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Microorganisms inactivation in wastewater by solar photo-Fenton at neutral pH

ABSTRACT

The global fresh water shortage, caused mainly by the drought and the pollution of sources, is one of the main environmental problems currently affecting the human race. Over the last few decades, water quality requirements for use in different activities has obliged us to find alternative solutions which requires a concerted effort at a scientific as well as political, economic and social level. In particular, treated wastewater recycling has come up recently as a source provision for sectors where high water quantities are consumed. In this regard, the main sector to benefit is agriculture, which produces the 60% of global food, according to Food and Agriculture Organization (FAO). As such, treatments that are able to guarantee water microbiological quality, as stated by governing law, are necessary and resolve disadvantages or problems with current treatments.

Amongst new technologies available for wastewater regeneration, a noteworthy point is the high level of efficiency in Advanced Oxidation Processes (AOPs). Furthermore, those that are capable of using sunlight as a radiation source are of special interest, particularly the homogeneous photocatalytic process, solar photo-Fenton. Generally speaking, in Fenton reactions, an oxidant agent (H_2O_2) reacts with a catalyst (Fe^{2+}) generating hydroxyl radicals, which are high oxidant and non-selective species causing the inactivation of several microorganisms. The presence of UV-A photons in sunlight leads to catalyst regeneration and the production of more hydroxyl radicals.

One of the main goals of this research work has been to improve the knowledge about the microorganism inactivation process through solar photo-Fenton at neutral pH, which has been scarcely studied, for application as a tertiary treatment in a wastewater treatment plant

(WWTP). With this aim in mind, the bacteria *Enterococcus faecalis* (Gram-positive microorganism) has been used as a fecal pollution indicator since it has not been studied in great detail until now. It is a more able and better indicator than *Escherichia coli* due to its higher resistance to disinfectant treatments. This bacterium has been used as model microorganism in order to study at length the effect of variables such as temperature, radiation intensity and the water matrix composition. Furthermore, the efficiency of solar photo-Fenton process at neutral pH under real conditions has been tested, using microorganisms from wastewater coming from a WWTP instead of a microorganism collection. Also the efficiency of the photo-Fenton treatment in other fecal pollution indicators, such as viruses, which had very little research dedicated to it, has now been investigated.

Studies of the effect of temperature and irradiance on bacteria inactivation through the photo-Fenton process have been carried out, whilst keeping constant other variables affecting the efficiency of the photocatalytic process, with the purpose of removing interference. Experimental results allowed a first order kinetic model to be achieved in each case, for the prediction of bacterial inactivation rate. Moreover, an in-depth study has been carried out to determine whether the irradiance or the accumulated energy is appropriate for the standardization and automation of solar disinfection treatments.

Another of the main parameters affecting the disinfection process by solar photo-Fenton is the water matrix composition. On that count, the interaction between simultaneous processes of microorganism disinfection and organic matter degradation has been studied, firstly evaluating the possible competence for the main oxidant species generated during the photo-Fenton process. Within this study a competitive effect has been tested. This greatly affects the disinfection results and proves that the concentrations of reactive agents are decisive. *E. faecalis* and resorcinol have been used as a pollution indicator and as an organic matter model, respectively. Secondly, based on reactive agent concentrations, at which the best results were attained, the effect of several water matrices on the *E. faecalis* inactivation has been evaluated: i) saline solution (0.9% NaCl), ii) distilled water with resorcinol as organic matter model and iii) WWTP secondary effluent simulated water. These experiments were carried out in 7L CPC photoreactors with the aim of gaining comparable results to those expected at large-scale. These results demonstrated that as the complexity of the organic matter composition increases, the disinfection kinetic improves, reducing the time needed for the treatment to achieve the detection limit. This feature makes the photo-Fenton process an option of interest for the use as tertiary treatment in WWTPs, where the complexity of the water matrix is high. Finally, a study of wild microorganism inactivation from a WWTP ("El Bobar", Almeria) secondary effluent by solar photo-Fenton at neutral pH was carried out in the same photoreactors. Several operation strategies were tested demonstrating that the detection limit was achieved in short treatment times. Furthermore, the efficiency of this process was evaluated under different seasonal pollutant conditions, establishing that treated water follows the microbiological quality parameters for regenerated water according to Spanish law for its subsequent recycling as irrigation water in agriculture.

Finally, a study of the mechanisms involved in virus inactivation by solar photo-Fenton has been also carried out. To this end, implicated processes in coliphage MS2 inactivation by photo-Fenton treatment (UV/Fe, UV/H₂O₂, H₂O₂/Fe and UV/H₂O₂/Fe) have been studied in

great detail. Also the influence of the main variables of the photo-Fenton process, such as the Fe oxidation state (Fe²⁺ or Fe³⁺), pH, Fe and H₂O₂ concentrations and UV radiation intensity, has been evaluated. The photo-Fenton process proved to be suitable in achieving low detection limits for this microorganism. Moreover, its efficiency in water from Lake Lemán (Switzerland) was verified, along with the inactivation of the human virus Echovirus.

The results obtained in this research reveal that the solar photo-Fenton process at neutral pH allows the inactivation of microorganisms from treated wastewater up to a biological load accepted by the law, following the specifications established in water recycling. In addition, short treatment times, combined with i) the use of sunlight as a radiation source, ii) a low reactive agents quantity and iii) the lack of toxic derivatives, makes the photo-Fenton process a suitable alternative for tertiary treatments currently used in urban WWTPs with the aim of regenerating treated water for it to be subsequently used in agriculture.