



Virus removal by iron oxide ceramic membranes



Maria M. Fidalgo de Cortalezzi^{a,d,*}, Maria V. Gallardo^a, Fernando Yrazu^a,
Guillermina J. Gentile^a, Oscar Opezzo^{a,b}, Ramon Pizarro^{a,b}, Hugo R. Poma^{c,d},
Verónica B. Rajal^{c,d}

^a Department of Chemical Engineering, Instituto Tecnológico de Buenos Aires, Avenue Madero 399, 1106 Buenos Aires, Argentina

^b Comisión Nacional de Energía Atómica, Unidad de Actividad Radiobiología, Avenue General Paz 1499, General San Martín, 1650 Buenos Aires, Argentina

^c INIQUI, Universidad Nacional de Salta, Avenue Bolivia 5150, 4400 Salta, Argentina

^d CONICET, Argentina

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ABSTRACT

Nanoporous iron oxide ceramics were studied for the removal of virus contamination from water. Supported and unsupported iron oxide nanostructured hematite was fabricated by a green chemistry route from ferroxane nanoparticles. The material had a surface area of approximately 30 m²/g and a mean pore size of 65 nm. Bacteriophage P22 was chosen as a model for human virus. The kinetics and equilibrium of the attachment process was investigated. P22 adsorption isotherms on iron oxide were described by the Freundlich equation. Batch experiments resulted in 1.5 LRVs. Removal proceeded rapidly for the first 7 h; next, a diffusion-limited stage occurred. Dynamic attachment experiments demanded extensive recirculation to achieve significant reduction levels. Up to 3 LRV were observed. The enhanced performance can be explained by the higher iron oxide area available and the facilitated access to inner porosity sites that were previously unavailable due to slow diffusion. The role of electrostatic interactions in the attachment mechanisms was confirmed by the dependence of the isotherm on the ionic strength of the suspension medium. P22 bacteriophage is expected to attach to the iron oxide by electrostatic forces up to a pH of 6.5. DLVO theory predicts moderately well the interaction energies between P22 particles themselves and between the phage and the ceramic. However, a slight underestimation of the P22–P22 repulsive forces was evident by comparison to the experimental data.

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Introduction

The availability of a safe water supply is an increasingly important priority for the welfare and development of human populations, challenged by the combined growth of demography, development, and waste. Parallel to this, researchers and authorities have been paying more attention in the last decades to the link between disease outbreaks and the presence of viral pathogens in drinking water sources [1]. The removal or inactivation of virus in water treatment is a challenging task: the technical difficulty resides in the fact that viruses and bacterial spores are much harder to eliminate -by common techniques such as microfiltration or chlorination for example- than bacterial

pathogens due to their smaller size and simpler physiology. Although bacteria and larger microorganisms can be removed by ultrafiltration or microfiltration, the removal of smaller viral particles is mainly controlled by electrostatic interactions and attachment [2,3]. Nanofiltration membranes meet the requirements for nanosize particle removal, but its use represents a significant increase in cost due to higher pressure requirements and lower produced water yields [4]. Consequently, there is a need to develop cost-efficient methods to achieve these goals, not only for large urban agglomerations but also for smaller rural populations and mobile applications.

Adsorption processes can effectively remove small contaminants as well as ionic constituents from water, avoiding energy consumption associated with pumping and disposal issues of concentrated streams generated [5,6]. Furthermore, bulk porous adsorbents, as opposed to suspended colloidal particles and co-precipitation schemes, offer the additional advantage of not requiring an extra separation stage added to the treatment process, and thus, rendering the system safer to handle, easier to use and applicable to mobile devices. In order for an adsorbent to be an

* Corresponding author. Current address: E2509 Laferre Hall, Department of Civil and Environmental Engineering, University of Missouri-Columbia, Columbia, MO 65211-2200, USA. Tel.: +1 573 884 6777.

E-mail addresses: fidalgom@missouri.edu, maria.fidalgo1@gmail.com (M.M. Fidalgo de Cortalezzi).

Nomenclature

a	radius of virus like particle (m)
A_{132}	combined Hamaker constant for bacteriophage P22 and hematite in water as medium (J)
A_{131}	Hamaker constant for bacteriophage P22 in water as medium (J)
A_{11}	Hamaker constant of virus like particle (J)
A_{22}	Hamaker constant of hematite (J)
A_{33}	Hamaker constant of water (J)
C	ion concentration (mol dm ⁻³)
DLVO	Derjaguin
Landau	Verwey and Overbeek
e	electron charge
h	separation between surfaces (m)
k	Boltzmann constant
n_{∞}	bulk number of ions (ions m ⁻³)
T	temperature (K)
U	electrophoretic mobility (m ² V ⁻¹ s ⁻¹)
V_{EDL}	electrical double layer interaction potential energy (J)
V_{TOTAL}	total colloidal interaction potential energy (J)
V_{vdW}	unretarded van der Waals interaction potential energy (J)
z	valence of symmetrical (z-z) electrolyte
z_i	valence of ion i including sign of charge
γ	reduced surface potential
ϵ	dielectric constant (F m ⁻¹)
ϵ	permittivity
ζ	zeta potential (V)
κ	Debye–Huckel reciprocal length (m ⁻¹)
μ	viscosity (Pa s)
φ	electrical surface potential (V)

efficient and economically sound option while still maintaining the above mentioned characteristics, it must be readily available on site, have a relatively high specific surface area, low cost, and no toxicity associated with its fabrication, use, or final disposal of the material itself or its degradation products.

Previous work successfully led to the developing of iron oxide nanostructured ceramic membranes [7,8], which have proven to be an efficient treatment for the removal of natural arsenic contamination [9] and promising for other ionic contaminants as well. These membranes are specially suited for their use as filters of both inorganic/organic and biological contaminants in rural settings where access to electricity or specialized supplies is compromised. There is evidence [10–12] that iron and iron oxides can effectively retain and/or inactivate viral particles. Furthermore, iron oxides or minerals having a substantial proportion of iron oxides have shown very high affinity for virus that seems to be independent of their type or structure [10,13], and therefore can become a general method for the removal of virus from natural waters, where microorganisms exhibit seasonal and special variability [14,15]. Metal oxide coatings on sand [16–19], glass fiber [20,21], ceramic filters [22,23], and diatomaceous earth [24] have been used in the removal and inactivation of virus from water.

Bacteriophages are recognized as model organisms for human virus [25,26]. P22 [27] and MS2 [13] phages have been used in attachment experiments as well as other phages [28]. P22 phage has an icosahedral capsid and an approximate diameter of 70 nm, which is within the same order of magnitude as picornaviruses, the wide family to which human enteroviruses belong (approximately 30 nm) [29–31].

The nanostructured iron oxide ceramic filter investigated in this work presents numerous advantages over previously studied systems: ease of operation, low fabrication cost, more compact due to the higher specific active area available, and no sludge generation. The attachment kinetics and equilibrium isotherm of bacteriophage P22 was investigated. The virus was characterized with respect to size and surface charge, as they are expected to play a key role in the removal process. A flow through ceramic filter was fabricated and tested for the continuous treatment of contaminated water. The mechanism and limitations of the processes were analyzed and discussed in light of the DLVO theory of colloidal interaction, as well as the aggregation conditions of the phage in natural waters.

Theory

DLVO theory, developed independently by Derjaguin and Landau (1941) and by Verwey and Overbeek (1948), explains colloid stability as well as attachment between colloids and surfaces, based on the van der Waals attraction and the electrical double layer repulsion.

When there is a particle in aqueous suspension that presents surface charge, as for example due to ionization of surface chemical groups, a layer of counterions will develop to balance the charge in the solution adjacent to the surface and an electrical double layer surrounding the particle will appear. As two particles approach, the double layers overlap, giving origin to interaction forces.

The electrical double layer interaction potential energy between two spheres can be calculated by the following expression [32]:

$$V_{EDL} = \frac{128\pi a_1 a_2 n_{\infty} kT}{(a_1 + a_2)\kappa^2} \gamma_1 \gamma_2 \exp(-\kappa h) \quad (1)$$

$$\gamma = \tanh \frac{ze\varphi}{4kT} \quad (2)$$

where V_{EDL} : electrical double layer interaction potential energy (J), a : radius of virus like particle (m), n_{∞} : bulk number of ions (ions m⁻³), k : Boltzmann constant (1.38 × 10⁻²³ J/K), T : temperature (K), κ : Debye–Huckel reciprocal length (m⁻¹), γ : reduced surface potential, z : valence of symmetrical (z-z) electrolyte, e : electron charge (−1.602 × 10⁻¹⁹ C), φ : electrical surface potential (V), h : separation between surfaces (m).

This equation is valid when $h \ll a$ and $\kappa h \gg 5$; in a symmetric electrolyte solution.

In aqueous solutions at 25 °C, κ can be calculated as [33]

$$\kappa = 2.32 \times 10^9 \sqrt{\sum C_i z_i^2} \quad (3)$$

where C_i : ion concentration (mol dm⁻³).

If there is a large difference between particle sizes, the bigger one is perceived as an infinite plate, and Eq. (1) reduces to:

$$V_{EDL} = \frac{128\pi a n_{\infty} kT}{\kappa^2} \gamma_1 \gamma_2 \exp(-\kappa h) \quad (4)$$

The electrical surface potential (φ) is commonly approximated by the zeta potential (potential at the shear plane) due to the impossibility to experimentally determine the first.

Attractive van der Waals interactions are the result of short-term magnetic forces that form between identical or different particles that may have the same, different or no net charge, due to transition dipoles, and depend on the geometry, properties of the particles and the medium in which they interact.

For two identical interacting spheres, the potential can be calculated according to [34]:

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