A comprehensive overview of microwave-assisted oxidation of industrial wastewaters

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Abstract
In the present study, recent progress in using microwave energy to enhance oxidative degradation of pollutants of poor biodegradability in industrial wastewaters is reviewed and evaluated, in order to assess the potential of microwave-induced oxidation as effective and viable remediation technique, alternative to conventional treatment procedures. Microwave radiation is actually emerging as an innovative technology, widely applied in various fields, as it offers many advantages for environmentally-friendlier processing. Microwave energy has already been employed in several environmental applications. In particular, the use of microwave technology to to overcome intrinsic drawbacks and improve oxidation/degradation methods for various wastes appears to be a challenging scientific area. Certainly, the oxidation efficiency is influenced by key operating factors including the microwave equipment and power level, irradiation time and temperature, oxidizing agent and catalyst loading, aeration, pH and initial pollutant concentration. In conclusion, microwave irradiation can contribute to enhancing the oxidation process, thus accelerating pollutant purification reactions, decreasing the consumption of chemicals and destroying microorganisms at low temperatures.

Keywords
Microwave energy; environment-friendly; oxidation; degradation; industrial wastewaters

INTRODUCTION

In the present work, recent advances in employing the microwave technology to enhance oxidation/degradation of pollutants, especially in industrial wastewaters, are reviewed and summarized, in order to assess their potential as viable and efficient remedial alternatives to conventional procedures.

Actually, microwave radiation is emerging as a novel technology that has gained widespread acceptance as an effective thermal method. Microwave energy offers many advantages, including a rapid heat transfer and a volumetric and selective heating, for energy-efficient, thus eco-friendlier, industrial processing over conventional techniques. Hence, microwave processes can be an effective and economic approach to reduce the treatment time considerably, thereby leading to substantial energy and cost savings. These characteristics provide sufficient motivation to promote the use of microwaves in "greener" processing. So far, microwave technology has already been employed in various environmental applications, including environmental and green chemistry, sintering for transformation of lignite ashes into ceramics, determination and/or oxidation/remediation of soil pollutants, solid waste treatment, anaerobic digestion feedstock pretreatment, disinfection of medical wastes, synthesis of shutle-like zinc oxide nanoparticles, and also pyrolysis for energy recovery and conversion of biomass into valuable energy products (Carrere et al. 2016; Peng et al. 2015; Mushtaq et al. 2015; Koo and Jeong 2015; Djahaniani et al. 2015; Tsukui and Rezende 2014; Karayannis et al. 2013; Horikoshi 2012).

For the degradation of various pollutants of poor biodegradability, such as antibiotic wastewater, pharmaceutical wastewater, aqueous phenanthrene, insecticides (e.g. imidacloprid), 17α-ethynylestradiol in secondary-treated wastewater, dispersive textile dyes, especially reactive azo dyes, brilliant blue, methylene blue etc., by their transformation into non-toxic substances, several advanced oxidation processes, including sono/-photo/-Fenton/-like degradation, heterogeneous UV or simulated solar photocatalytic processes, photochemical treatment with H_2O_2/UV, photoelectrocatalytic oxidation, and even high-frequency ultrasound degradation, have recently received much attention and have been increasingly applied as potentially efficient treatment methods, using new photocatalysts such as TiO_2 with photocatalytic activity influenced of gamma-irradiation, Fe(III)/TiO_2-montmorillonite or even TiO_2/Ti electrodes (Alvarez-Corena et al. 2016; Fatimah et al. 2015; Frontistis et al. 2015; Soutsas et al. 2010; Philippidis et al. 2009). Actually, the end effect of these processes is the production of hydroxyl radicals (˙OH) that have a very strong oxidation potential thus being powerful oxidizing agents. For assessing the quality of effluents prior to discharge, chemical oxygen demand (COD), estimating the amount of oxidizable organic matter in effluents, is the parameter usually monitored (Amanatidou et al. 2012; Moustakas and Malamis 2015). However, the energy consumption and operational cost of these oxidation methods can vary from pollutant to pollutant and also depends on their loading rates, especially in full-scale operation of industrial interest. Moreover, some of these methods require chemical reagents that may cause secondary pollution. Besides, traditional chemical oxidation processes, such as ozonation and chlorination often used in wastewater reclamation, may result in by-products potentially altering the toxic and mutagenic properties of effluents (Kalavrouziotis et al. 2011; Shang et al. 2006). Therefore, the use of microwave irradiation to improve degradation processes for many types of wastes and overcome the aforementioned drawbacks emerges to be a challenging scientific area.

MICROWAVE-ASSISTED OXIDATION OF INDUSTRIAL WASTEWATERS

The most commonly used microwave frequency for industrial purposes (2.45 GHz) corresponds to significant penetration depth within most of the materials, and therefore it is suitable for most reaction conditions. In fact, microwave energy can contribute to accelerating pollutant purification reactions, enhancing the oxidation process and diminishing hazardous products formation, while also decreasing the consumption of chemicals. It is demonstrated in the following reported literature that the assistance of microwave irradiation, not only consists in a beneficial thermal effect alone, as the same result cannot be achieved by conventional caloric methods, but also in an increased formation of hydroxyl radicals that is likely due to some sort of surface restructuring induced by the microwave energy. Such non-thermal effects of microwave are able to enhance various chemical reactions, treat wastewater systems, and destroy microorganisms at low temperatures.

Particularly in TiO_2 photocatalytic oxidation of various pollutants, the influence of microwaves can provide significant enhancement of photodegradation kinetics, as assessed by several recent studies. In particular, a microwave non-thermal effect involving lattice distortion and oxygen vacancies in an appropriately treated TiO_2 photocatalyst beneficially affects the catalyst photoactivity, thus accelerating reaction kinetics during microwave-assisted photodegradation of 4-chlorophenol under various irradiation conditions (Horikoshi et al. 2013). It should be noted that the UV/H_2O_2/MW combined process appears even faster than other process variations (MW, H_2O_2, H_2O_2/MW, and UV/MW) for the color removal of an azo-dye (tartrazine) solution (Parolin et al. 2013). In another research, the combined A-TiO_2/AC/MW process (nano-sized anatase or rutile TiO_2-supported activated carbon), which was developed for the microwave (MW)-induced degradation of parathion, displayed many advantages in comparison with the MW/AC degradation, and the supported A-TiO_2/AC showed higher MW catalytic activity than R-TiO_2/AC, thus providing a promising approach for the removal of parathion in wastewater treatment applications (Zhang, Z. et al. 2013). A synergy effect upon application of microwave energy together with UV irradiation, ozone, and a photocatalyst seems to play an important role in the O_3-assisted photocatalysis, resulting in the highest rate constant, being 4.5 times that obtained with MW/UV/photocatalyst combination and more than 6 times that obtained by injection only of ozone (Lee et al. 2015). Besides, a novel microwave-enhanced photocatalytic membrane distillation process was proposed for the treatment of organic wastewater containing inorganic ions. Coupling microwave energy with UV irradiation effectively decreased the deposit on membrane surface by destabilizing complex compounds, and the beneficial effect of microwaves on the membrane distillation process was proved to be sufficient for a COD removal rate higher than 96% when applied to coal gasification wastewater treatment (Wang et al. 2016). Also, microwave discharge electrodeless lamps were investigated as light source in the presence of H_2O_2 for the photooxidation of guaiacol to value-added carboxylic acids. It should be noted that the production of chemicals from lignin sources, such as guaiacol, by green and sustainable technologies is examined as a promising alternative route for the substitution of fossil fuels. This combined microwave-assisted heating and photooxidation with special ultraviolet lamps in the presence of hydrogen peroxide led to a more effective degradation of guaiacol than with conventional photocatalysis, thus possibly being a promising green and economically feasible alternative approach for lignin oxidation to produce organic acids (Zhang et al. 2016).

Also, Fenton (Fe^{2+}/H_2O_2) and/or Fenton-like (Fe^{3+}/H_2O_2) process assisted by microwave energy, appears to be an innovative and less expensive solution, with advantages of highly effective and fast processing, for the treatment of wastewater containing organic contaminants, while also permitting to optimize the operational parameters, especially to reduce the necessary iron catalyst concentrations, thus satisfying environmental demands. Indeed, by the assistance of microwave irradiation, oxidative degradation of amoxicillin was significantly improved over classical Fenton's reaction (Homem et al. 2013). Moreover, the application of microwave-enhanced Fenton oxidation noticeably accelerated the conversion of methylene blue, due to the fast and selective heating of water and hydrogen peroxide molecules (Liu et al. 2013). Also, a certain synergy between microwaves and Fenton oxidation system could shorten the reaction time, greatly improve the COD removal rate wastewater and even reduce the dosage of Fenton reagent, for dye wastewater containing the weak acid brilliant red B and the anionic surfactant SDS (You at al. 2013). Such synergistic effect resulting in promotion of dye degradation in dye wastewater was investigated systematically even in the application of microwave irradiation/Fenton oxidation coupling coagulation process (Zhang, G. et al. 2013). Furthermore, the microwave-enhanced Fenton reaction followed by hydroxide precipitation was investigated in the oxidation of EDTA, which can form very stable complexes with heavy metal ions, greatly inhibiting conventional metal-removal technologies. The COD reduction order achieved, from high to low, was found to be Cu(II)-CNi(II)-CEDTA ≈ Cu(II)-CEDTA >Ni(II)-CEDTA (Lin et al. 2016).

A non-thermal mechanism based in advanced oxidation pathways through holes and $^{\cdot}OH$ radicals generation was mainly proposed for the removal of pyridine (a toxic and volatile N-containing organic pollutant, occurring in effluents from herbicides and pesticides manufacturing industries) by microwave irradiation, this appearing to be an effective alternative degradation technique (Zhang et al. 2016). For the microwave-induced degradation of another herbicide, atrazine, the role of surface chemistry in mineral micropores of a solvent-sorbate-sorbent system was also investigated. Actually, surface chemistry affects the interactions of sorbate and solvent molecules with the pore wall surfaces of microporous minerals, thus influencing the transmission and absorption of microwave energy (Hu and Cheng 2013). The aforementioned substantial performance improvements in the oxidation of several organic pollutants by coupling microwave energy to advanced oxidation processes would easily justify additional capital and electricity costs, although the degradation mechanism and variation in the formation of intermediate species remains relatively obscure (Nascimento and Azevedo 2013).

Besides, it seems a promising approach to combine microwave irradiation with chemical oxidation (MW/H_2O_2) and $MW/S_2O_8^{2-}$) for the treatment of biological waste sludge in order to degrade adsorbed micropollutants that can be problematic for the safe reuse or disposal of biosolids (Bilgin Oncu and Akmehmet Balcioglu 2013). Indeed, the advantages offered by the application of microwave irradiation techniques over conventional methods for effective sludge treatment and generation of environmentally clean and value-added end products are also outlined in other relevant studies (Jang and Ahn 2013; Mehdizadeh et al. 2013; Tyagi and Lo 2013).

Moreover, for the rapid activation of persulfate and thus of production of $SO_4^{-\bullet}$, a powerful oxidant, the combination of microwave irradiation with catalytic ion Ag+ is showed to be an efficient method for the degradation of dimethyl phthalate (a pollutant of concern in aqua media) (Sun et al. 2013). From the contribution of both microwave irradiation and persulfate oxidation in treating landfill leachate, specific effects occur, usually additive and even synergistic or exceptionally antagonistic (Chou et al. 2013a). In further study regarding the treatment of landfill leachate, the previous researchers compared formation and degradation behaviors of organic acids under microwave oxidation process and under conventional heating oxidation and explored derivative mechanisms of organic acids in microwave-assisted oxidation (Chou et al. 2013b).

Activated carbon has also been extensively used in microwave catalysis technology for environmental pollution abatement (Li et al. 2013). Particularly for the pretreatment of persistent old-age landfill leachate, microwave-assisted catalytic oxidation in the presence of activated carbon displayed superior treatment effectiveness compared to each separate process (Li et al. 2013). In petroleum industry, a significant synergetic effect is observed between microwave irradiation and Fe^0/granular activated carbon micro-electrolysis system for the pretreatment of refinery wastewater. Actually, the biodegradability of the wastewater was improved by approximately 60% (in terms of COD removal efficiency) by the microwave-enhanced micro-electrolysis treatment, thus rendering the pretreatment process favorable for the subsequent biological process (Qin and Gong 2014). Also, milder reaction conditions can be applied and higher rates be achieved for petroleum oxidative desulfurization under microwave treatment compared to conventional heating technologies (Shang et al. 2013).

Furthermore, under microwave-irradiation, MnO_2, a relatively mild reagent, is reported to act not only as a microwave absorber but also an efficient oxidizer for the high-yield oxidation of arylmethylene compounds to the corresponding aldehydes and ketones as well as benzylic ethers to esters (Nammalwar et al. 2013) or in sulphuric acid solution for the rapid removal of polychlorinated biphenyls from soil contaminated by capacitor oil (Lin et al. 2013). Also, ε-MnO_2 (akhtenskite) is proved to be a kind of excellent microwave catalyst for the degradation of tetracycline in water by microwave-induced oxidation (Peng et al. 2014).

Recently, a new microwave catalytic oxidation process was reported, based on two kinds of catalysts, the commercially available activated carbon (AC) and Mn_2O_3 nanoparticle modified activated carbon (Mn_2O_3/AC), without adding any oxidant, for the degradation of 4-nitrophenol, one of the most important derivatives of phenol, intermediate of pesticide, pharmaceuticals, dyes, oil refineries, and organic chemical manufacturing, which can be largely harmful for humans even at low concentration. Actually, the new Mn_2O_3/AC catalyst showed much higher catalytic activity than pure AC and Mn_2O_3 particles. Indeed, 4-NP degradation efficiency reached 99.6%, corresponding to 93.5% TOC removal under optimal conditions, which was attributed to the generation of ·OH radicals due to the microwave 'photoelectric effect' (Yin et al. 2016). Furthermore, microwave catalyst of pristine Bi_2O_3 was developed for the highly efficient treatment of p-nitrophenol (PNP) polluted water by microwave catalytic oxidation process (Qiu et al. 2016).

CONCLUSIONS

- Microwave energy can contribute to enhancing the oxidation process of organic pollutants in industrial wastewaters, thus accelerating purification reactions, decreasing the consumption of chemicals and destroying microorganisms at low temperatures.
- Microwave equipment and power level, irradiation time and temperature, oxidizing agent and catalyst loading, aeration, pH and initial pollutant concentration are key operating factors influencing the oxidation efficiency.
- In conclusion, microwave-assisted oxidation processes appear to be promising alternatives for the treatment of pollutants of poor biodegradability. It should be noticed, however, that full-scale application for real industrial wastewaters still remains underway.

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