



Effect of μM Fe addition, mild heat and solar UV on sulfate radical-mediated inactivation of bacteria, viruses, and micropollutant degradation in water

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ABSTRACT

In this work, solar disinfection (SODIS) was enhanced by moderate addition of Fe and sodium peroxydisulfate (PDS), under solar light. A systematic assessment of the activating factors was performed, firstly isolated, then in pairs and concluded in the combined Fe/heat/solar UV-PDS activation process. Solar light was the most effective (single) activator, and its combination with Fe and heat (double activation) yielded high level of synergies (up to $S = 2.13$). The triple activation was able to reduce the bacterial load up to 6-log in less than 1 h, similarly to the photo-Fenton process done in comparison (SODIS alone: >5 h). Fe-oxides were suitable activators of PDS under the same conditions while the presence of organic matter enhanced bacterial inactivation by the triple activated PDS process. The degradation of a (selected) mixture of micropollutants (i.e. drugs, pesticides) was also achieved in similar order of magnitude, and faster than the photo-Fenton process. Finally, the removal of a viral pathogen indicator (MS2 bacteriophage) was attained at minute-range residence times. The aforementioned facts indicate the suitability of the mild, combined process, as a potential SODIS enhancement, producing safe drinking water for sunny and especially for developing countries.

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1. Introduction

Nowadays, more than 10% of world's population does not have an access to improved source of drinking water (McGuigan et al., 2012). This lack of safe drinking water in a large proportion of world's population is responsible for many disorders' development and illnesses in less developed countries (Fewtrell et al., 2005). Point-of-use solutions to this context and potent processes capable to treat drinking water effectively, in order to achieve higher water quality and safety of consumption for the populations are necessary. Considering that these processes will be used especially in developing countries, they have to be easy to use, safe, cheap and sustainable with the environmental situation at the point-of-use

(McGuigan et al., 2012; Fewtrell et al., 2005; Clasen et al., 2007).

Solar disinfection (SODIS) is a process in which the potential of solar light is used to disinfect drinking water. This developing process takes advantage of the ability of solar light to kill pathogens through genome damages and intracellular oxidative reactions (Giannakis, 2018) and can be enhanced by the addition of various chemicals, optimizing of container volume and exposure to solar light (McGuigan et al., 2012; Kehoe et al., 2001). In the SODIS context, processes such as solar-assisted advanced oxidation processes (AOPs) present great potential. The AOPs use the potential of a chemical oxidant species to disinfect water by inactivating potentially pathogenic species such as bacteria, viruses or yeasts (Giannakis et al., 2016, 2016c, 2017a, 2017b) and degrading various harmful organic contaminants to treat drinking water. A widely studied AOP is the photo-Fenton (PF) process in which hydroxyl radicals (HO^\bullet) are generated using light, iron and hydrogen peroxide (H_2O_2) and act as oxidant species in the treatment of water (Giannakis et al., 2016a; b). Photo-Fenton process can be carried out using solar light, enhancing the efficiency of SODIS,

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given that Fe is almost omnipresent in natural waters and H₂O₂ can be added and/or is in situ-generated (Vione et al., 2014).

Sulfate radicals (SO₄^{•−}), as hydroxyl radicals, present a great potential for an alternative advanced oxidation treatment for water (Devi et al., 2016; Xiao et al., 2016). In comparison with photo-Fenton process, where hydrogen peroxide is the initiator of hydroxyl radical species, sulfate radicals can be generated in different pathways from peroxymonosulfate (PMS) and peroxydisulfate (PDS), both acting as initiators (Oh et al., 2016). These initiators have to be activated in order to generate sulfate radicals. Moreover, the sulfate radicals' redox potential is close to hydroxyl radicals' (2.6 V) (Ghanbari and Moradi, 2017; Liang et al., 2008), which makes SO₄^{•−} a good candidate for water treatment. There are many activation factors such as heat (Ji et al., 2015; Tsitonaki et al., 2008), light (Cui et al., 2016; Garkusheva et al., 2017), transition metals (Oh et al., 2016; Anipsitakis and Dionysiou, 2004; Rodríguez-Chueca et al., 2017a) and others, each of them with specific efficiencies. Table 1 summarizes the known activators of PDS. The interest in using sulfate radicals instead of HO[•] arises from characteristics which are favorable to the use of sulfate-based radicals, such as the selectivity to oxidation (Anipsitakis and Dionysiou, 2004; Yu et al., 2016) and also the higher stability compared to H₂O₂, that allows the initiator to diffuse at wider lengths in the subsurface before microorganism activation.

So far, a significant amount of works has been published on contaminant removal using sulfate radicals and treating large variety of compounds, such as pharmaceuticals (Ahmed et al., 2012), alcohols (Oh et al., 2009) and others. The by-products generated depend on the contaminants present, but the other species produced (CO₂, H₂O and SO₄^{2−}) are harmless. This makes the process interesting for application in drinking water treatment. Compared to organic contaminants, the effect of sulfate radicals on microorganisms is much less studied. The effectiveness of transition metal/UV activated processes on *Escherichia coli* inactivation is reported for different water conditions (Garkusheva et al., 2017; Rodríguez-Chueca et al., 2017b; Sun et al., 2016). There is however, an important lack of studies on solar and mild thermal/chemical activation of persulfate, for bacterial disinfection, which, given the nature of the in-situ SODIS treatment, would be greatly beneficial.

In this study, the enhancement of the SODIS process using sulfate radicals in mild conditions is evaluated. Solar light, low amounts of iron and mild heat activation of sodium peroxydisulfate (Na₂S₂O₈) are investigated to build a combined process where these three factors are optimized to disinfect simulated contaminated drinking water (*Escherichia coli*) in the context of point-of-use technology in developing countries. The effect of the most commonly encountered parameters, such as reactant concentrations, organic matter presence, and the use of natural iron oxides as iron source is also studied. The combined process for bacterial disinfection is compared to the well-known photo-Fenton AOP and briefly evaluated on virus and micropollutants.

2. Materials and methods

2.1. Chemicals and reagents

Chemicals and reagents were used as received. A list of the

reagents used within this work are provided in the following table (Table 2). MilliQ water (Millipore Elix Advantage 3, Millipore AG) is used for the preparation of the aqueous solutions (15.8 MΩ cm).

2.2. Experimental setup

In order to assess bacterial inactivation, cylindrical double-wall Pyrex glass reactors (outer diameter 7.5 cm, inner diameter 6.5 cm, height 9 cm, no UV-B transmission) are used. The reactors are provided with a double-wall, serving as a heating jacket which allows control of the reaction temperature, while all reactions are carried under constant stirring (350 rpm). However, with the heating jacket being relatively thick and opaque, light is allowed to penetrate the reacting volume mostly through the top of the reactors, offering an exposed surface of about 30 cm²; the cover was made of borosilicate glass with limited UVB transmittance (see Supplementary Fig. S1). Before every experimental run, the reactors are sterilized by autoclaving (FVG3, Fedegari Autoklaven AG). After each experiment, reactors are washed using acid to ensure iron removal, ethanol to remove any other contaminant and finally rinsed with deionized water in abundant quantities.

During the experiments, simulated solar light exposure is ensured using a bench-scale solar simulator (Suntest CPS, Heraeus), employing a 1500 W air-cooled Xenon lamp with infrared and UV-C cut-off filters. The light intensity applied within the experiments (900 W/m²) is monitored by a pyranometer (CM6b, Kipp & Zonen) and corresponds to a typical daylight intensity in the areas candidate for SODIS.

Temperature control is ensured using water as calorific fluid. A pump guarantees the water flow through reactor's jacket in a closed cycle including a water basin which is kept at the desired temperature thanks to a thermostat (F25 Refrigerated/Heating Circulator, JULABO GmbH).

2.3. Microbial methods

2.3.1. *Escherichia coli* K12

All disinfection experiments are carried out using *Escherichia coli* strain K12 (Deutsche Sammlung von Mikroorganismen und Zellkulturen, DSMZ No. 498) as bacterial species. This strain is non-pathogenic and allows a good approximation of wild-type *E. coli*, which is a well-known indicator bacterium for enteric pathogens. *E. coli* strain storage is assured in cryo-vials containing 20% glycerol at −20 °C.

Bacterial pre-cultures are obtained by spreading 20 μL of the strain onto Plate Count Agar (PCA; Merck) followed by 24 h incubation at 37 °C (Heraeus Instruments). A grown colony is then re-plated and spread on a new PCA plate and incubated for additional 24 h at 37 °C. This 48 h process results in a master plate which is stored at 4 °C and can be used during about 2 weeks.

In order to prepare bacterial stock solution from the master plate, Luria-Bertani (LB) broth is first prepared and then heat-sterilized in an autoclave. A colony from the master plate is then inoculated into a sterile PE Eppendorf flask containing 5 mL of the LB broth prepared. The flask is agitated aerobically for 8 h at 750 rpm and under a temperature of 37 °C. Then 2.5 mL are extracted from the Eppendorf flask and diluted into a sterile 500 mL

Table 1
Indicative rates of sulfate radical-generating systems in the vicinity of our study.

Activation factor	Initiator	Radical species	Rate constant (k) or quantum yield (Q)	Ref
Light	PDS	SO ₄ ^{•−} (2x)	Q = 0.55 mol/Einstein (simulated solar light; 25°C)	Khandarkhaeva et al., 2015; Mark et al., 1990
Heat	PDS	SO ₄ ^{•−} (2x)	k = 1 · 10 ^{−7} s ^{−1} (25°C) up to k = 5.7 · 10 ^{−5} s ^{−1} (70°C)	Liang et al., 2008
Transition metals	PDS	SO ₄ ^{•−}	k = 20–27 M ^{−1} s ^{−1} (Fe ²⁺ activation)	Liang et al., 2008; Bianco et al., 2017

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