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Water Research 39 (2005) 3098–3108

WATER RESEARCH

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Comparison of electrocoagulation and chemical coagulation pretreatment for enhanced virus removal using microfiltration membranes

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Received 20 December 2004; received in revised form 17 May 2005; accepted 18 May 2005

Abstract

This research studied virus removal by iron electrocoagulation (EC) followed by microfiltration (MF) in water treatment using the MS2 bacteriophage as a tracer virus. In the absence of EC, MF alone achieved less than a 0.5-log removal of MS2 virus, but, as the iron-coagulant dosage increased, the log virus removal increased dramatically. More than 4-log virus removal, as required by the Surface Water Treatment Rule, was achieved with 6–9 mg/L Fe^{3+}. The experimental data indicated that at lower iron dosages and pH ($<\sim$8 mg Fe/L and pH 6.3 and 7.3) negatively charged MS2 viruses first adsorbed onto the positively charged iron hydroxide floc particles before being removed by MF. At higher iron dosages and pH ($>\sim$9 mg Fe/L and pH 8.3), virus removal was attributed predominantly to enmeshment and subsequent removal by MF. Additionally, the experimental data showed no obvious influence of ionic strength in the natural water range of 10^{-7}–10^{-2} M on MS2 virus removal by EC-MF. Finally, EC pretreatment significantly outperformed chemical coagulation pretreatment for virus removal. The proposed mechanism for this improved performance by EC is that locally higher iron and virus concentrations and locally lower pH near the anode improved MS2 enmeshment by iron flocs as well as adsorption of MS2 viruses onto the iron floc particles.

Keywords: Microfiltration; Coagulation; Electrocoagulation; Virus; Bacteriophage; Water treatment; Membrane filtration

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1. Introduction

Microfiltration (MF) has been used to completely or significantly remove turbidity, bacteria, and protozoa from water and wastewater (Jacangelo et al., 1991; Mallevialle et al., 1996; Madaeni, 1999). However, MF alone is not an efficient barrier for virus removal, because viruses are typically smaller than its pores. Several published studies have reported partial (0.2- to 3-log) virus removal by MF (Coffey et al., 1993; Madaeni et al., 1995; Urase et al., 1996; Roberts, 1997), which is significantly less than the 4-log virus removal mandated by the Surface Water Treatment Rule (SWTR). Therefore, MF by itself cannot meet the SWTR virus removal requirement even though the turbidity and protozoa regulations can be met easily.

The addition of a coagulant prior to membrane filtration has been suggested to generally improve product water quality and reduce membrane fouling by coagulating the dissolved organic matter ahead of the

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0043-1354/$ - see front matter
doi:10.1016/j.watres.2005.05.020

filter (Lahoussine-Turcaud et al., 1990; Wiesner et al., 1992). Usually, chemical coagulation (CC) with iron or aluminum salt is used, however electrocoagulation (EC) is another possibility. To our knowledge, this is the first report of EC pretreatment for MF.

EC has been widely studied in water and wastewater treatment to remove heavy metals, organics, bacteria, hardness, turbidity, and other contaminants (Horner and Duffey, 1983; Tsouris et al., 2001; Can et al., 2003; Mills, 2000). In the EC process, the electrodes are consumed as the coagulant is generated and precipitated; no liquid chemical is added; alkalinity is not consumed; and pH adjustment is not needed. Additionally, compared with CC, the EC process reportedly requires less coagulant and produces less sludge (Horner and Duffey, 1983; Mills, 2000). According to one estimate, the space required for EC is less than CC because EC does not require chemical storage, dilution, and rapid mixing (Mills, 2000). Because EC systems typically use solid iron or aluminum anodes rather than corrosive iron or aluminum salt solutions, EC units can be more easily incorporated into "packaged" plants and transportable water treatment plants for use in remote areas or in emergency water supply treatment, which was one of the driving forces to undertake this research.

The objectives of this study were to (a) evaluate the efficacy of iron EC-MF for virus removal, (b) investigate the effect of EC-MF operating parameters on virus removal, (c) compare virus removal by EC-MF and CC-MF, and (d) suggest possible mechanisms for virus removal by EC-MF.

2. Materials and methods

2.1. Virus

The indicator-organism virus used throughout the study was the bacterial virus, MS2. It is 0.025 μm in size, icosahedral in shape, and contains a single strand of ribonucleic acid with 3569 nucleotides (Valegård et al., 1990). The virus stock was bought from American Type Culture Collection (ATCC # 15597-B1). Because bacterial viruses are also called bacteriophages (Brock and Madigan, 1991), we use the terms "virus" and "bacteriophage" interchangeably to refer to MS2.

2.2. MS2 virus propagation and MS2 virus assay

The procedure of MS2 virus propagation and MS2 virus assay was described in our previous paper (Zhu et al., 2004). MS2 samples were assayed using a modified agar-overlay technique, which counts viable viruses using *Escherichia coli* as the host bacterium.

2.3. Test water

Three synthetic fresh waters, roughly characterized as low ($I = 10^{-7}$ M), medium ($I = 6 \times 10^{-3}$ M), and high ionic strength ($I = 1.8 \times 10^{-2}$ M), were used in this research. These were prepared from deionized (DI) water and reagent-grade chemicals to simulate fresh water containing monovalent and divalent cations, and alkalinity. The low-ionic-strength water was DI water, and the high-ionic-strength water was DI water spiked with 3.0 mM $NaHCO_3$, and 10 mM $CaCl_2$. The medium-ionic-strength water used in most of the experiments consisted of DI water spiked with 3.0 mM $NaHCO_3$, and 1.0 mM $CaCl_2$. Its pH was 8.3 without any adjustments.

2.4. Membrane microfilter

All filtration experiments were carried out using a hydrophilic, 0.22-μm pore size, modified polyvinylidene fluoride (PVDF) membrane filter (Durapore, Millipore Corp. Bedford, MA). A fresh membrane with an effective filtration area of 4.1 cm^2 was used for each experiment. The manufacturer confirmed that its surface was negatively charged in the pH range of our experiments, 6.3–8.3.

2.5. EC unit

A bench-scale EC unit with three anode–cathode pairs was designed and built for this research (Fig. 1). It consisted of a 200-mL active-volume, flow-through electrode chamber with 22-cm long rod-shaped iron anodes and porous cylindrical stainless steel cathodes. The total anode surface area was 100 cm^2 and the current density was typically 0.25 mA/cm^2. By adjusting the operating current and flow rate of source water, the desired iron concentration was obtained.

As can be seen in Fig. 1, the recycle pump discharged into the annular area between the cathode and anode of each of the three cathode-anode pairs in order to flush the anode where iron coagulant was continuously

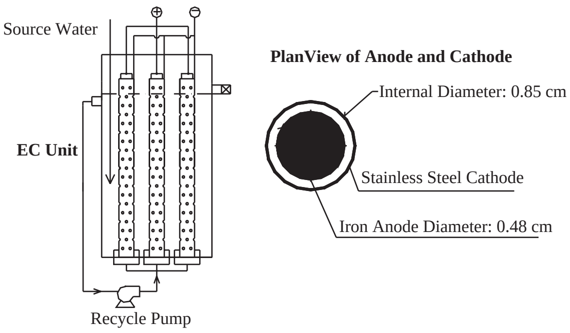

Fig. 1. Diagram of batch bench-scale EC system.