Sonochemical catalytic removal of antibiotics using converging ultrasound and cavitation agent with life cycle assessment

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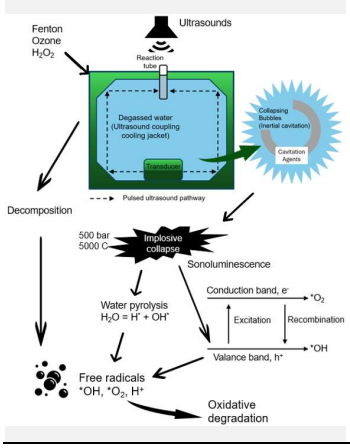

Antibiotic exposure to wastewater, such as tetracycline, has profound environmental and health implications. Yet conventional treatment methods are not fully effective, and the potential environmental impact of traditional AOP applications is not generally considered. Alternative strategies, such as sonocatalytic oxidative processes, are gaining interest. In this work, we investigated the degradation of tetracycline using a combination of cavitation agent (CA) and sonochemical reactor with converging ultrasound and UV, which resulted in a six times higher reaction rate compared to either standalone UV or standalone ultrasonic treatment. Accounting for the impact of energy consumption and the material required to remove a certain amount of pollutant, the use of CA allowed a 38% reduction in CO_2 footprint and proved to be more competitive in 14 of 18 indicators of environmental impact. It is crucial to consider both the environmental impact associated with material acquisition and synthesis and the reaction rate of contaminant degradation in order to fully understand the impact of new AOP technologies.

Introduction

There is current concern over human and environmental overexposure to micropollutants and the lack of effective treatment technologies. The principal aim of this project is to investigate and optimize novel sonochemical methods to remove micropollutants in wastewater. The newly proposed methods should be highly efficient, have high economic competitiveness and be environmentally friendly. The primary technology studied in this project is applying the nanostructured cavitation agent (CA) to stimulate cavitation and free radical generation. Antibiotics account for a significant fraction of all kinds of micropollutants. Up to 90% of the antibiotics are still active after human excretion and are released into wastewater discharges, causing a significant environmental burden to water bodies. It has been shown that some antibiotics still affect microbes under a concentration of 10 μg/L [1]. Tetracycline is chosen as the primary object of the current study, as it is the second most used antibiotic [2]. Researchers usually neglect the environmental impact of material synthesis and application. This study aims both to demonstrate approaches to improve the overall removal rate and to compare the life cycle environmental impact of using CA to enhance sonochemical AOPs. Finally we discuss broader environmental and social issues.

Material and Methods

The nano-flower ZnO and hollowed TiO2 were selected as sono-photocatalytic cavitation agents and synthesized using a previously reported method [3,4]. They are light-sensitive and are commonly utilized for photocatalytic degradation [5]. The experimental objectives were to optimize the system and operational parameters to enhance the number of cavitation events, to investigate the effect of using CA to generate free radicals under cavitation, and to evaluate the potential to treat other micropollutants. A bespoke sonoreactor with converging ultrasound and conventional ultrasound tips generates the ultrasound to excite the cavitation [6,7]. The test solution is 40 ppm of tetracycline. The life cycle assessment (LCA) process, as delineated by ISO 14044/40 guidelines, evaluates the environmental performance of sonolytic and sonocatalytic (with CA) removal of tetracycline from Cradle to Grave within the LCA system boundary. All raw material data were sourced from the Ecoinvent 3.9 database via SimaPro v9.5 software, and the ReCiPe 2016 environmental impact assessment method was used [8]. The material consumption and energy utilization estimates during CA synthesis were based on global market and UK scenarios, respectively.

Results and Discussion

The control experiments shown below in Figure 1a confirmed that the tetracycline degradation occurred and was observed within 10 minutes. The degradation by sonolysis and photolysis alone was 5.98% and 5.47%, respectively. By applying the UV and ultrasound irradiation simultaneously, the degradation was very slightly elevated to 6.75%. The degradation rate was significantly enhanced with the addition of 0.5 mg/ml of CA nanoparticles, where the CA was sensitive to both ultrasound and ultraviolet light. Nanoparticles act as an intermediate agent to stimulate the free radical generation and further break down the benzene rings in tetracycline. The

sono-catalytic and photo-catalytic removal of tetracycline increased significantly to 32.05% and 29.42%, respectively. Under the synergism between ultrasound and UV light, the removal rate increased even more to 38.25% in 10 minutes. This demonstrated that the CA makes a considerable contribution to boosting the degradation.

Figure 1b shows the catalytic and non-catalytic degradation after 180 seconds when using the converging ultrasound. The non-catalytic reaction degraded 3.12% of tetracycline. Applying the CA as a catalyst significantly enhanced the degradation rate to 14.08% in 180s. The results show that the system design performed well in degrading antibiotics in a short period of time. In principle, the CA with a specially designed nanostructure provides the opportunity to stimulate cavitation and yield a higher number of free radicals to enhance the reaction rate. The intensive cavitation would also lead to sonoluminescence in which light is emitted. The CA is also sensitive to light, which helps to provide free radicals via a photocatalytic mechanism. Even though a sonocatalyst will yield an advanced oxidation rate, the environmental impact of material acquisition and synthesis is usually not considered by investigators. This final section undertook a comparative analysis of the life cycle environmental impact of utilizing CA in micropollutant degradation. Figure 1c illustrates 18 midpoint indicators of life cycle environmental impacts when treating the same amount of tetracycline. Applying CA exhibited less environmental impact in 14 out of 18 indicators, owing to a reduced electricity consumption (41%). However, the remaining four indicators (FE, HCT, MRS and FSS) showed a higher impact than sonolysis. This disparity can be attributed to greater chemical consumption in the CA synthesis. Exploring alternative synthesis methods, such as substituting titanium with more environmentally friendly materials, may present new opportunities for catalyst preparation to achieve a lower environmental impact.

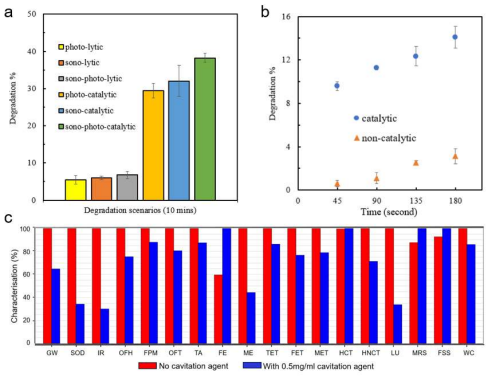

Figure 1. (a) Tetracycline degradation under different scenarios (b) Degradation of tetracycline with and without cavitation agent after 180s of irradiation (c) 18 midpoint environmental impact comparisons with and without cavitation agent

Table 1. Life cycle impact indicators for sonolytic degradation

	Midpoint impact category	Indicators		Midpoint impact category	Indicators
GW	Global warming	kg CO2eq	TET	Terrestrial ecotoxicity	kg 1,4-DCB
SOD	Stratospheric ozone depletion	kg CFC11eq	FET	Freshwater ecotoxicity	kg 1,4-DCB
IR	Ionizing radiation	kBq Co-60 eq	MET	Marine ecotoxicity	kg 1,4-DCB
OFH	Ozone formation, Human health	kg NOx eq	HCT	Human carcinogenic toxicity	kg 1,4-DCB
FPM	Fine particulate matter formation	kg PM2.5 eq	HNCT	Human non-carcinogenic toxicity	kg 1,4-DCB
OFT	Ozone formation, Terrestrial ecosystems	kg NOx eq	LU	Land use	m2a crop eq
TA	Terrestrial acidification	kg SO2 eq	MRS	Mineral resource scarcity	kg Cu eq
FE	Freshwater eutrophication	kg P eq	FSS	Fossil resource scarcity	kg oil eq
ME	Marine eutrophication	kg N eq	WC	Water consumption	m3

Conclusions

Combining converging ultrasound, UV and cavitation agents would significantly improve the oxidation reaction rate of Tetracycline by six times compared to standalone UV or sono-reactions. Even though the use of CA reduced CO_2 emissions by 38%, four environmental indicators showed higher impacts. Both the environmental impact of material acquisition and synthesis and the degradation rate of reaction should be considered when evaluating an AOP.

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