Chapter

Application of Advanced Oxidation Process in the Food Industry

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Abstract

Wastewater in the food industry contains recalcitrant organic compounds and a certain degree of toxicity. Present wastewater treatment plants are insufficient in dealing with the increasing complexity of effluents from modern food industries. Improperly treaded wastewaters can lead to spoil soil and are threads to aquatic life. The reaction of these recalcitrant chemicals with reactive radicals is an efficient treatment strategy. Researchers have proposed advanced oxidation processes (AOPs) that generate reactive radicals including ozonation, UV irradiation, (photo-) Fenton process, etc. This chapter reviews laboratory-scale and pilot-scale AOPs to incorporate with conventional pre-treatment methods and to evaluate their effectiveness and factors including operation condition and catalysts to optimize the process. Further research related to novel catalyst synthesis and cost evaluation of pilot-scale study is suggested.

Keywords: advanced oxidation processes (AOPs), food industry wastewaters, physicochemical methods, biodegradability improvement, combined treatment

1. Introduction

Water is a widely used resource in the food industry such as sanitary water for food processing, as a raw material, and for equipment cleaning. The large consumption of water leads to a corresponding generation of wastewater. Wastewater treatment from a diverse industry is technically more challenging than the treatment of domestic wastewater whose characteristics are largely similar. The various utilizations of water in the industries result in complex compositions of the resulting wastewater [1], which is often a thread to human, plant, and aquatic life [2].

The complexity of wastewaters from the food industry is a more severe issue than its amount. The effluents typically include large total load of organic pollutants such as colored and recalcitrant compounds with high organic loads (chemical oxygen demand, COD), proteins or fat, and chemicals used during processing [1], some with considerable levels of toxicity [3]. Current municipal wastewater treatment plants with the remaining capacity for population growth are not designed to handle the characteristics of such influent loading [4], creating a demand of complementary technologies in addition to traditional treatments.

Advanced oxidation processes (AOPs) generate highly reactive radical (**Table 1**). As one of the most active oxidizing agents, hydroxyl radical is generally

Oxidizing agent	**Relative oxidation activity**
Positively charged hole on titanium dioxide, TiO_2^+	2.35
Hydroxyl radical	2.05
Atomic oxygen	1.78
Ozone	1.52
Hydrogen peroxide	1.31
Permanganate	1.24
Hypochlorous acid	1.1
Chlorine	1

Table 1.
Relative oxidation activity of some oxidizing agents [5].

Organic compound	**Rate constant [$M^{-1}\ s^{-1}$]**	
	O_3	**HO**
Alcohols	10^{-2}–1	10^8–10^9
Aromatics	1–10^2	10^8–10^{10}
Chlorinated alkenes	10^3–10^4	10^9–10^{11}
Ketones	1	10^9–10^{10}
N-containing organics	10–10^2	10^8–10^{10}
Phenols	10^3	10^9–10^{10}

Table 2.
Reaction rate constants for ozone and hydroxyl radical for organic compounds [7].

used in AOPs. It reacts nonselectively with organic compounds [6] which are resistant to conventional oxidation methods as high reaction rate (**Table 2**).

AOPs are widely used in winery wastewater, olive oil mill wastewater, and slaughterhouse wastewater. In addition to degradation of extra chemical compounds directly, AOPs are used to degrade cellular contents to disinfect wastewater with high organic nature in the dairy industry [8].

However, AOPs are capital-intensive since they include the use of expensive regents such as ozone and hydrogen peroxide, as well as the cost of equipment including sources of ultraviolet light. Hence AOPs are often combined with traditional treatments to overcome this drawback. The combination technologies reduce toxicity and increase the biodegradability of wastewater effluent after AOPs, which is more suitable for inexpensive biological process.

As a complementary of previous reviews [1, 9], this review categorizes relevant AOP applications in the food sector into winery, olive oil mill, meat industries, dairy, and food dye sections. This work summarizes state-of-the-art wastewater treatment technologies in food industries by AOPs both at laboratory-scale and pilot-scale after 2016. Current treatment technologies are also introduced.

2. Introduction to current wastewater treatment technologies in the food industry

Current wastewater treatment in the food industry is evolving from traditional treatments by focusing on the degradation of excessive organic matters, the recovery of profitable by-products, and water reusability. Typically wastewater treatment consists of three common stages: primary treatment such as physicochemical treatments partially removing solids; secondary treatment aimed at removing biodegradable organics and suspended solids using biological methods including aerobic and anaerobic processes; and tertiary treatments designed to remove residual compounds of particular concern to the given plant. Current technology alternatives to AOPs for wastewater treatment in the food industry are introduced briefly.

2.1 Evaporation

Natural evaporation during summer can be applied to olive oil mill wastewater (OMW) stored in lagoons, which reduces the volume of waste but results in odor and soil pollution. Alternatively, vacuum evaporation can reduce the volume of wastewater at low temperature and pressure; two streams including distillate and concentrate are separated which can then easily be handled with common treatments [10].

2.2 Land application

The application of wastewater as organic fertilizers has been investigated. High organic loadings in wastewater provide nutrients for soil but can also result in soil contamination. For lower amounts of wastewater, soil can act as biofilter of OMW degradation [11]. In contrary to direct application, composting is more applicable due to the elimination of phenols [12].

2.3 Physicochemical treatments

Colloidal particles in wastewater result in filtering devices clog. To prevent the malfunction of equipment for biological treatment and to meet the discharge requirement, technologies called physicochemical treatments are in demand to eliminate these particles.

The addition of flocculation destabilizes colloidal dispersions which count for the majority of the total dispersed solids (TDS), turbidity, and part of COD. Moreover, lime treatment and adsorption are also practical technologies [13]. Such treatments increase biodegradability during biological treatment and reduce residual components including organic load, color, and metal content after biological treatment [14]. For further study, alternative coagulants are promising to achieve higher removal efficiency, less pollution, and lower health risks [15].

2.4 Membrane technologies

Membrane technology is applied when phytotoxic recalcitrant pollutants invalidate biological treatment [16] or for recovery of valuable products [17]. With the advantage of high efficiency, simple equipment, and convenient operation, membrane technologies are beneficial to small plants that cannot afford high investments. However, membrane fouling needs to be reduced to increase economic feasibility. Pulido et al. reviewed current membrane technologies addressing the efforts of decreasing fouling issues in OMW [16].

According to the comprehensive review papers of winery wastewater treatment, membrane bioreactors are competitive considering water reuse [14, 15].

2.5 Summary

The choice of treatment solution is based on the specific characteristic of wastewater. The combination of complementary treatments is promising to achieve discharge regulation. More detailed current technologies and their comparison are reviewed elsewhere [18].

The advances in wastewater treatment in the food industry are driven by the rising awareness of environment protection in the public. At present, for example, various reactors are being designed for olive mill wastewater treatment, while most of them were not treated before 2004 in the EU [10]. However, the remaining challenges after current treatment call for extra treatment technologies in future research, which might be addressed through AOPs.

3. Application of AOPs in the food industry

3.1 Wineries

In 2016, the worldwide total alcohol consumption was equal to 6.4 liters of pure alcohol per person aged 15 years and older [19]. Accordingly, winery wastewater originates from multiple steps in the production process. One of the origins is fresh water used for cleaning, including washing of equipment and facilities [20], while other streams are directly related to wine, such as the effluent from filtration units and off products [15].

Winery wastewater contains organic matters such as ethanol, sugars, organic acids, phenolic compounds, etc. [15]. Moreover, there are toxic compounds from pesticides used on grapes [22]. Winery wastewater discharged without any treatment would result in soil pollution. General sewage treatment facilities are not sufficient to process the wastewater which leads to acid pH and high COD [27]. The treatment for winery wastewater has to be specific due to the differences between each winery, such as the type of wine, unique component and volume of the wastewater, etc. [28]. Even in the same winery, the effluent is unstable according to the operation period and season [29].

A recent review [15] summarizes the processes currently applied and/or tested for the treatment including physicochemical, biological, membrane filtration and separation, advanced oxidation processes, and combined biological and advanced oxidation processes. Physicochemical processes (i.e., coagulation/flocculation, EC) can be used as pre-treatment to lower TSS. Biological treatment processes using membrane bioreactors (MBR) are promising and are efficient in reducing organic load, while AOPs, especially photo-Fenton as post-treatment in combination with other treatment, show higher efficiency in reducing COD. Mechanism and restriction of each method are reviewed elsewhere [30], also providing a guidance for choosing suitable method according to different matrices. Applications of AOPs in the winery industry since 2016 are summarized in **Table 3**.

The combined electro-Fenton processes and photolysis processes showed higher efficiency in degradation rate and energy consumption because of synergetic effect. Such processes can be implemented in separate steps or can be integrated together. Díez et al. proposed different sequential reactors which are divided into two sections. Wastewater processed in the electro-Fenton section generates Fe complexes, which are decomposed by light radiation in the photolysis section. They initially proposed a two-chamber cubic reactor, which suffered from foam formation during aeration. The foam reduces the irradiation volume; hence the reactor was changed

Winery wastewater origin	Treatment process	Observations	Reference
Simulated winery wastewater (SWW) from commercial wine. COD 14.43 g/L, TOC 3.726 g/L, maximum wavelength 522 nm, color intensity (CI) 1.31, browning index (BI) 0.55, pH 3.9	Photo-electro-Fenton process	TOC reduced to 61% (mercury lamp) and 65% (LED lamp). DC, CI, and BI reduced totally. (optimized condition: electrode gap, 2 cm electric field 15 V; reaction time, 180 min) TOC: 68.14% (RWW1), 68.77% (RWW2)	[21]
Simulated (SE1, SE2) and real (RE) COD 10,940 mg/L TOC 4427 mg/L pH 3.2 COD; TOC of SE2 is about 8 times of SE1 (82,050, 33,200). pH 2.9 COD (86100) of real WW (60100) is similar to SE2; TOC is twofolds of SE2. pH 3.4	Photo-electro-Fenton process	SE1(200 min) Color: 97.49% TOC: 53.01% G-PTFE cathode, flow rate: 2 ml/min SE2: Color: 93% TOC: 65.7% RE: Color: 60% TOC: 51% 5 V, reaction time 1.3 h	[22]
local white wine producer (Spain) pH 4.08 TOC 55 g L^{-1}	Sequential two-column electro-Fenton-photolytic reactor	Color: 65% TOC: 67%, 64%^{P} COD: 77%, 74%^{P} IC_{50}: 76.5%, 41%^{P} M Na_2SO_4, 75 mg L^{-1} of Fe^{3+}, pH 2 and 5 V Reaction time: 12 h^{P}: with pesticides	[22]
Undurraga® Winery Company (Talagante, Santiago de Chile) COD 3490 mg/L TOC 1320 mg/L NTU 15.2	Anodic oxidation	Eliminate COD, TOC, and NTU totally 50 mM of NaCl or Na_2SO_4 and apply higher-density currents (60 mA /cm^2) Reaction time 420 min pH 8.30 ± 0.20	[23]
Douro red wine diluted samples COD 513 mg/L TOC 143 mg/L pH 4.0	Solar radiation-assisted sulfate radical-based advanced oxidation processes (SR-AOP)	TOC: around 50% COD: 75% (pH 4.5, PMS, Fe (II)) Reaction time 3 h	[24]
Wine cellar in the Douro region in the north of Portugal pH = 4.37, COD 600 mg/L, BOD_5 145 mg/L, BOD_5/COD 0.24. TOC 166 mg/L	UV-C assisted sulfate radical-based advanced oxidation processes (SR-AOP)	COD 96%, TOC 71% Reaction time 240 min. pH 7. Consumed 25 mM $S_2O_8^{2-}$ UV-C = 254 nm	[25]
pH = 3.8, COD 2.128 g/L, BOD_5 974 mg/L, TOC 825 mg/L	Adsorption and photo-Fenton process Ca-smectite as adsorbent and catalyst support	TOC reduced 90% in total, by 54% (adsorption) and 36% (photo-Fenton) pH = 4, H_2O_2 98 MM, catalyst 6 g/L	[26]

Table 3.
Recent application of AOPs in the winery industry.

into two divided columns. The economic efficiency was increased by decreasing the voltage from 15 to 5 V [21, 22]. In order to minimize the electrode gap, the reactor system is designed in two columns connected vertically. The efficiency for degradation of TOC and decreasing color increased in treating real winery wastewater compared to initial laboratory studies.

Both studies investigating simulated winery wastewater using column reactors eliminate color totally. Other than color, TOC reduction rates are both nearly 65%. However, the efficiency reduced with an increased concentration of TOC, which reduces $\cdot OH$ from UV radiation, and accumulated carboxylic acids inhibit regeneration of Fe(II) by reacting with Fe(III) [22].

Anodic oxidation with boron-doped diamond (BDD) electrode is able to process real effluents with high concentration of COD (3490 mg/L), TOC (1320 mg/L), and more than 40 organic compounds [23]. The addition of electrolytes including NaCl and Na_2SO_4 and the application of high-density currents (60 mA/cm^2) increased the efficiency of oxidation, leading to total mineralization in 400 min. The higher efficiency of this process results from the larger number of radicals which have two origins: (i) $HO\cdot$ radicals are weakly bounded on BDD electrode with high O_2-overpotential; (ii) addition of salt generates other radicals such as $S_2O_8^{2-}$ and active chlorine species.

Solar-driven sulfate radical is also a promising tertiary treatment [24]. This process has the advantage of independence from pH. The addition of transition metal as catalyst including Fe(II) and Co(II) shows that Fe(II) performs best when considering environmental implications and efficacy. Furthermore, the reduction of TOC could be enhanced from around 50–71% by combination of UV radiation and thermally activated persulfate (TAP) [25]. It indicates higher efficiency of $S_2O_8^{2-}$ than H_2O_2 for removal of COD (96%) and organic matters, as the UV-C/$S_2O_8^{2-}$ process generates two kinds of radicals, $HO\cdot$ and $SO_4^{-}\cdot$,where the yield of $SO_4^{-}\cdot$ is higher and $SO_4^{-}\cdot$ can be converted into $HO\cdot$.

Natural Ca-smectite can be used for removal [26]. It works both as adsorbent and catalyst support saturated with Fenton. The adsorption capacity can be predicated by Jovanovich isothermal model. For photo-Fenton process' operation condition regarding pH, H_2O_2 concentration and catalyst dosage were optimized, yielding 90% of TOC removal with 54% due to adsorption and 36% due to the photo-Fenton process. However, catalyst regeneration analysis in three consecutive cycles shows reduction to 57%.

Based on the complex organic components and toxicity of winery wastewater, each method aims at increasing the effective radicals. Further study related to reactor optimization, utilization of multiple radicals, and novel catalyst synthesis is worthwhile.

3.2 Olive oil mills

The high demand of water in olive oil production processes results in large amounts of wastewater ranging from 0.5 to 1.68 m^3 per ton with large amounts of semisolid or slurry wastes.

Factors influencing the characteristics of olive oil mill wastewater (OMW) include (i) composition of vegetation water, (ii) olive oil extraction process, and (iii) storage time [30]. Moreover, phenolic compounds result in toxicity of the effluents. However, properly treated OMW produces nutrients (N, P, K) for plants [31]. Therefore, pre-treatment steps are necessary to economically increase the efficiency of the following treatment steps, as summarized in **Table 4**.

A photo-Fenton with medium pressure UV-lamp process was developed for mixture of real OMW [32]. This process showed the ability of fast degradation of pollutants (>90% in 30 mins). The wavelength of UV light did not influence the reduction rates. Another factor is the concentration of H_2O_2. The total nitrogen content removal rate is not related to the concentration of H_2O_2 but the reaction

Olive oil mill wastewater origin	Treatment process	Observations	Reference
Olive oil mill in the province of Seville (Spain) mixture of WOW (olives washing) and WOOW (olive oil washing) pH 5.98; COD 7060 mg/L; TOC 918 mg/L; BOD_5 685 mg/L; turbidity (FTU) 1390	Photo-Fenton	Nitrogen content removed 62.5–75.5%, COD = 95.7 ± 0.53%, TOC 96.3%, total phenolic compounds = 93.6 ± 2.5%, total carbon = 94.0 ± 1.2%, total organic carbon = 96.3 ± 0.6%, total nitrogen = 74.9 ± 6.8%, turbidity = 92.5 ± 1.9%. pH = 3, T = 20°C, catalyst = 3 g/L, reaction time 5–30 min, agitation rate = 600 rpm	[32]
Adjusted olive oil mill (mean value) samples from Cyprus, Israel, Jordan, and Portugal. pH 5.2 EC 12.5 mS/cm COD 25.0 g/L BOD_5 5.1 g/L DOC 6 g/L TSS 24 g/L TP 4.2 g/L TPh 4.2 g/L	Coagulation/flocculation followed by solar photo-Fenton oxidation	Coagulation/flocculation TSS 90%, COD 40%, DOC 11% Solar photo-Fenton oxidation COD 94%, DOC 43%, BOD_5 86%, TSS 96%, TPh 99.8% Reaction time 70 min	[17]
Badajoz (Spain) pH 4.9 COD 6450 mgO_2/L TN 42 mg/L TP 21 mg/L TSS 3190 mg/L BOD_5 2130 mg O_2/L BOD_5 /COD 0.33	Coagulation/flocculation followed by Fenton oxidation and biological treatment (only industry)	Lab Coagulation/flocculation COD 38% + 75% TSS 40% Industry COD 95% Flow rate $1.5m^3\ h^{-1}$ Reaction time 60 days	[33]
COD 1344 ± 22 mg/L; TN 22.4 ± 1.4 mg/L; total phosphate 19.1 ± 1.6 mg/L; phenols 5.89 ± 0.8 mg/L; pH 6.9; conductivity 1260 ± 32 μS/cm Color, expressed as Pt/Co units 2310 ± 9 Olive Oil washing wastewater after filtration Mersin, Turkey	Combined electrocoagulation (ECR)-photocatalytic (PCR) degradation system	**ECR + PCR** COD 88% Phenol (ECR: 88%) 100% Color 100% Reaction time **PCR + ECR** COD 78% Phenol 93.2% (PCR: 40.7%) **ECR:** voltage, 12.5 V; CD, 12.9 A/m2; pH, 6.9; reaction time, 120 min; electrode type, Al **PCR:** catalyst loading, 1 g/L; catalyst type, ZnO; pH, 7.7; reaction time, 120 min; light source, (UVA)	[34]
COD 33927 ± 200 mg/L DOC 10120 mg/L Phenol 164 ± 13 mg/L Color 13,350 ± 120 (Pt-Co) Acid-cracked wastewater from olive production factory. Kahramanmaras, Turkey	Combined ozone/Fenton process	Color 51.6% +21% DOC 27.9% + 49% CODs 58% +22% Phenol 100%	[35]

Olive oil mill wastewater origin	Treatment process	Observations	Reference
TOC 23231–34,050 mgL^{-1} COD 62400–82,400 mgL^{-1} Color 369 Pt-Co pH 4.5 BOD_5/COD 0.144 100 times diluted sample wastewater from olive production factory. Gemlik region, Turkey	Microwave (MW) activated persulfate	TOC 100% COD 63.38% Color 94.85% Optical density 121.7% PS: 266 g L^{-1} Reaction time 23.58 min Power level 567 W Initial pH 2	[36]

Table 4.
Recent application of AOPs on OMW.

time. The removal efficiency decreased with longer reaction time since more N_2 is fixed with abundant CO_2. The conversion rates of TOC have all reached more than 90% and increased slightly with increasing H_2O_2 concentrations.

Solar photo-Fenton oxidation pretreated by coagulation/flocculation achieved 90% TSS removal at optimal dose of $FeSO_4{\cdot}7H_2O$ [17]. After coagulation, solar photo-Fenton oxidation removed up to 94% of COD. In this process, Fe^{2+} should be chosen to promote the regeneration of Fe(II). Otherwise, at higher Fe(II) concentration, the penetration efficiency of light decreases. At lower Fe(II) concentration, it accelerates consumption of H_2O_2 without the generation of ·OH, which also explained the low efficiency in high H_2O_2 concentration. Furthermore, the solar photo-Fenton-treated effluents contained fewer toxic matters and lead to higher nutrient uptake by plant. It appears to an efficient and economic technology for plants which can take advantage of sunshine. A similar technology was applied without solar photocatalysis, and the removal efficiency of COD decreased to 75% [33]. However, it increased to 95% after 60 days biological treatment since the process increases the biodegradability. Similar effects for different dosages of Fe^{2+} were observed. Scale-up experiment in industry indicates that adjusting the reagent dosages by monitoring the concentration of COD is necessary to optimize efficiency and cost. Compared with lab scale, decreased efficiency of degradation might be a result of the complexity of real matrices.

Optimal operation condition for electrocoagulation (ECR) and photocatalytic (PCR) process were investigated [34]. For ECR, neutral pH and higher current density are chosen to optimize the solubility of aluminum hydroxide, which enhances electrocoagulation of COD, phenol, and color. For PCR, different types of catalyst, including ZnO and TiO_2, had similar removal efficiency of COD and color. But ZnO has higher efficiency in phenol removal while also having a cost advantage. Sequential application of ECR before PCR results in higher degradation in COD and phenol.

Fenton process followed by ozonation has also been optimized [35]. Considering removal efficiency and cost of ozone, 90 min was found to be the optimal reaction time. However, subsequent biological treatment processes are needed to meet regulatory guidelines for COD and color.

Oxidation of OMW by microwave (MW)-activated persulfate is a promising pre- or post-treatment step [36]. Based on dielectric heating principle, MW irradiation is efficient in heating, which activates persulfate and breaks down phenolic compounds which have dark brown color. Hence MW counts for main removal of TOC, operating cost, and a portion of color reduction, while pH is the dominant factor in reducing color.

Rich in nutrient and color, more OMW is degraded by Fenton process in combination with pre-treatment technologies after 2016, compared with previous comprehensive literature review [13]. Factors involved in Fenton process are investigated extensively, including dosage of reactants and catalyst, pH, and reaction time. For further study, different combination of conventional pre-treatment methods and Fenton process can be analyzed. In addition, great effort for scale-up study and cost evaluation is needed.

3.3 Meat processing industry

The meat processing industry produces large volumes of slaughterhouse wastewater (SWW). SWW contains elevated amount of organic matter, suspended solids, oil, grease, and toxic matters [37]. Anaerobic treatment is efficient in removing organic matter with low costs in addition to the generation of methane. However, complementary treatment is necessary for the effluents to meet the required discharge limits [38]. As post-treatment method, AOPs are a promising supplementary for the entire process in removing non-biodegradable organics and inactivated microorganisms producing hazardous by-products [37] as summarized in **Table 5**.

UV/H_2O_2 has been applied to degrade slaughterhouse effluents pretreated by anaerobic baffled reactor (ABR) and an aerobic activated sludge (AS) reactor connected sequentially [37]. RSM was used to optimize the operation condition in order to maximize removal efficiency of TOC and TN and CH_4 yield, as well as minimize H_2O_2 in effluent.

This process can be enhanced by silver-doped TiO_2 nanoparticles as photocatalyst [42]. Electrons captured by silver react with oxygen more efficiently than those combined with electron hole in TiO_2. However, application of silver and further treatment for separation of photocatalyst make it an uneconomic technology for practical use.

The combined UV-C/H_2O_2-VUV was studied with the similar method used by Bustillo-Lecompte, but a Box–Behnken design was used instead of the CCD [39]. The combined process removed more TOC and minimized H_2O_2 simultaneously compared with individual processes. Furthermore, this technology performs better if applied after biological treatment, since the respirometry analyses indicated low biodegradable degree. Wastewater pretreated by anaerobic digestion can yield up to 90% COD removal but remains yellow in color after 30 days [41]. Further treatment is necessary to remove more COD and eliminate color compounds, which can be achieved by SPEF with BDD anode. SPEF results in higher decline of absorbance and COD than photolysis only and less cost compared with electron-Fenton (EF) [42] and Fenton process only [40].

Compared with conventional disinfection methods, cold plasma technology has the advantage of less by-product and high removal efficiency [43]. This technology achieved high removal efficiency of pathogens, organics, and inorganic pollutants at the same time with enough hydraulic retention time. Furthermore, other operation condition for cold plasma can be investigated.

Various AOPs combined with biological treatment methods have been applied. Treatment of wastewater from meat processing industry meets specific challenge including methane production. Different experiment strategies are performed to study the factors. Cost evaluation in scale-up plant is needed considering both removal efficiency and overall economy, including cost and profits of CH_4.

3.4 Dairy wastewater

The consumption of dairy production in Canada increases gradually. Correspondingly, more wastewaters need to be treated. Dairy industries' wastewaters come from processing milk and system management and typically contain large

Meat processing plants wastewater origin	Treatment process	Observations	Reference
BOD 1209 mg/L COD 4221 mg/L TN 427 mg/L TOC 546 mg/L TP 50 mg/L TSS 1164 mg/L pH 6.95 Slaughterhouse effluents in Ontario, Canada	An anaerobic baffled reactor (ABR), followed by an aerobic activated sludge (AS) reactor, and a UV/H_2O_2 photoreactor (ABR-AS-UV/H_2O_2 processes)	TOC 97.8% H_2O_2 residual 1.3% TOC 50 mg/L, flow rate 15 mL/min, H_2O_2 dosage 344 mg/L, pH 7.2	[37]
Slaughterhouse effluents in Ontario, Canada	Combined UV-C/H_2O_2-VUV	TOC 46.19% H_2O_2 residual 1.05% TOC 213 mg/L H_2O_2 dosage 450 mg/L Irradiation time 9 min	[39]
pH 7–8.3 COD 25–32 g/L Volatile solid (VS) 3.31–9 g/L TS 5.32–11.5 g/L Total coliforms MPN/100 mL 90x105 Fecal coliforms MPN/100 mL 20 × 105 EC 550–900 μS/cm TDS 0.9–1 g/L Color 800–1197 Pt/Co SVI 900–950 mL/g	Fenton process	TS 50% VS 61% COD 53% Color 61% pH = 3 Reaction time 150 min $c(H_2O_2) = 4000$ mg/L	[40]
TOC 132 mg/L COD 480 mg/L BOD5 267 mg/L Slaughterhouse in Puente Alto, Santiago, Chile	Anaerobic digestion followed by solar photoelectron-Fenton (SPEF)	COD 97% Turbidity and solids total removal CH4 accumulation 90 mL pH 3 Fe^{2+} 1 mM Reaction time 180 min	[41]
pH 6.687 BOD 1078.45 mg/L COD 2024.5 mg/L N 74.8 mg/L Slaughterhouse effluent from Lahore, Pakistan	Photocatalytic oxidation assisted with TiO_2 and silver-doped TiO_2 nanoparticles	BOD 95% COD 87% N 74% Ag-TiO_2-H_2O_2 under UV (400 Watt)	[42]
COD 7.4 g/L TN 609.9 mg/L TP 44.1 mg/L T-Fe 66.9 mg/L Toxic unit (TU) 13.8 Total coliforms 267,000 CFU/ml Slaughterhouse effluent from Nonsan, Korea (average)	Cold plasma	COD 78–93% TN 51–92% TP 35–83% T-Fe 93% Bacteria 98% TU 96%	[43]

Table 5.
Recent application of AOPs on meat processing industry.

amounts of organic materials, suspended solids, oils, salts, and fats [44]. The following AOPs (**Table 6**) are studied to degrade excessive pollutants.

Kinetic models describing the COD degradation of flotation/ozonation processes have been developed and compared [45]. The highest kinetic constant can be achieved for ozonation at acidic medium. The results imply that pre-treatment is necessary to remove the scavengers of HO· in milk. Moreover, participation of casein at low pH might also contribute to the removal of COD. The process could be enhanced further by the addition of H_2O_2 [46], which was then optimized via a CCD (pH, dosage of H_2O_2, ozone, and catalyst). Ozonation is also efficient in decomposing antibiotic in milk based on electrophilic attack by O_3 rather than generated HO· [52].

RSM has been applied to optimize the operation condition of electro-Fenton process with iron electrodes [47]. In order to maximize the removal rate of color and COD, five factors are investigated, including reaction time, current density, pH, H_2O_2/DW (mL/L), and molar ratio. The results reveal strong interactions between pH and molar ratio, as well as between pH and current density. Increasing the current density below the optimal point accelerates Fe^{2+} regenerated from Fe^{3+}, but higher current density results in the generation of O_2 and H_2. Longer reaction times increase removal efficiency, while high pH results in iron ion precipitation, and low pH affects H_2O_2 decomposition into ·OH. Similar process analyzed via RSM was done for Fenton only processes [48]. The electro-Fenton process has the advantage of less consumption of chemicals. Besides removal of chemicals, disinfection of dairy effluent has also been studied [51]. The electrocoagulation process followed by electro-Fenton (EF) or UVA-assisted photoelectro-Fenton (PEF) is suitable for this objective. The results show that EF and PEF are more efficient in inactivation. PEF is better than EF because of UVA radiation.

TiO_2 has also been used as a catalyst to disinfect dairy wastewater [8] which is used as an irrigation water resource. The results show that disinfection efficiency is higher with the addition of TiO_2 and that oxygen accelerates the photocatalysis process. The removal efficiencies of COD, BOD, and SS increased with TiO_2 as photocatalyst illuminated by UV light combined with biological treatment compared with photocatalytic UV reactor only [49].

AOPs in dairy wastewater often operate with biological treatment methods. The operation conditions are optimized by RSM and well explained. Other than elimination of general TOC, COD, and BOD, dairy wastewater treatment also requires disinfection for further application.

3.5 Miscellaneous food industry wastewaters

The diversity of the food industry results in a multitude of wastewater effluents. They are often rich in refractory organics and color compounds. The following table presents existing technologies in other types of food industries (**Table 7**). AOPs have also shown capability for degradation of these effluents.

Molasses used in the production of baker's yeast result in dark color and high organic load of wastewater. The color compound is recalcitrant to aerobic and anaerobic processing. Ultrasonic irradiation assisted by TiO_2-ZnO decolorized the effluent by 25% [53] after optimization (factors: reaction temperature, catalyst composite and calcination temperature, and catalyst load). However, the COD did not change because ultrasonic irradiation transformed organics into smaller intermediate.

In beverage production, 1.72 L wastewater is produced for every 1 L beverage [54]. Photocatalytic processes have been used to degrade synthesized beverage wastewater effluent [54], showing advantages of cerium doped ZnO and favorable

<table>
<tr><th>Dairy wastewater origin</th><th>Treatment process</th><th>Observations</th><th>Reference</th></tr>
<tr><td>COD 2000 mg/L
Simulate WW: whole milk + distilled water</td><td>Flotation/ozonation</td><td>More than 80%</td><td>[45]</td></tr>
<tr><td>COD 2000 mg/L
Simulate WW: whole milk + distilled water</td><td>Flotation followed by O_3/H_2O_2 and O_3/Mn^{2+}</td><td>64.5%
O_3 42.9 mg/L
H_2O_2 1071.5 mg/L
pH 10.9</td><td>[46]</td></tr>
<tr><td>COD 2527 mg/l
Color 100
pH 6.27
Conductivity 210 μs/cm
TDS 981 mg/L</td><td>electro-Fenton process</td><td><table>
<tr><th>Factors</th><th>COD</th><th>Color</th></tr>
<tr><td>Removal</td><td>91.76</td><td>95.22</td></tr>
<tr><td>Current density (mA/cm^2)</td><td>56</td><td>55.1</td></tr>
<tr><td>Reaction time (min)</td><td>90</td><td>86</td></tr>
<tr><td>pH</td><td>7.52</td><td>7.48</td></tr>
<tr><td>Molar ratio H_2O_2/Fe^{2+}</td><td>3.965</td><td>3.987</td></tr>
<tr><td>H_2O_2/DW (mL/L)</td><td>0.898</td><td>0.907</td></tr>
</table></td><td>[47]</td></tr>
<tr><td>COD 6055 mg/L
TS 11900 mg/L
TSS 1320 mg/L
Color (Pt-Co) 1700
pH 5.7
Conductivity 6 mS/cm
Wastewater treatment plant produces milk, yogurt, and butter</td><td>Electro-Fenton process</td><td>electro-Fenton
COD 72%
Orthophosphate 88%
SS 92%
Color removal efficiencies 92.5%
H_2O_2/COD ratio 2, current density 32 mA/cm2, pH 2.4 and reaction time 45 min</td><td>[48]</td></tr>
<tr><td>pH 6.5–7.5
Turbidity 2–5 NTU
DO 6.5–7.5 mg/L
Microbes 2300e2900 CFU·mL^{-1}
Dairy wastewater after treated by activated sludge process (extended aeration)</td><td>Solar photocatalysis (ph-C S)
concentrated solar photocatalysis (ph-C CS)
solar photolysis (ph-L S)
concentrated solar photolysis (ph-L CS)
TiO_2 as catalyst</td><td>Disinfection efficiency
ph-C S 41%
ph-C CS 97%
ph-L S 10.5%
ph-L CS 68.9%
Reaction time 30 min</td><td>[8]</td></tr>
<tr><td>COD 876 ± 255 mg/L
BOD 33 ± 10 mg/L
SS 580 ± 159 mg/L
Conductivity 1.6 ± 0.4 mS/cm
pH 7.9
DO 1.8 ± 0.3 mg/L
Dairy effluent after a three-step piggery wastewater treatment (TPWT) system, involving (1) solid/liquid separation, (2) anaerobic treatment, and (3) aerobic</td><td>Photocatalytic UV reactor (UVR) followed by bioreactor</td><td>Photocatalytic + biological
COD 72%
BOD 98%
SS 79%
UV only
COD 53%
BOD 62%
SS 62%</td><td>[49]</td></tr>
</table>

Dairy wastewater origin	Treatment process	Observations	Reference
treatment (activated sludge basin with a final clarifier)			
Wastewater after anaerobic and aerobic ponds	Ultraviolet/persulfate (UV/PS) oxidation		[50]
pH 5.7 ± 0.2 Conductivity 2.95 ± 0.12 mS/cm TOC 1416 ± 24 mg C L^{-1}	Electrocoagulation (EC) with Fe electrodes followed by electro-Fenton (EF) or UVA-assisted photoelectro-Fenton (PEF) with BDD or RuO_2-based anode	Not available	[51]

Table 6.
Recent application of AOPs in the dairy industry.

Wastewater origin	Treatment process	Observations	Reference
Absorbance at 400 nm 0.3–0.4 pH 5–6 COD 4800–5400 mg/L Baker's yeast factory effluent (Turkey)	Ultrasonic irradiation TiO_2-ZnO as sonocatalyst	Decolorization 25% COD 22.4% (no reduction by ultrasound) Ultrasonic irradiation 20 kHz, 200 W TiO_2/ZnO 4:1 molar ratio (0.15 g/L) 700°C for 60 min	[53]
COD (mg/L) 500–3000 pH 4.7 EC (μs/cm^2) 1845 Turbidity (NTU) 100 Total dissolved solids (TDS) (mg/L) 850 Synthesized beverage industry effluent	UV and solar illumination assisted by synthesized catalyst, immobilized cerium doped ZnO	Photodegradation efficiency* Catalyst dosage 1–5% 35.33–51.56% (UV) 19.86–32.45% (visible light) COD initial concentration 500–3000 mg/L 65.14–21.9% (UV) 42.13–10.12% (visible light) Reaction time 120 min	[54]
COD 15290 ± 855 mg/L TSS 14950 ± 2400 mg/L Palm oil mill effluent (POME) from Rantau, Malaysia	Coagulation by chitosan, addition of ferrous sulfate ($FeSO_4$), chitosan with hydrogen peroxide (H_2O_2), and chitosan with Fenton oxidation	COD 82.82 ± 1.71% TSS 89.92 ± 0.48% Chitosan (2500 mg/L) with H_2O_2 (500 mg/L) pH 7 Reaction time 15 min mixing +1 h sedimentation	[55]
pH 3.6–4.5 Conductivity (uS/cm 20 C) 450–550 TSS (mg/L) 240–280 COD (mg/L) 10,000 BOD_5 (mg/L) 4246–5252 DOC (mg/L) 4218–4260 NTU 130–160 Color 450–100 BOD_5/COD 0.43–0.53 Diluted citrus effluents from Cuba	1. Ozone-based processes (O_3, O_3/OH^-, O_3/UV, O_3/H_2O_2, and $O_3/UV/H_2O_2$) 2. Solar photo-Fenton treatment	1. Ozone-based processes COD 15.7% DOC 10.9% pH 7 Ozone 1.9 g/L UV 254 nm Reaction time 150 min H_2O_2 1017 mg/L 1. Solar photo-Fenton treatment COD 77% DOC 53%	[56]

Calculation based on adsorption reading through spectrophotometry.

Table 7.
Recent application of AOPs in miscellaneous food industries.

effect of an acidic environment. Large amounts of wastewater effluents result from the fast growth of the palm oil industry [55]. AOPs utilizing $FeSO_4$ or H_2O_2 and the contribution of chitosan as flocculant have been investigated for such water. Process optimization revealed opposite fluctuation of removal efficiency for COD and TSS with different combination of reagents. Furthermore, chitosan (2500 mg/L) with H_2O_2 (500 mg/L) results in the best removal efficiency as post-treatment of anaerobically digested POME [55].

The effluents of citrus fruit processing facilities are characterized by acidity, presence of essential oils, and toxicity. Diluted real citrus juice wastewater was investigated with various ozonation process, and it was found that the solar-Fenton process is better both in removal efficiency and economic perspective [56].

In the coffee industry, wastewater effluent treatments have recently been reviewed elsewhere [57]. The main characteristics of these streams are the presence of various color compounds and macromolecules. AOPs have been proven to be efficient in combination with biological treatment [58]. However, considering the nature of color compounds, ion exchange is a more promising field.

Wastewater treatment for different food or beverages should be adjusted according to the characteristics of different effluents such as complex color compounds (yeast and coffee) or low pH (juice). Pre-treatment such as coagulation or anaerobic digestion is favorable.

3.6 Food dyes

Apart from wastewater effluents from food processing factories directly, municipal wastewater contains some refractory compounds from food industries. Food dyes are extensively used in fruit juices and sweets products as food additives. The colored effluents from such industries need to be treated carefully before discharge. Otherwise they will lead to issues such as pollution on esthetic grounds and interference of light transmission [59].

Food dye	Treatment process	K_{app}	Reference
Ponceau 4R	Electro-oxidation 1. Electrogenerated H_2O_2 ($EO–H_2O_2$) 2. Electro-Fenton (EF) 3. Photoelectro-Fenton (PEF)	(10^{-2} min^{-1}) 1. 2.72 ± 0.41 2. 12.31 ± 0.53 3. 13.35 ± 0.66 (real water matrices)	[61]
Amaranth food dye (AM)	Heterogeneous E-Fenton process with synthesized $Fe_3 - xCu_xO_4$ ($0 \leq x \leq 0.25$) NPs	4.2×10^{-2} min^{-1} (x = 0.25)	[62]
Brilliant blue FCF (BBF)	Fe_3O_4-TiO_2(FTNs) assisted with different UV light	0.059 min^{-1} (FTNs/UVA/PMS)	[63]
Carmoisine (E22)	UVA-LEDs/PMS/Fe^{2+}	0.1553 min^{-1}	[64]
Tartrazine	Visible light photo-Fenton oxidation with three bismuth oxyhalide catalysts	0.0026–0.06 min^{-1} Temperature from 30–70°C	[65]
Sunset yellow FCF (SY)	Electrochemical assisted with palladium-ruthenium nanoparticles incorporated with carbon aerogel (Pd-Ru/CA)	0.295 s^{-1}	[66]
Tartrazine yellow (TT) and brilliant blue (BB)	Photocatalytic process	0.016 min^{-1}	[67]

Table 8.
Recent application of AOPs in food dyes.

Azo compounds are widely used in food industries because of its brilliant shades, relative low cost, and simple manufacture [60]. However, azo compounds are resistant to conventional treatment due to one or more azo bonds [61]. Hence the following AOPs (**Table 8**) are applied to degrade specific food dyes.

Fenton process is widely used. Thiam et al. proposed the routes for ponceau 4R degradation. The Fenton process was found to provide fast ·OH production and stability despite the interference of real water matrices [61].

Fenton processes can be enhanced by various catalysts. In order to circumvent the pH limitation of Fenton processes, catalysts with immobilized iron ions are synthesized to avoid precipitation. The combination of magnetite (Fe_3O_4) and copper presents high specific surface area and synergic effects between Cu^{2+}/Cu^{+} and Fe^{3+}/Fe^{2+}. Moreover, it is easy to separate [62]. Similarly, Fe_3O_4-TiO_2 (FTNs) [63] as well as metal doped bismuth oxyhalide catalysts (BiOCl, Cu-BiOCl, and Fe-BiOCl) have been used to degrade BBF [65].

Compared with conventional UVA, UVA-LED are more efficient and cost beneficial. The implementation of UVA-LED accelerates the regeneration of radicals. [64]. Zazouli et al. also suggest that UVC, a high energy UV source, is more efficient in decomposing PMS into ·OH and $SO_4{\cdot}^-$ than UVA [63]. The replacement of batch reactors by flow reactors is another promising modification [67].

In dealing with a specific food dye, catalysts are synthesized to assist AOPs. The operation conditions such as catalyst dosage and UV source have to be considered. Further studies are required related to economic catalyst development and ease of separation and recovery.

4. Practical application of AOPs

Wastewater treatment in the food industry with AOPs is a flourishing field. Nevertheless, pilot-scale plants are not widely studied. A number of studies implement AOPs in sequential approaches with a complete solution for the treatment and reuse of a complex wastewater from food industries.

Yalılı Kılıç et al. investigated the pilot-scale treatment of olive oil mill wastewater [68]. The combination of physicochemical treatment, ultrafiltration, and $O_3/H_2O_2/UV$ presented the most favorable results of pollutant removal. However, cost evaluation indicates that the absence of ozone is more economic without removal efficiency diminishment. Another study applied Fenton process in a CSTR at pilot scale [69]. The results highlight the importance of operation condition optimization in actual plant condition. However, such a process is not affordable for small facilities generating large amount of pollutants.

In the winery wastewater treatment field, ozone-based AOPs (O_3/UV and $O_3/UV/H_2O_2$) at pilot-scale bubble column reactors have shown high efficiency in TOC removal. Moreover, addition of H_2O_2 is favorable considering overall costs [70]. One of the origins of winery wastewater is cork boiling water. AOPs including solar photo-Fenton and ozone have been applied to the effluents pre-treated by physicochemical methods. [71] The pre-treatment reduces the additional benefits of AOPs such as increased biodegradability and toxicity reduction. Further assessments of overall costs remain to be investigated.

5. Conclusion

In the food industry, wastewater effluents including complex organics are generated from various food processing steps and equipment maintenance procedures.

Moreover, the concentration of containments varies according to the type of food. This situation leads to an urgent demand for AOPs as complementary technologies to traditional wastewater treatment which is insufficient to process excess pollutants. The effluents from AOPs are more biodegradable for biological treatments, and hence the addition of AOPs as pre-treatment or post-treatment is a promising and economical solution for processing various wastewaters in the food industry.

Further research on catalysts that increase the amounts of effective radicals, simple separation procedure, and high recovery rates is suggested. Reactor design and optimization is another promising field. Due to the small number of practical applications, additional pilot-scale studies with AOPs are also recommended. Large-scale studies can provide overall cost evaluation including capital cost, operation cost, and possible profits from by-products. Such integrated economic assessment in real plants will be valuable guidance for future research.

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