



## MS2 bacteriophage inactivation using a N-doped TiO<sub>2</sub>-coated photocatalytic membrane reactor: Influence of water-quality parameters

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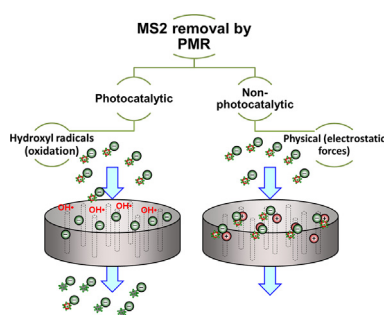
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### HIGHLIGHTS

- Photocatalytic Al<sub>2</sub>O<sub>3</sub> membrane was found efficient for virus removal.
- MS2 removal by PMR was strongly affected by water quality.
- Alkaline pH and NOM were main inhibitors to MS2 removal by PMR.
- Calcium increased MS2 removal through charge-neutralization or cation-bridging.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

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### ABSTRACT

Alternative water-disinfection processes are needed due to the formation of harmful disinfection byproducts during traditional chlorination. Depth filtration using microfiltration membranes has the advantage of applying low transmembrane pressure. However, removal of small nanoparticles such as viruses cannot be based on size-exclusion or sieving due to the large pore sizes. Combining membrane filtration and advanced oxidation processes such as photocatalysis may provide high water quality in a single step by combined filtration and photocatalytic inactivation. This hybrid process, where a photocatalyst immobilized onto a membrane substrate is termed photocatalytic membrane reactors (PMRs). The efficiency of a N-doped TiO<sub>2</sub>-coated Al<sub>2</sub>O<sub>3</sub> PMR for removal of MS2 bacteriophage was investigated with different water qualities. MS2 inactivation was determined by individual addition of cations, anions and natural organic matter. The combined effect of water content on the disinfection potential of the suggested PMR was examined using a natural surface water source. Virus removal by the PMR under irradiation was  $4.9 \pm 0.1 \log$  (> 99.99%) in natural surface water. However, results revealed complex virus–PMR interactions even before exposure to the light source. Removal of MS2 by the PMR in a complex water matrix was driven by electrostatic forces in addition to photocatalytic inactivation. While alkaline pH of water resulted in poor interaction and low removal of MS2 by PMR, Ca<sup>2+</sup> addition lead to increased MS2 removal. High affinity to interaction with Ca<sup>2+</sup> was observed for both MS2 and PMR zeta potentials; moving towards more positive zeta values. Therefore, “salt bridge” effect is the most likely route to reduced virus–membrane electrostatic repulsion.

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## 1. Introduction

Concerns about the formation of potentially mutagenic and carcinogenic disinfection byproducts during water chlorination [1] have led to the development of alternative disinfection processes involving, for example, ozone, chlorine dioxide, ultraviolet (UV) and membrane filtration. Many of these processes require expensive chemicals, costly equipment or high energy consumption. Microfiltration (MF) membrane systems (with pores in the 0.1–10  $\mu\text{m}$  range) offer quick and selective separation of suspended particles and larger pathogenic microorganisms while operating at low pressures. However, contaminants, including micropollutants, small bacteria and viruses, are poorly removed from water by MF alone. One way to overcome the low removal of viruses and micropollutants via MF is to design a hybrid process with MF and advanced oxidation processes (AOPs).

In photocatalysis, classified as a heterogeneous AOP, a semiconductor (catalyst) is activated with sunlight irradiation. Photocatalysis could potentially enable effective virus inactivation in drinking water while limiting the formation of disinfection byproducts [2].  $\text{TiO}_2$  is the most common photocatalyst applied to remove xenobiotics (e.g. pharmaceuticals) from water [3] and to inactivate microorganisms such as bacteria [4] and viruses [4–6]. A hybrid photocatalytic membrane reactor (PMR) combining both membrane filtration and photocatalysis can address multiple functions beyond physical separation, such as degradation of organic pollutants [7,8], inactivation of microorganisms [9,10] and self-antibiofouling action [11]. PMRs can be operated in two configurations [12]: (a) photocatalyst in a slurry, limited by catalyst leaching and settling, requires separation/recycling of the catalyst from the treated water, or (b) photocatalyst immobilized on a porous membrane and activated by light. A PMR developed in our previous work using  $\text{Al}_2\text{O}_3$  MF membranes as a substrate for sol-gel-coated N-doped  $\text{TiO}_2$  photocatalyst was superior by 90% with respect to oxidation of the persistent pollutant carbamazepine, with flow through the membrane pores relative to flow on top of the membrane due to enhanced mass transfer and increased surface area. Doping  $\text{TiO}_2$  with nitrogen resulted in a coating with anatase structure and N content of 0.3 at.%, predominantly located at interstitial sites in the  $\text{TiO}_2$ . The modification of  $\text{TiO}_2$  facilitated extended photocatalytic activity to UVA and visible light, allowing more efficient utilization of solar light [8,13,14]. The aforesaid developed PMR will be further examined in this work to determine its disinfection efficiency.

Bacteriophages are a common viral indicator for water-disinfection efficacy. Although bacteriophages infect bacteria, they can serve as models or surrogates for human viruses in assessments of water quality [15]. The MS2 virus, which is an F-RNA coliphage, is similar to polio and hepatitis viruses in size ( $\sim 25$  nm [16]), shape (icosahedron) and all have a single-stranded RNA genome. The MS2 phage possesses unique physicochemical properties, which include its small size, negative charge under a wide pH range (isoelectric point [IEP]  $\sim 3.9$  [17]) and relative hydrophobicity [18]. All of this makes MS2 and similar viruses the worst-case scenario for virus removal by traditional membrane filtration [19]. MS2 is resistant to UV treatment, with no significant disinfection at wavelengths greater than 295 nm [20]; however, it is highly sensitive to hydroxyl radical oxidation [5,20]. Inactivation of MS2 bacteriophages is induced by denaturing the protein of the capsid, whose structure is simple and rigid, and thus requires high oxidation power [21].

Guo et al. [9] were the first to report photocatalytic disinfection of viruses by an immobilized PMR configuration. Inactivation of P22 bacteriophage in ultrapure water was found to be considerably more effective using PMR (a photocatalytic UV–MF hybrid) than either UV disinfection, MF or a non-photocatalytic UV–MF hybrid process. However, photocatalytic efficiency is expected to be lower for natural vs.

deionized water. The chemical composition of different water bodies—inorganic and organic substances—will have a substantial impact on photocatalytic process efficiency due to these substances' potential photoactive and radical-scavenging properties [22,23]. Removal of microorganisms by conventional membrane filtration is predominantly physical; however, the physicochemical properties of the virus and the membrane, and the origin of the interaction between them, may be more significant for MF membranes where small organisms like viruses are concerned [19,24]. Physical adsorption plays an important role in the removal of viruses by MF membranes (non-photocatalytic and photocatalytic) and is also strongly affected by water chemistry (e.g. organic matter, ionic composition) [24]. The chemical composition of different water types can vary significantly and therefore, it is necessary to determine the influence of the dominant species on both the non-photocatalytic virus removal and the photocatalytic inactivation efficiency of the PMR proposed herein. To date, there have been no reports on viral disinfection efficiency using a PMR in a complex water matrix, such as natural water. The goal of this study was to examine the impact of water-quality parameters on the inactivation of MS2 virus using PMR based on previously developed N-doped  $\text{TiO}_2$ -coated or non-coated  $\text{Al}_2\text{O}_3$  MF membranes with or without activation by solar UV–visible light. The following water-quality parameters were examined: pH and alkalinity, chloride, calcium, magnesium and natural organic matter (NOM) contents, in buffered water and natural surface water. Non-photocatalytic (without irradiation) and photocatalytic (with irradiation) MS2 removal (see Appendix Fig. A.1), driven by virus–membrane interactions under different water qualities, was linked to the physicochemical properties of the virus and the coated and non-coated membrane using MS2 nanoparticles and transmembrane zeta potential analysis.

## 2. Materials and methods

### 2.1. Materials and reagents

Sodium phosphate monobasic and sodium phosphate dibasic (Sigma Aldrich, Germany) were used together to make pH 6 and pH 7 buffers. Boric acid and sodium tetraborate (Sigma Aldrich, Germany) were used together to make a pH 8.5 buffer. Alkalinity of the water was modified using  $\text{NaHCO}_3$  (Sigma Aldrich, Germany). Chloride was added by dissolving NaCl (Sigma-Aldrich, Germany). Calcium ( $\text{Ca}^{2+}$ ) and magnesium ( $\text{Mg}^{2+}$ ) were added by dissolving  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  and  $\text{MgSO}_4$  (Sigma-Aldrich, Germany), respectively. Suwannee River natural organic matter (SRNOM) (International Humic Substances Society – IHSS, St. Paul, MN) was used to simulate NOM. A stock solution of SRNOM ( $500 \text{ mg L}^{-1}$ ) was prepared following Luster et al. [14]. All chemicals were used as obtained. Stock solutions were prepared by dissolving each compound in DI water (Direct-Q3 UV System, Millipore – France).

### 2.2. Preparation of N-doped $\text{TiO}_2$ thin film coatings on membranes

Commercial flat  $\alpha\text{-Al}_2\text{O}_3$  MF membranes with a nominal pore size of 800 nm (Nanostone Water, Germany, formerly KSM Water GmbH) were selected as the substrates for the sol-gel N-doped  $\text{TiO}_2$  coating. The membrane sheets were cut to give an effective filtration area of  $2.2 \times 4.2 \text{ cm}^2$ , with a thickness of 6.6 mm. The N-doped  $\text{TiO}_2$  coatings were prepared by sol-gel deposition using tetrabutylorthotitanate (25 mL), isopropanol (32 mL), ammonium hydroxide (32%, 2.4 mL) and triethanolamine (15 mL) as precursors. The coatings were applied by a drop-coating method using a pipette (2.5 mL corresponding to  $0.12 \text{ mL cm}^{-2}$  of membrane area). A 1 mL of solution was deposited at the sample center using a graduated Eppendorf pipette. The solution quickly spread out on the surface forming a homogeneous film. In a few

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