettuce grown under controlled conditions. *Journal of Hazardous Materials 305*,139-148. https://doi.org/10.1016/j.jhazmat.2015.11.039

Huston, P. L., Pignatello, J. J. (1999). Degradation of selected pesticide active ingredients and commercial formulations in water by the photo-assisted Fenton reaction. *Water Research 33*, 1238-1246. https://doi.org/10.1021/es980969b

Hutchinson, S. A. (1996). The effect of pH, temperature and reactants on the thermal and non-thermal degradation of the high-intensity sweeteners: Alitame and sucralose. Ph.D. Dissertation, Rutgers University, New Brunswick, New Jersey.

Hutchinson, S. A., Ho, G. S., Ho, C. T. (1999). Stability and degradation of the high-intensity sweeteners: Aspartame, Alitame, and Sucralose. *Food Reviews International 15*, 249-261. https://doi.org/10.1080/87559129909541189

Inyang, M., Flowers, R., McAvoy, D., Dickenson, E. (2016). Biotransformation of trace organic compounds by activated sludge from a biological nutrient removal treatment system. *Bioresource Technology 216*, 778-784. https://doi:10.1016/j.biortech.2016.05.124

John, B. A., Wood, S. G., Hawkins, D. R. (2000). The pharmacokinetics and metabolism of sucralose in the rabbit. *Food and Chemical Toxicology 38*, S111-S113. https://doi.org/10.1016/S0278-6915(00)00033-8

Kohli, R. (2015). Chapter 2-UV-Ozone cleaning for removal of surface contaminants. Development in surface contamination and cleaning. Development in surface Contamination and Cleaning. *William Andrew Publishing 71-104*. https://doi.org/10.1016/B978-0-323-29961-9.00002-8

Loos, R., Gawlik, B. M., Boettcher, K., Locoro, G., Contini, S., Bidoglio, G. (2009). Sucralose screening in European surface waters using a solid-phase extraction-liquid chromatography-triple quadrupole mass spectrometry method. *Journal of Chromatography A 1216*, 1126-1131. https://doi.org/10.1016/j.chroma.2008.12.048

Loret, C., Schumm, S., Pudney, P. D. A., Frith, W. J., Fryer, P. J. (2005). Phase separation and molecular weight fractionation behaviour of maltodextrin/agarose mixtures. *Food Hydrocolloids 19*, 557-565. https://doi.org/10.1016/j.foodhyd.2004.10.030

Lubick, N. (May 1, 2008). Artificial sweetener persists in the environment. *Environmental Science & Technology 3125*. https://pubs.acs.org/sharingguidelines for options on how to legitimately share published articles.

Luo, J., Zhang, Q., Cao, M., Wu, L., Cao, J., Fang, F., Li, C., Xue, Z., Feng, Q. (2019). Ecotoxicity and environmental fates of newly recognized contaminants-artificial sweeteners: A review. *Science of The Total Environment 653*, 1149-1160. https://doi.org/10.1016/j.scitotenv.2018.10.445

Magnuson, B. A., Roberts, A., Nestmann, E. R. (2017). Critical review of the current literature on the safety of sucralose. *Food Chemical Toxicology 106*, 324-355. https://doi.org/10.1016/j.fct.2017.05.047

Max, J. J., Chapados, C. (2009). Isotope effects in liquid water by infrared spectroscopy. III. H2O and D_2O spectra from 6000 to 0 cm^{-1}. *The Journal of Chemical Physics 13*, 184505. https://doi.org/10.1063/1.3258646

Morones-Esquivel, M. M., Núñez-Núñez, C. M., González-Burciaga, L. A., Hernández-Mendoza, J. L., Osorio-Revilla, G. I., Proal-Nájera, J. B. (2020). Kinetics and statistical approach for 2,5-dichlorophenol degradation in short reaction times by solar TiO_2/glass photocatalysis. *Revista Mexicana de Ingeniería Química 19*, 555-568. https://doi.org/10.24275/rmiq/Cat732

Muruganandham, M., Suri, R. P. S., Jafari, S., Sillanpää, M., Lee, G. J., Wu, J. J., Swaminathan, M. (2014). Review article: Recent development in homogeneous advanced oxidation processes for water and wastewater

treatment. *International Journal of Photoenergy 2014*, 1-21. https://doi.org/10.1155/2014/821674

Naidu, R., Arias Espana, V. A., Liu, Y., Jit, J. (2016). Emerging contaminants in the environment: Risk-based analysis for better management. *Chemosphere 154*, 350-357. https://doi.org/10.1016/j.chemosphere.2016.03.068

Oppenheimer, J. A., Badruzzaman, M., Jacangelo, J. G. (2012). Differentiating sources of anthropogenic loading to impaired water bodies utilizing ratios of sucralose and other microconstituents. *Water Resource 46*, 5904-5916. https://doi.org/10.1016/j.watres.2012.07.060

Papoutsakis, S., Pulgarin, C., Oller, I., Sánchez-Moreno, R., Malato, S. (2016). Enhancement of the Fenton and photo-Fenton processes by components found in wastewater from the industrial processing of natural products: The possibilities of cork boiling wastewater reuse. *Chemical Engineering Journal 304*, 890-896. https://doi.org/10.1016/j.cej.2016.07.021

Paulino, A. T., Fajardo, A. R., Junior, A. P., Muniz, E. C., Tambourgi, E. B. (2011). Two-step synthesis and properties of a magnetic-field-sensitive modified moltodextrin-based hydrogel. *Polymer International 60*, 1324-1333. https://doi.org/10.1002/pi.3084

Pignatello, J. J., Liu, D., Huston, P. (1999). Evidence for an additional oxidant in the photoassisted Fenton reaction. *Environmental Science & Technology 33*, 1832-1839. https://doi.org/10.1021/es980969b

Pignatello, J. J., Oliveros, E., MacKay, A. (2006). Advanced oxidation processes for organic contaminant destruction based on the fenton reaction and related chemistry. *Critical Reviews in Environmental Science and Technology 36*, 1-84. https://doi.org/10.1080/10643380500326564

Reutergardh, L. B., Langphasuk, M. (1997). Photocatalytic decolourization of reactive azo dye: a comparison between TiO_2 and CdS photocatalysis. *Chemosphere 35*, 585-596. https://doi.org/10.1016/S0045-6535(97)00122-7

Santiago-Adame, R., Medina-Torres, L., Gallegos-Infante, J. A., Calderas, F., González-Laredo, R. F., Rocha-Guzmán, N. E., Ochoa-Martínez, L. A., Bernad-Bernad, M. J. (2015). Spray drying-microencapsulation of cinnamon infusions (*Cinnamomun zeylanicum*) with maltodextrin. *LWT-Food Science and Technology 64*, 571-577. https://doi.org/10.1016/j.lwt.2015.06.020

Saucedo-Vence, K., Elizalde-Velázquez, A., Dublán-Garcia, O., Galar-Martínez, M., Islas-Flores, H., SanJuan-Reyes, N., García-Medina, S., Hernández-Navarro, M. D., Gómez-Olivan, L. M. (2017). Toxicological hazard induced by sucralose to environmentally relevant concentrations in common carp (*Cyprinus carpio*). *The Science of The Total Environment 575*, 47-357. https://doi.org/10.1016/j.scitotenv.2016.09.230

Scheurer, M., Stork, F. R., Brauch, H. J., Lange, F. T. (2010). Performance of conventional multi-barrier drinking water treatment plants for the removal of four artificial sweeteners. *Water Research 44*, 573-3584. http://dx.doi.org/10.1016/j.apcatb.2016.08.035

Schiffman, S. S., Abou-Donia, M. B. (2012). Sucralose revisited: Rebuttal of two papers about Splenda safety. *Regulatory Toxicology and Pharmacology 63*, 505-508. https://doi.10.1016/j.yrtph.2012.05.002

Schiffman, S.S., Rother, K. I. (2013). Sucralose, a synthetic organochlorine sweetener: overview of biological issues. *Journal of toxicology and Environmental Health Part B 16*, 399-451. https://doi.10.1080/10937404.2013.842523

Sharma, B. M., Bečanová, J., Scheringer, M., Sharma, A., Bharat, G. K., Whitehead, P. G., Klánová, J., Nizzetto, L. (2019). Health and ecological risk assessment of emerging contaminants (pharmaceuticals, personal care products, and artificial sweeteners) in surface and groundwater (drinking water) in the Ganges River Basin, India. *The Science of The Total Environment 646*, 1459-1467. https://doi.org/10.1016/j.scitotenv.2018.07.235

Silva, M. S., Lizama, B. C. (2009). Chlorbromuron urea herbicide removal by electro-Fenton reaction in aqueous effluents. *Water Research*

43, 33-40. https://doi.org/10.1016/j.watres.2008.09.036

Smrčková, P., Horsky, J., Šárka, E., Koláček, J., Netopilík, M., Walterová, Z., Krulič, Z., Synytsya, A., Hručková, K. (2013). Hydrolysis of wheat B-starch and characterization of acetylated maltodextrin. *Carbohydrate Polymers 98*, 43-49. https://doi.org/10.1016/j.carbpol.2013.04.065

Sucralose-FDA. (1998). *Final Rule- Food Additives Permitted Addition to Food for Human Consumption*. United States: Food and Drug Administration. Federal Register/ Vol 63, No. 64/Friday, April 3, 1998/ Rules and Regulations, 16417-16433. https://www.govinfo.gov/content/pkg/FR-1998-04-03/pdf/98-8750.pdf

Suez, J., Korel, T., Zilberman-Schapira, G., Thaiss, C. A., Maza, O., Istaeli, D., Zmora, N., Gilad, S., Weinberger, A., Kuperman, Y., Harmelin, A., Kolodkin-Gal, I., Shapiro, H., Halpern, Z., Segal, E., Elinav, E. (2014). Artificial sweeteners induce glucose intolerance by altering the gut microbiota. *Nature 154*, 181-186. https://doi.10.1038/nature13793

Tabani, H., Mahyari, M., Sahragard, A., Fakhari, A. R., Shaabani, A. (2015). Evaluation of sulfated maltodextrin as a novel anionic chiral selector for the enantioseparation of basic chiral groups by capillary electrophoresis. *Electrophoresis 36*, 305-311. https://doi.org/10.1002/elps.201400370

Tran, N. H., Nguyen, V. T., Urase, T., Ngo, H. H. (2014). Role of nitrification in the biodegradation of selected artificial sweetening agents in biological wastewater treatment process. *Bioresource Technology 161*, 40-46. https://doi.org/10.1016/j.biortech.2014.02.116

Walling, C. Fenton's reagent revisited. (1975). *Accounts of Chemical Research 8*, 125-131. https://doi.org/10.1021/ar50088a003

Wang, F., van Halem, D., van de Hoek, J. P. (2016). The fate of H_2O_2 during managed aquifer recharge: A residual from advanced oxidation processes for drinking water production. *Chemosphere 148*, 263-269. https://doi.org/10.1016/j.chemosphere.2016.01.046

Wiklund, A. K., Breitholtz, M., Bengtsson, B. E., Adolfsson-Erici, M. (2012). Sucralose-An ecotoxicological challenger? *Chemosphere 86*, 50-55. https://doi.org/10.1016/j.chemosphere.2011.08.049

Wood, S. G., John, B. A., Hawkins, D. R. (2000). The pharmacokinetics and metabolism of sucralose in the dog. *Food and Chemical Toxicology 38*, S99-S106. https://doi.org/10.1016/S0278-6915(00)00031-4