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Triclosan as model system for the adsorption on recycled adsorbent materials

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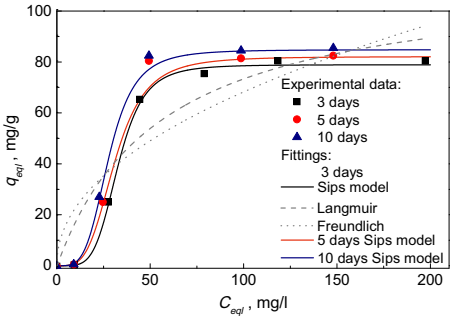
HIGHLIGHTS

- Triclosan adsorption is studied to qualify activated carbon sorbents.
- Experimental isotherms are fitted by Langmuir, Freundlich and Sips isotherms models.
- Contact time is a crucial sorption parameter.
- Maximum efficiency of GP5 (88%) is obtained after 10 days of adsorption.
- Non-linear Sips isotherm provides suitable fitting results.

GRAPHICAL ABSTRACT



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ABSTRACT

The adsorption of triclosan as model system was studied to qualify activated carbon sorbents recycled from gas masks (civilian gas mask GP5). The triclosan equilibrium concentration was measured spectrophotometrically, the morphology of the activated carbon characterized by scanning electron microscopy, and the amount of the adsorbed triclosan on the activated carbon quantified by a mass balance method. Experimental isotherms were fitted by Langmuir, Freundlich and Sips adsorption models. It was obtained that the contact time is a crucial sorption parameter that provides information on the optimum adsorption efficiency. It was shown that the maximum efficiency of GP5 (88%) is obtained after 10 days of adsorption at a maximal concentration of triclosan and carbon loading 1 mg/l. No significant adsorption efficiency differences were measured after 5 and 10 days of adsorption. The non-linear Sips isotherm, a combined Freundlich–Langmuir model, provides suitable fitting results. The observed remarkable adsorption capacity of activated carbon (GP5) towards triclosan adsorption (~85 mg/g) makes it a viable solution for wastewater treatment.

1. Introduction

Triclosan (5-chloro-2-(2,4-dichlorophenoxy)-phenol) is a common synthetic antimicrobial agent that has been incorporated into more than 700 different industrial and personal care products. Today, as antimicrobial active component triclosan can be found

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Table 1
Properties of activated carbon GP5, where A—adsorption capacity towards iodine, M_c—moisture content, V_{total}—the total pore volume, A_c—ash content D_b—bulk density.

A	M_c	V_{total}	A_c	D_b
%	%	cm^3/g	%	g/cm^3
44	14.05	0.62	19.51	0.598

in consumer care products such as toothpaste, mouthwash and soaps, as well as in household cleaners and even in textiles, such as sportswear, bed clothes, shoes, and carpets. The content was found to be in the range of 0.1–0.3% [1–3]. The widespread use of triclosan provides a number of pathways to enter the environment. Laboratory tests have shown that it is toxic to aquatic species. The chemical properties of triclosan indicate that it may also bio-accumulate and persist in the environment [4–6].

Moreover, upon exposure to sunlight or UV light, environmental triclosan can be transformed into more toxic compounds such as chlorinated dioxins [7]. In addition, reports suggest that the incineration of textile products containing triclosan may result in the formation of dioxin-like substances. The primary disposal route for many of these ingredients is the municipal wastewater collection and treatment system and finally the aquatic environment. Due to the uncertainty in estimating the environmental release via this pathway, no predicted environmental concentration (PEC) in surface waters has been derived.

Biodegradation of triclosan in the environment and wastewater has recently become an important research topic [1–23]. In a recent study it was reported that approximately 79% of triclosan can be removed via a biological wastewater treatment processes [3], suggesting that (i) biodegradation can be an important removal mechanism in wastewater, and (ii) triclosan-degrading bacteria are present in the activated sludge. Removal of about 90% was measured in wastewater treatment plants (WWTP) employing conventional activated sludge processes of which 40–60% were due to biodegradation while the remainder was caused by sorption to the sludge [3,4,8,9]. Hence, most removal occurs due to biodegradation processes [8–10].

Activated carbons are traditional and effective adsorbents for the treatment of industrial wastewater [11]. The advantages of coal based carbons can be seen in their ability to remove toxic organic compounds from industrial and municipal wastewater and potable water as well [12,13]. That is why it is crucial to study the adsorption of triclosan as a model system by various sorbents. In this paper we present results of the adsorption of triclosan on a sorbent obtained from production wastes. As sorbent we used activated carbon GP5 which was recycled from civilian gas masks GP5.

2. Experiment

The adsorption experiments of triclosan were performed from ethanol solutions instead of using standard aqueous solutions [14,15]. This allowed a large increase in the concentration of the adsorbate in the solution due to its 100% solubility in ethanol. Thus, the adsorption capacity of activated carbon GP5 towards triclosan could be fully explored.

As sorbent we used recycled activated carbon GP5 from civilian gas masks GP5, taken as it was. The properties of this recycled activated carbon were given in Table 1. Low temperature nitrogen adsorption analyzed by the BET theory was used to find V_{total} and D_b.

Scanning electron microscopy, SEM (Zeiss Gemini LEO 1550) was employed to analyze the structure of the adsorbent. For the characterization, a droplet of the aqueous particle suspension was p;1;laced on a sample holder and left to dry at atmospheric pressure and room temperature and was then sputtered with a gold/palladium mixture to avoid electron charging of the samples during SEM analysis. The samples were studied using an operating voltage of 3 kV and different magnifications.

Triclosan (97%) was purchased from Aldrich. The adsorption process of triclosan was studied in a static mode at the conditions of the adsorption equilibrium in the "triclosan-activated carbon" system. The adsorption experiment was performed atthe following way: at various triclosan concentrations (from 10 to 400 ppm) and a fixed adsorbent loading (1 mg/l) the adsorption isotherms was measured between 3–10 days in order to find the equilibrium values. This process was carried out at constant temperature ($24 \pm 1\,^{\circ}C$). In order to quantify adsorbed amount, the adsorbent was removed from "triclosan-activated carbon" solutions by filtration through membrane filters with a pore size of 0.45 μm. Then triclosan equilibrium concentration was measured spectrophotometrically at a Cary 50 Cons at a fixed wavelength of 280 nm [24,25].

The amount of the adsorbed triclosan on the activated carbon was quantified by a mass balance method.

The adsorption capacity of the carbon (q_{eql}) can be expressed in terms of the triclosan amount adsorbed per unit of sorbent mass, i.e. the uptake (mg/g) is given by:

$$q_{eql} = \frac{(C_{init} - C_{eql})}{m} \quad (1)$$

The sorption efficiency of the system ($Rem\%$) indicated by the percentage of removed triclosan relative to the initial amount, i.e. Rem in% is given by

$$Rem\% = \frac{(C_{init} - C_{eql})}{C_{init}} \times 100 \quad (2)$$

where C_{init} and C_{eql} are, respectively, the initial and equilibrium concentrations of triclosan in solution (mg/l) and m is the carbon dosage (g/l).

As an adsorption model we used the nonlinear Sips (Freundlich–Langmuir) isotherm model [26] in order to obtain a more accurate representation of the adsorption equilibria parameters:

$$q_{eql} = \frac{q_s(K_s C_{eql})^{1/n}}{1 + (K_s C_{eql})^{1/n}} \quad (3)$$

where q_s is the adsorption capacity of the adsorbent, K_s is the equilibrium constant and n is the index of heterogeneity. The parameter n characterizes the interaction between adsorbate and adsorbent, and its magnitude increases with the heterogeneity of the system. It should be noted that for $n = 1$ Eq. (3) becomes a Langmuir type equation. Alternatively, for either C_{eql} or K_s approaching 0, this isotherm reduces to the well-known Freundlich isotherm [27].

3. Results and discussion

In order to determine the concentration of triclosan in the experimental solutions, a linear calibration curve was determined based on absorption spectra (within 230–800 nm) of triclosan/ethanol model solutions with known concentrations in the range between 10 and 400 mg/l. The values of the absorbance intensity were taken at 280 nm which corresponds to the triclosan absorption [25,26]. The obtained calibration curve of triclosan obeyed the Beer-Lambert-Bouguer law.

Fig. 1 represents the SEM images of GP5 activated carbon at different magnifications. Primary particles of a broad size distribution around 300 nm seem to form amorphous agglomerates in the micrometer range, leading to a porous structure which together with micropores attributes for the total pore volume of 0.62 cm^3/g (Table 1).

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