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Characterization of a low cost *Lagenaria vulgaris* based carbon for ranitidine removal from aqueous solutions

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Abstract Practical aspects of *Lagenaria vulgaris* shell conversion to activated carbon were examined along with its use in ranitidine adsorption. Kinetics and isotherms of adsorption onto *Lagenaria vulgaris* carbon (LVC) were correlated to several theoretical adsorption models. The best fit was found in the case of Langmuir and pseudo-second-order model indicating monolayer adsorption. The influence of pH under kinetic study showed slightly hindered adsorption below pH 4. The optimal adsorbent dosage was set to 1 g/L. LVC was characterized by several complementary techniques, including wet chemical techniques such as Boehm's titrations and determination of pH_{PZC} and pH of LVC, which revealed neutral nature of the adsorbent. N₂ sorptometry determined specific surface area of 665 m²/g and significant ratio of micropores in the sample with maximum wall's diameter of 2.2 nm. Fourier transform infrared spectroscopy (FTIR) confirmed the role of lignin and cellulose in the formation of the final LVC structure. Porous structure of the material was proved by using scanning electron microscopy. Preparation of LVC material drew attention as an easy and low-cost process for production of a highly efficient adsorbent which

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exhibited fast kinetics of ranitidine removal in the first minutes of contacting and large adsorption capacity (315.5 mg/g) at equilibrium.

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1. Introduction

It has been shown that Ranitidine is one of the most popular drugs on the planet used on daily base due to frequent gastric problems in humans today. It is manufactured in tons annually and easily incorporated in spontaneous mechanism of environmental pollution, especially in urban areas. Ranitidine is being excreted by urine and feces, parent compound or metabolites reach the sewage system, wastewater treatment plant remove them only partially (Jones et al., 2005), so it eventually ends up in surface water, mainly rivers. In addition, unwanted or expired ranitidine doses are disposed from households and hospitals on daily basis directly to the sewage systems. In the study conducted in Italy, ranitidine was identified in a group of pharmaceuticals with a “priority” for the environment (Castiglioni et al., 2006). This group encounters daily loads of 50–500 mg for 1000 inhabitants in the influents and estimated residual loads, while after its removal in sewage treatment plants and in the river it drops to 25–280 mg for 1000 inhabitants daily, which results from 1.5 to 16 kg/day if we extrapolate this data to the whole of Italy. Its overall presence in water bodies all over the world is significant.

Ranitidine (Fig. 1) is a histamine H_2 receptor antagonist which is prescribed for the treatment of duodenal and gastric ulceration since it efficiently decreases the amount of acid produced in the stomach (Pffaffen and Ortiz, 2010). Ranitidine is a furan derivative, with a nitroamine urea polar group and dimethylamine group enhancing the basic character of its heterocyclic moiety. It is metabolized by demethylation, N- and S-oxidation, but after oral administration, about 30–70% of it is eliminated unchanged in urine within 24 h (Vediappan and Lee, 2011).

Pharmaceuticals cannot be absolutely removed in sewage treatment plants using conventional techniques such as bioremediation and physicochemical treatments, including coagulation, volatilization, sedimentation and filtration (Sires and Brillas, 2012). On the other hand, adsorption has shown promising results. Up to now, adsorption of various drugs onto zeolites (Fukahori et al., 2011), chitosan (Kyzas et al., 2013), clays (Figuerola et al., 2004) and activated carbons prepared from different residues (Cabrita et al., 2010) was reported. Photoassisted degradation of many pollutants, including ranitidine, is an efficient technique, but it deserves special plants and expensive equipment (Addamo et al., 2005).

Powdered and granular activated carbons are considered the most potent adsorbents used commercially. However, they are expensive (Salleha et al., 2011) which imposes the need for searching a cheaper preparation procedures or precursors. Importantly, the resulting properties of the carbon depend on the used precursor (Menendez-Diaz and Martin-Gullon, 2006). Recently, many interesting alternatives have been proposed in the scientific literature, including preparation of activated carbons from livestock sewage sludge (Wu et al., 2014), aguaje and olive fruit stones (Obregón-Valencia and Rosario Sun-Kou, 2014), reedy grass leaves (Xu et al., 2014), orange peel (Fernandez et al., 2014), Golden Bamboo (González and Pliego-Cuervo, 2014), and marigold straw (Qin et al., 2014). The main goal of such studies is to examine the possibilities for producing novel and cheaper forms of carbon adsorbents with standard or even better properties.

We examined activated carbon derived from *Lagenaria vulgaris* as a suitable adsorbent for the small scale ranitidine adsorption under laboratory conditions. *L. vulgaris* is a climbing, hardy plant mainly grown on alluvial sandy soil and it belongs to the family *Cucurbitaceae*. Although it has a wide medicinal use (Ghule et al., 2009), it is mainly used as a container for water and food worldwide, known as “The Bottle Gourd” (Decker-Walters et al., 2004). The outer shell of the plant is composed of lignin and cellulose (Mitić-Stojanović et al., 2012). Such structure is recognized as one of the key features in production of activated carbons.

The goal of this study was to examine practical options for conversion of *L. vulgaris* to activated carbon and its possible use in ranitidine removal by adsorption from contaminated water sources. Kinetics, isotherms, effect of pH of ranitidine adsorption and adsorbent dosage of LVC were investigated under batch conditions. Detailed characterization of adsorbent was performed in order to acquire as much information as possible about its structure and reactivity.

2. Materials and methods

2.1. Adsorbent preparation

Carbon adsorbent was derived from *L. vulgaris* shell by the thermo-chemical procedure. Initially, the dried fruit was rinsed with tap and then demineralized water in order to remove dust and soil from it. Small pieces of broken shell were manually

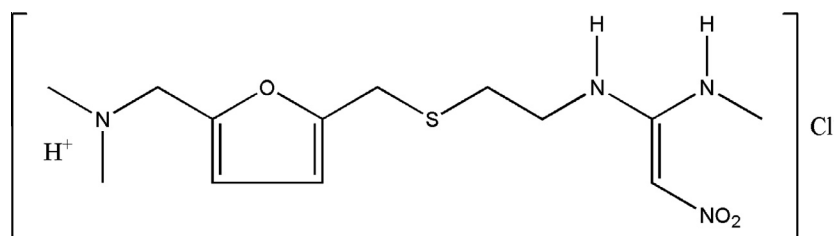


Figure 1 Molecular structure of ranitidine hydrochloride.

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