



Ranking nano-enabled hybrid media for simultaneous removal of contaminants with different chemistries: Pseudo-equilibrium sorption tests versus column tests

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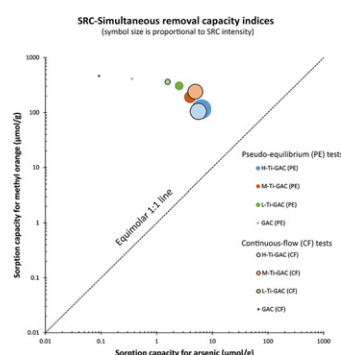
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HIGHLIGHTS

- Pseudo-equilibrium and continuous flow column testing provide the same sorbent ranking trends.
- Simultaneous Removal Capacity (SRC) indices are statistically the same for each tested media.
- GAC with the highest TiO₂ content exhibited highest SRC for simultaneous contaminant removal.

GRAPHICAL ABSTRACT



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ABSTRACT

The underlying hypothesis of this study was that pseudo-equilibrium and column testing conditions would provide the same sorbent ranking trends although the values of sorbents' performance descriptors (e.g. sorption capacity) may vary because of different kinetics and competition effects induced by the two testing approaches. To address this hypothesis, nano-enabled hybrid media were fabricated and its removal performances were assessed for two model contaminants under multi-point batch pseudo-equilibrium and continuous-flow conditions. Calculation of simultaneous removal capacity indices (SRC) demonstrated that the more resource demanding continuous-flow tests are able to generate the same performance rankings as the ones obtained by conducting the simpler pseudo-equilibrium tests. Furthermore, continuous overlap between the 98% confidence boundaries for each SRC index trend, not only validated the hypothesis that both testing conditions provide the same ranking trends, but also pointed that SRC indices are statistically the same for each media, regardless of employed method. In scenarios where rapid screening of new media is required to obtain the best performing synthesis formulation, use of pseudo-equilibrium tests proved to be reliable. Considering that kinetics induced effects on sorption capacity must not be neglected, more resource demanding column test could be conducted only with the top performing media that exhibit the highest sorption capacity.

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1. Introduction

In absence of existing centralized water treatment and distribution infrastructure, conventional water treatment technologies are frequently rendered a less desirable option for providing potable water in small and rural communities (Brame et al., 2011; Cotruvo et al., 2014; Silverstein, 2005). Because of the high techno-economic costs associated with centralized infrastructure construction and operation, many of these communities have shifted their focus towards implementing smaller point-of-use or point-of-entry water treatment systems as a solution for their potable water needs (Berg, 2015; Cotruvo and Cotruvo, 2003; Hamouda et al., 2010; Jones and Joy, 2006). Together with promulgation of more stringent water quality regulations, this has spurred intensive research efforts to develop inexpensive and better performing, small-scale water treatment technologies (Westerhoff et al., 2016). Led by the advent of nanotechnology, much of the research efforts focused on employing the unique properties of nanomaterials to modify existing best available technologies (BAT) and create improved hybrids that are capable of simultaneously removing multiple contaminants with different chemistries. Being the BAT for removing organic contaminants from water, granular activated carbon (GAC) and activated carbon fibers have been a frequent subject of many studies that have explored its modification by introducing metal (hydr)oxide nanoparticles into its pores to create hybrid sorbents capable of also removing inorganic contaminants, like arsenic for example (Chen et al., 2007; Hristovski et al., 2009b; Jang et al., 2008; Sandoval et al., 2011).

Development of a well-performing nano-enabled hybrid sorbent, however, represents a tedious and long task that typically follows a sequence of fabrication, characterization, and testing processes under different conditions to generate a ranking series of the best performing media and optimize the process (Cooper et al., 2010; Hristovski et al., 2009a). Before the best performing hybrid sorbent could be selected for pilot testing and consequent commercialization, a number of different formulations need to be fabricated in sufficient quantities to conduct adequate performance tests, ranging from batch to continuous-flow conditions (Chwirka et al., 2000; Garcia et al., 2017; Hristovski et al., 2008a; Westerhoff et al., 2006). This frequently represents a cost and time-prohibitive process, which necessitates optimization to obtain all the relevant information and make the right decisions with minimum invested resources. The quality and quantity of obtained information from the specific tests directly correlates to the level of required resources to conduct it. For example, single-point pseudo-equilibrium sorption batch tests are suitable for delineating whether the fabricated hybrid sorbent (or ion-exchange resin) exhibits any affinity for removing a targeted set of contaminants. Typically, these types of tests represent a first level of screening, but they do not provide any substantial data beyond simple “litmus test” to quickly evaluate whether a sorbent removes (or not) the contaminants of interest (Hristovski et al., 2007). A more resource demanding multi-point batch sorption tests have the ability to provide more than a single-point sorption tests. When properly conducted and coupled with adequate isotherm models, valuable information could be obtained. Specifically, the results of these tests could describe (1) contaminants' sorption capacity under pseudo-equilibrium conditions, (2) affinity of the sorbent (ion-exchange) for removing the contaminant under different sorbent/sorbate ratios and thermodynamic conditions, and (3) performance under high and low initial contaminant concentrations (Crittenden et al., 2005; Hristovski and Markovski, 2017). Although these batch tests provide less information than the column tests, the generated data is sufficient to conduct rapid screening and ranking of the sorbent's capacity to remove the targeted contaminants. When properly conducted, continuous-flow column tests have the ability to provide much better description of the sorption performance, including information related to contaminant kinetics, potential full-scale operational problems, and sorption (or ion-exchange) capacity, including kinetics induced competition (Garcia et al., 2017; Hristovski and Markovski, 2017). Unfortunately, continuous-flow

column tests are more resource demanding, which frequently is cost and time prohibitive when the performance of sorbents fabricated via different synthesis formulations needs to be screened (Crittenden et al., 1991; Hristovski et al., 2007). In brief, column tests typically require longer testing times and greater quantities of different sorbent materials.

From the practical perspective, the higher data resolution (i.e. more information) provided by continuous-flow column tests is not required when simply screening the sorption performance of a set of media fabricated via different formulation. The end goal of screening is to determine the media performance ranking trends and select the top sorbents for further evaluation. However, a knowledge gap exists that limits the extrapolation of the sorbent rankings obtained under pseudo-equilibrium conditions to rankings obtained under continuous-flow conditions. The underlying hypothesis, which stems from this research gap, is that both testing conditions would provide the same ranking trends although the values of the sorbents' performance descriptors (e.g. sorption capacity) may vary because of different kinetics and competition effects induced by the two testing approaches.

To validate this hypothesis, four task-oriented objectives were achieved: (1) nano-enabled hybrid media were fabricated by impregnation of GAC with three different quantities of TiO₂ nanoparticles; (2) removal performances of the pristine GAC and hybrid Ti-GAC media for two model contaminants were assessed under multi-point batch pseudo-equilibrium and continuous-flow conditions and sorption capacities were estimated under these conditions; (3) simultaneous removal capacity (SRC) indices of all media were estimated; and (4) ranking media's SRC indices for removal of the two model contaminants were performed. Considering the ability of TiO₂ nano-enabled GAC media to simultaneously remove organics and inorganics (Guan et al., 2012; Yao et al., 2012), two sorbates with different chemistries, arsenic (As) and methyl orange (MO), were used as model contaminants.

2. Experimental approach

2.1. Fabrication and characterization of sorbent media

A commercially available macroporous lignite-based GAC (Norit HydroDarco 3000 - HD 3000) was impregnated with three different quantities of TiO₂ nanoparticles following the synthesis and characterization methods described by Garcia et al. (2017) and Elton et al. (2013). Table 1 summarizes designation, the content of TiO₂ and particle porosity of tested media.

2.2. Determining single sorption capacities of media for model contaminants under multi-point pseudo-equilibrium conditions

To assess pseudo-equilibrium sorption capacity for pristine GAC and hybrid Ti-GAC media, multi-point batch sorption tests were conducted with arsenic and methyl orange in realistic conditions. The data was fitted with the Freundlich isotherm model to characterize and quantify the sorption capacity.

The removal of arsenic was evaluated in NSF 53 model challenge water matrix that simulates realistic groundwater conditions that might

Table 1
Designation, content of titanium dioxide and particle porosity of tested media (Garcia et al., 2017).

Designation	TiO ₂ content (%)	Particle porosity (%)
GAC	0.00	0.80
L-Ti-GAC	7.94 ± 1.5	0.80
M-Ti-GAC	20.22 ± 0.2	0.78
H-Ti-GAC	31.46 ± 2.5	0.75

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