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ERYTHROSINE B IN THE ENVIRONMENT. REMOVAL PROCESSES

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Abstract: *This paper presents a comparison of the resultes obteined for different methods used for the decolorization of the food dye Erythrosine B.*
Erythrosine B is a red odorless powder used in food industry as a colloring substance. Erythrosine B, also known as E 127, consists essentially of disodium 2-(2,4,5,7-tetraiodo-6-oxido-3-oxoxanthen-9-yl) benzoate monohydrate and subsidiary coloring matters together with water, sodium chloride and sodium sulphate as the principal uncolored components. The methods considered in this paper were sorption, biodegradation and photodegradation. Sorption demonstrated good removal efficiency in the presence of low-cost activated carbon from agro-waste but the treatment increases the operation cost. Because of Erythrosine B toxicity aerobic biodegradation processes showed to be inefficient in the most studies. Considering this, Erythrosine B degradation could be performed by photodegradation process using an adequate catalyst in order to reduce the operation cost.

Keywords: *food dyes, performance comparison, removal processes*

1. Introduction

Food coloring is a substance that is added to food or drink to change its color. Erythrosine B (FD & C Red No. 3) is the only xanthene dye listed for use in food and ingested drugs. It is exclusively authorised for use in cocktail and candied cherries, and Bigarreaux cherries (94/36/EC).
The paper presents an overview including:
- Erythrosine B presentation in terms of its use as a xanthene food coloring substance;
- the most used methods used for the possible decolorization of aqueous solutions containing the food dye Erythrosine B.

1.1. Erythrosine B (E 127), a coloring substance in EU

Synthetic food colors, or coal-tar/ petroleum colors, represent a special class of dyes with application in food industries.
The main dye categories used in food industry are:
- azo dyes, such as: tartrazine (FD&Cyellow no. 5)., ponceau (FD&C red no. 4), sunset yellow (FD&C yellow no. 6);

- non azo dyes, like brilliant blue (FD&C blue no. 1), erythrosine (FD&C red no. 3), indigotin (FD&C blue no. 2).

Unlike other food additives, these compounds need to be tested and certified by the official chemical examination bodies. Dyes approved by the Food Dye and Coloring Act (FD&C) are coal tar derivatives, contain aromatic rings. **Erythrosine B (E 127)** is a red odorless powder or granules with a calculated Log P (octanol-water) of 4.95 at 25°C, which is soluble in water (≤ 9% w/w) and ethanol [2]. The molecular weight of E 127 is 879.84 g mol^{-1}. It's full chemical name is disodium *2-(2,4,5,7-tetraiodo-6-oxido-3-oxoxanthen-9-yl)benzoate*.

Erythrosine B structural formula is presented in Fig. 1. At least 78 synonyms of the compound are in use. As European Food Safety Authority (2011) related the most commonly synonyms used in literature for Erythrosine are: CI Food Red 14, FD & C Red No. 3, C.I. 45430, INS No. 127 and Erythrosine sodium [1]. Erythrosine B is a red odorless powder or granules with a calculated Log P (octanol-water) of 4.95 at 25°C, which is soluble in water (≤ 9% w/w) and ethanol [2]. Specifications have been defined in the Directive 2008/128/EC7 and by JECFA (2006) (Table 1) [3].

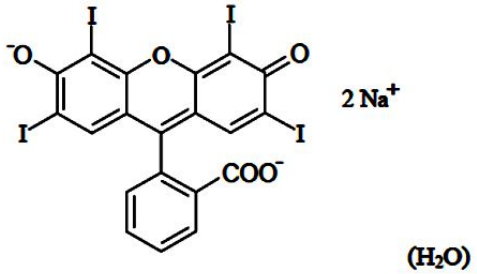


Figure 1. Chemical structure of Erythrosine B

Erythrosine B (E 127) consists essentially of disodium 2-(2,4,5,7-tetraiodo-6-oxido-3-oxoxanthen-9-yl) benzoate monohydrate and subsidiary coloring matters together with water, sodium chloride and sodium sulphate as the principal uncolored components. The phosphorescence of Erythrosine B is due to the xanthene ring with four iodine atoms. Erythrosine B has phosphorescence emission time scale of 10^{-5}s to 10^{-3}s corresponding to motion in glassy environment and is sensitive to oxygen [4].

Table 1
Specification for Erythrosine according to Commission Directive 2008/128/EC and JECFA (2006) [3]

Purity	*Commission Directive 2008/128/EC*	*JECFA (2006)*
Inorganic iodides calculated as sodium iodide	≤ 0.1%	≤0.1%
Fluorescein	≤20 mg kg^{-1}	<20 mg kg-1
Subsidiary colouring matters	≤ 4.0%	≤4.0%
Water insoluble matter	≤0.2%	≤0.2%
Ether extractable matter	≤0.2%[a]	≤0.2%[b]
Arsenic	<3 mg kg^{-1}	-
Lead	≤10 mg kg^{-1}	≤2 mg kg-1
Zinc	-	≤50 mg kg -1
Mercury	≤ 1 mg kg^{-1}	-
Cadmium	≤ 1 mg kg^{-1}	-
Heavy metals as Pb	≤ 40 mg kg^{-1}	-
Loss on drying at 135°C together with chloride and sulphate calculated as sodium salts	-	≤13%
Tri-iodoresorcinol	≤0.2%	≤0.2%
2-(2,4-dihydroxy-3,5-diodobenzoyl) benzoic acid	≤0.2%	≤0.2%

[a] from a solution of pH from 7 through 8
[b] from a solution of pH not less than 7

Erythrosine B, like other xanthene compounds, exhibits unusual spectroscopic and photochemical properties like huge absorption coefficient or molar extinction coefficients (λ_{max}: ε_{524nm} = 67,282 M^{-1} cm^{-1} for Erythrosine B; ε_{510nm} = 60,826 M^{-1} cm^{-1} for Eosin Y) in the visible region and a high tendency for intersystem crossing to produce a photochemically active triplet excited state [5].

1.2. Short overview on the synthesis process and analysis

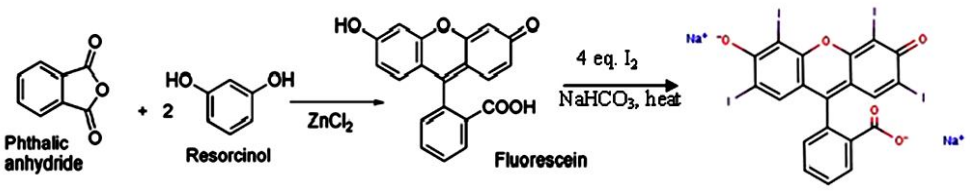
Erythrosine B, the tetraiodo- analogue of fluorescein, is produced by iodination (the reaction of iodine or potassium iodate in an ethanolic solution converted to the sodium salt) of fluorescein followed by the condensation of resorcinol with phthalic anhydride (Eq. 1) [6].
Erythrosine B may be converted to the corresponding aluminium lake by reacting aluminium oxide with coloring matter [1]. Undried aluminium oxide is usually freshly prepared by reacting aluminium sulphate or aluminium chloride with sodium carbonate or sodium bicarbonate or aqueous ammonia. Following lake formation, the product is filtered, washed with water and dried [3].
In the literature few methods for the determination of Erythrosine B in foods are described. These methods include High Performance Liquid Chromatography (HPLC) and capillary electrophoresis [7].

1.3. Stability, reaction and fate in the environment

A low number of data on the fate and reaction of Erythrosine B in food is available. In general, the dyes used as additives are changeable in combination with oxidizing/reducing compounds in food. Dyes depend on the existence of a conjugated unsaturated system.
Any compound which modifies the system will affect the dye (e.g. oxidising or reducing agents, sugars, acids, and salts) [8]. For exemple when cherries colored with Erythrosine B are stored in uncoated steel cans, fluorescein is readily formed (the production of fluorescein from Erythrosine B occurs in the presence of iron and/or tin and free organic acid as a result of electrochemical reduction in the can [9]).

1.4. Use and toxicology

Authorized use levels have been defined in the Directive 94/36/EC8 on colors for use in foodstuffs (Table 2). As a synthetic food coloring substance Erythrosine B is permitted in the EU for certain limited uses only.
Table 2 give the main foodstuffs that are permitted to contain Erythrosine B up to specified maximum permitted levels (MPLs) set by Directive 94/36/EC.
Erythrosine B has been used as a food dye since its approval by the U.S. Department of Agriculture in 1907.
It is used in maraschino cherries, sausage casings, oral drugs, baked goods, and candies. Erythrosine B has been evaluated several times by JECFA and by the Scientific Committee for Food.



(1)

Table 2
Food Dye Certification [10]

Food dye	*Pounds of total dye certified*	*Percentage of total*
Blue 1	711 659	4.7
Blue 2	550 883	3.7
Citrus Red 2	1 764	0
Green 3	15 817	0.1
Orange B	0	0
Erythrosine B	216 235	1.4
Red 40	6 203 374	41.3
Yellow 3	3 765 331	25
Yellow 6	3 338 351	23.7
Total	15 016 634	100

Table 3
Maximum permitted usage levels of Erythrosine B according to European Parliament and Council Directive 94/36/EC [11]

Foodstuffs	**Maximum Permitted Level (mg/kg)**
Cocktail cherries and candied cherries	200
Bigarreaux cherries in syrup and in cocktail	150

The US Code of Federal Regulation, states that FD&C Red No.3 (Erythrosine B) may be safely used as a coloring in general foods in amounts consistent with Good Manufacturing Practice (GMP) [12].
Erythrosine B is permitted in the USA for general use in sweets and foods marketed to children such as candies, popsicles, cake frosting and cake-decorating gels.
The report provided by United States Food and Drug Administration (US FDA) indicate that under experimental conditions Erythrosine B at high dose levels (4% in the diet) can affect the level of circulating thyroid hormones in rats, thus leading to an increase in the incidence of thyroid tumors. The response of the US FDA in 1990 was withdrawal of permission to use Erythrosine B lakes (salts), but not Erythrosine B, in all foods, drugs and cosmetics, and to withdraw the use of Erythrosine in cosmetics and externally applied drugs [1].
In Australia, the Code restricts the use of Erythrosine B in foods. It is used just to preserved cherries (maraschino cherries), cocktail cherries or glace cherries. The dye is used prior to processing.
Food Standards Australia New Zealand (FSANZ) have proposed extending the permitted uses to products such as icing and frostings used in other foods that are more widely consumed (e.g. cakes, biscuits, fancy breads) [13].
In EU, Erythrosine B is used in manufacturing photographic plates, for microscopic stains, pharmaceuticals and cosmetics. In June 2010, the Scientific Committee on Consumer Safety published an opinion on Erythrosine use in toothpaste products. GlaxoSmithKline (GSK) considered Erythrosine B safe for consumers when used as a colorant in toothpaste products with a maximum concentration of 0.0025% (25 ppm) and estimated exposure from this use to be 0.0002 mg/kg bw/day (Table 4) [1].

Table 4
Toxicological parameter of xanthene dye Erythrosine B [13]

	Toxicological parameter	***Value [mg kg^{-1}]***
Accepted Daily Intake = 0 - 0.1 mg kg^{-1} bw^{-1} day^{-1}	LD_{50} (rat), intraperitoneal	300
	LD_{Lo} (rat), intravenous	200
	LD_{Lo} (rabbit), intravenous	200
NOEL = 60 mg $person^{-1}$ day^{-1} (mg kg^{-1} bw^{-1} day^{-1})	LD_{Lo} (mouse), oral	2500
	LD_{50} (mouse), intravenous	370

2. Removal of Erythrosine B from wastewater

Food once was colored only with natural dyes, coming mainly from various plants.

By the 19th Century, colors began to derive from other chemicals so that they came into use with sometimes serious health consequences.

Erythrosine B is still permanently listed for use in ingested drugs and food, such as baked goods, cherries, dairy products, desserts, dietary supplements, food seasonings, jellies, jams, and vegetable products.

Acid dye like Erythrosine B can be lost in effluents in percentage varying from 5 to 20%, since low adsorption occurs for acid and reactive dyes, while high adsorption occurs with basic and direct dyes and high to medium for disperse dyes [14; 15].

Ryvolova et al. [16] and Ramakrishnan et al. [17] found that the concentration of colorants in foods and drugs, respectively were between 0.3 and 0.03 mg mL^{-1}. In this situation, it was estimated that a high amount of dye results as waste and has to be treated (Table 5) [16; 17].

Erythrosine B contained in aqueous solutions was subjected to different decolorization methods, including physical, chemical, and biological processes. Some of them are discusses below.

Table 5
Amount of Erythrosine B contained in different products

Product	*Erythrosine B ($\mu g\ mL^{-1}$)*
Cherry	235
Cream biscuits	316.65
Gems	177.8
Candies	36.74
Ibuprofen	5.6 (µg/tablet)

2.1. Sorption

Sorption is widely used for dye removal from wastewaters. Activated carbon, the universal adsorbent used for pollutant removal, although reasonably effective at removing dyes from aqueous streams, needs either regeneration or disposal, once it is fully loaded. Other limitations are the high cost and 10-15% loss of adsorbent during reactivation [14].

In recent years, many investigations have been undertaken to evaluate inexpensive alternative materials of biological origin (biosorbents) as potential adsorbents for dyes, which include feathers [18], de-oiled mustard [19], montmorillonite [20], bottom ash and de-oiled soya [21], fungi [22].

Due to its low cost and widespread availability, biomass has been extensively investigated as sorbent for removing color, with promising results**.** In this case, biomass refers to dead plant and animal matter resulting from agriculture, forest, fermentation and shellfish by-products or wastes.

Other unconventional biosorbents used for sorptive removal of different dye classes are pinus bark powder [23], hazelnut shells [24], nut shells [25], or bagasse pith [26]. The main mechanisms found responsible for the decolorization of aqueous solutions containing dyes are adsorption and ion exchange.

Agricultural wastes can be good sorbents for the removal of pollutants, since they involve reduced costs for operation and waste disposal, providing a cheap alternative to existing commercial activated carbons.

Vegetal hull/husk was applied as a raw material for producing sorbents with the following advantages:

(1) appropriate chemical composition;
(2) low cost;
(3) high dispersity;
(4) scaly structure and developed porous surface ensuring a high surface-to-volume ratio [27].

Several studies reported chemical modifications of celluloses and ligno-celluloses extracted from cotton waste, sawdust and corn stalks in order to increase the number of active centers for dye immobilization on sorbent surface [28].

Table 6
Sorbents tested for xanthene dye removal

Dye	*Initial dye conc. ($x\ 10^{-5}$ mol L^{-1})*	*Sorbent*	*q ($x\ 10^{-5}$ mol g^{-1})*	*Ref.*
Rhodamine B	2 - 200	fungi	8.15	[22]
	5 - 50	activated carbon	4.57	[33]
	4 - 400	algae	16.7	[34]
	10 - 50	baryte	34.22	[35]
Eosin Y	-	sun-dried jute fiber	4.55	[36]
	1 - 70	chitosan hydrobeads	11.57	[37]
Erythrosine B	1 – 6	bottom ash de-oiled soya	2.37 1.20	[21]
	1 – 6	hen feathers	2.31	[18]
	1 - 9	de-oiled mustard activated carbon	13.15 19.9	[19]
	5 - 10	montmorillonite	1.5	[20]
	5 - 40	Activated carbon	47.28	[38]

Adsorption of reactive dyes by sawdust chars and activated carbon [29]; methylene blue by waste *Rosa canina* sp. seeds [30]; anionic dyes by modified coir pith [31]; and methylene red by acid-hydrolysed beech sawdust [32] has also been reported.
Table 6 presents a summary of the biosorbents used for xanthene dye removal.

2.2. Biodegradation

Biological processes involve the aerobic (presence of oxygen) or anaerobic (absence of oxygen) degradation of organic substances by microorganisms.
Anaerobic biological reduction of dyes has been investigated from different perspectives, i.e. degradation and color removal. Anaerobic reducing conditions found in the environment include sediments at the bottom of streams of certain sections of landfills where there is no oxygen. Anaerobic bioremediation of soluble dyes to undergo decolorization by breaking them into less toxic compounds has been widely investigated [14]. The decolorization in the case of azo dyes occurs due to azo reduction [39].
Additional carbon is required for decolorization to proceed the process at a viable rate: this is converted into methane, hydrogen sulphide and carbon dioxide [40]. This additional carbon source may be a limiting factor from a commercial perspective. In many situations, decolorization of reactive dyes under anaerobic conditions is due to the action of a 'reductase' enzyme. If complete mineralization occurs, conversion of organic contaminants into methane and oxygen leads to production of bioga,s which is a major attraction because of heat, power and reduced energy costs [14].
In the case of toxic compounds interactions with living microorganisms two pathways can be considered:
i) when biodegradation starts with chemical interactions, the sorption rate limits the biodegradation;
ii) when biodegradation starts after some lag period, the chemical has time to sorb before degradation begins, and slow sorption causes biodegradation to be desorption-rate limited [41].
However, the toxicity of certain dyes inhibits the complete mineralization.
Xanthene dyes have been reported to be toxic to various species tested in laboratory conditions. Several reports on enzymatic oxidation of xanthene dyes are available [42; 43; 44; 45; 46; 47]. Enzymatic reduction of xanthene compounds was not reported.

Borgerding and Hites (1994) reported the presence of xanthene dye Erythrosine B in wastewater from food and cosmetic industry and affirmed that the dye was adsorbed by the sludge [48]. Itoh and Yatome (2004) studied the decolorization of six xanthene dyes by a white rot fungus, *Coriolus versicolor* but only three of them were degraded [45]. Table 7 presents a summary of xanthene dyes removal by enzymatic mechanism.

2.3. Photodegradation

Because processes like adsorption or biodegradation can generate secondary pollutants, AOP like photocatalytic degradation represented an alternative for the removal of hazardous xanthene dye from effluents [49].

AOPs are effective for detoxification and mineralization of dyes from wastewaters and research studies have shown promising results as these processes appear to have the ability to completely decolorize and partially mineralize the pollutants from dye-industry in short reaction time [50; 5].

Table 7
Xanthene dyes removal by biodegradation

Dye	*Initial dye conc. ($x\ 10^{-5}$ mol L^{-1})*	*Biomass*	*R (%) r ($\mu M\ min^{-1}\ mg^{-1}$)**	*Ref.*
Fluorescein	10	Fungus *Coriolus versicolor*	85.0 10.2	[45]
4 - Amino fluorescein	10	Fungus *Coriolus versicolor*	95.0 6.7	
5 - Amino fluorescein	10	Fungus *Coriolus versicolor*	91.9 7.2	
Rhodamine B	10	Fungus *Coriolus versicolor*	0	
	5	Laccase Mediator System (LMS)	80%	[46]
	40	LiP of fungus *Phanerochaete chrysosporium*	46.0	[44]
Rhodamine 123	10	Fungus *Coriolus versicolor*	0	[45]
Rose bengal	-	Fungus strain *Aspergillus Wentii*	89.3	[43]
Erythrosine B	10	Mold *Neurospora crassa*	-	[47]
	-	Aerobic sludge	0	[48]
	30	Anaerobic granular sludge	20 – 70 1.5 h^{-1}*	[38]

*****first order degradation rate

Table 8
Xanthene dye removal by chemical degradation process

Dye	*Initial dye conc. ($x\ 10^{-5}$ mol L^{-1})*	*Catalyst (UV)*	*R (%)*	*Ref.*
Eosin Y	10	TiO_2 P25	78	[51]
Rhodamine B	5	0.65 g L^{-1} TiO_2	80	[52]
	1	SiO_2@TIO_2	90	[53]
Rhodamine 6G	5	1 g L^{-1} ZnO	80	[54]
Fluorescein		ZnO	44.4	[55]
Phloxine B	10	0.04 g L^{-1} TiO_2	82	[19]
Erythrosine B	10	2 g L^{-1} TiO_2 P25	35	[56]
	10	g L^{-1} ZnO	60	
	10	0.5 g L^{-1} TiO_2 Aeroxide	99	[57]
	20	Electrochemical degradation	95	[58]

Photodegradation is based on the degradation of a molecule mediated by the absorption of photons. Photocatalytic degradation of several xanthene contaminants using large bandgap semiconductor particles (such as TiO_2, ZnO, WO_3) has been extensively studied.
Table 8 shows data on xanthene dye photodegradation in the presence of different catalysts.

3. Comparison of physico-chemical and bological processes applied for the decontamination of aqueous sollutions polluted with Erythrosine B

Physico-chemical and biological methods that are effective in the elimination of dye have been studied. Each process has its own constraints in terms of cost, feasibility, practicability, reliability, stability, environmental impact, sludge production, operational difficulty, pre-treatment requirements, the extent of the organic removal and potential toxic by-products. Even if a process is reported to be successful in decolorizing a particular effluent, the same may not be able to other type of colored aqueous solution. The use of a single process may not be efficient in the complete decolorization of the polluted effluent.
The comparison of different processes used for dyes removal from aqueous solution is of interest to establish the conditions for removal performance (the most efficient experimental conditions for the elimination of compound from solution) and to provide useful information for the essential aspects of the combination of different processes [59].
Sorption, biodegradation and photodegradation processes where tested to evaluate Erythrosine B removal from aqueous solution.
Table 9 summarizes the conditions and the performances of the processes obteined for 300 mg L^{-1} Erythrosine B (the higer dye concentration tested) removal from aqueous solution.
Sorption process resulted in 65% – 90% removal efficiency at 50°C and natural pH of the solution (pH=5.6) for a contact time of around 21h [60].
Photodegradation using fixed or suspended TiO_2 yielded very high color removal (95%), within the same contact time like sorption study also at natural pH and resulting nontoxic products.
Anaerobic biodegradation treatment has demonstrated to be affected by Erythrosine B toxicity at high concentration (0.4 mM) and low color removal was achieved for the same biomass amount as with sorption study (20 g L^{-1}) [38].
Sorption process demonstrated to have good removal efficiency especially at higher temperature (50°C) where the dye amount adsorbed per sorbent unit was between 14.1 and 16.4 mg g^{-1} for BH and PSH respectively. Sorption of Erythrosine B demonstrated a ~5-fold increase of dye adsorption in the presence of low-cost activated carbon in the study conducted by Jain and Sikarwar (2009) but agro-waste treatment increases the operation cost [19].

Table 9
Erythrosine B removal using physico-chemical and biological methods under different condition

Process applied	*Experimental condition*	*Amount of dye uptake (mg g^{-1})*	*Removal efficiency (%)*	*Observation*
Sorption using agro-waste	**C_i=300 mg L^{-1} Contact time = 21h**	**14.1 - 16.4 mg g^{-1} (BH and PSH)**	**65 – 80 (BH and PSH)**	**$C_{sorbent}$= 20 g L^{-1} t=50°C**
Anaerobic Biodegrdation		**3 mg g^{-1}**	**30**	**$C_{biomass}$= 20 g L^{-1} t=37°C**
Photocatalytic degradation		**-**	**95**	**C_{TiO2}= 0.5 g L^{-1} t_i=25°C**

The results obtained for biodegradation study confirmed the biomass capacity to adsorb Erythrosine B based on the biomass color at the end of the study. Similar result was obtained by Chamam et al. (2007) for a sulphuric textile dye, Cassulfon CMR, using activated sludge for the batch sorption test [61].

For the range of Erythrosine B concentrations studied (30 and 300 mg L^{-1}) dye photodegradation in the presence of TiO_2 represent the most efficient method than sorption and biodegradation in the removal of Ery B in aqueous solution. Ong et al. (2009) compared the sorption and photodegradation of BB3 and RO16 and observed also a higher efficiency for photodegradation using TiO_2 that in the sorption study using modified rice hull [62].

The decolorization rate was observed to be different for each system, for the first 60 min of reaction the relative order evaluated was: sorption > photodegradation >> biodegradation.

The combination of different methods for the removal of dyes from aqueous solution sounds to be an ideal solution for the present need of time. A combination of AOP and biological treatment showed better results for the color removal from dyeing effluents containing hazardous compounds [63].

The scientific knowledge represented an important element for managing the direction and improvement of techniques applied for dye removal from effluents. In view of the need for a technically and economically satisfying treatment technology, an abundance of technologies were proposed and tested [40].

4. Conclusions

Erythrosine B or FD&C Red No.3 is a cherry-pink/red synthetic coal tar dye. It is most popularly used as food coloring and a host of other applications, such as printing inks, biological stain, and for extraction-photometric determination of K, Cd, Pb, Mn, Zn, Ag. Erythrosine B was the subject of different decolorization methods applied to aqueous solutions containing dye. Sorption demonstrated good removal efficiency in the presence of low-cost activated carbon from agro-waste but the treatment increases the operation cost. Because of Erythrosine B toxicity aerobic biodegradation processes showed to be inefficient in the most studies. Considering this, Erythrosine B degradation could be performed by photodegradation process using an adequate catalyst in order to reduce the operation cost (with irradiation time).

5. Acknowledgments

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