



Effects of power of ultrasound on removal of Cu(II) ions by xanthated *Lagenaria vulgaris* shell



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ABSTRACT

This study was aimed at removal of Cu(II) ions from aqueous solutions by ultrasound-assisted sorption onto xanthated *Lagenaria vulgaris* shell (xLVB). Sorption kinetics, isotherms and ultrasonic power were investigated. Sorption isotherm data were best fitted by the Langmuir model. The acoustic power was an important factor for the enhancement of the removal of copper. The maximum sorption capacity in the presence of ultrasound was much greater than in absence of ultrasound. The sorption process in the presence and absence of ultrasound obeyed to the pseudo-second order kinetics, indicating that the rate-limiting step might be a chemical reaction between the sorbent and the sorbate. The intraparticle diffusion and Boyd model indicated that the intraparticle diffusion was not the only rate-limiting step in the sorption of Cu(II) ions on xLVB but that the other processes also control the rate of sorption.

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1. Introduction

One of the most important global environmental problems is water pollution, especially pollution by heavy metals. The improper disposal of heavy metals leads to pollution. This is especially significant because of the facts that heavy metals are non-degradable, hence are accumulated in living organisms by food chains. Copper and its salts are widely used industrial chemicals as agricultural fungicides, petroleum refining, electroplating baths, mining etc. (Huang et al., 2007; Al-Asheh et al., 2002). Excessive sorption of copper can cause serious health problems such as heart, liver, kidney, pancreas and brain damage, intestinal distress and anemia (Abu-Al-Rub et al., 2006). The toxic heavy metals must be effectively removed from the wastewaters, because of its numerous consequences to human and overall environment. The most common used methods for reducing heavy metal concentration are chemical precipitation, chemical oxidation and/or reduction, filtration, ion exchange, electrochemical treatment, application of membrane technology and evaporation recovery (Canizares et al., 2005; Feng et al., 2000; Kumano et al., 2006; Ozaki et al., 2002; Trivunac and Stevanovic, 2006). However, there are numerous disadvantages of these technology processes: incomplete metal removal, high costs

because of requirements for expensive equipment and monitoring system, low efficiency (Brady et al., 1999). Alternative technology for conventional wastewater treatment is sorption, as efficient, clean and cheap technology.

Although in literature there are no numerous information and studies about the removal of metal ions from aqueous solution by sorption combined with ultrasound, this technique is used in industry, especially in wastewater treatment.

Present study deals with investigation of ultrasound-assisted biosorption of Cu(II) ions from aqueous solutions on xanthated *Lagenaria vulgaris* shell. Therefore, the overall objective of the study was to evaluate the effects of power of ultrasound on the process equilibrium and kinetics and to define the best isotherm and kinetic sorption models. Also, there are no records about the influence of US on Cu(II) ions removal by xanthated biomass.

2. Experimental

2.1. Reagents and biosorbent

All chemicals were of reagent grade and used without further refinement. HNO₃, NaOH, CS₂ and Cu(NO₃)₂ were purchased from Merck (Germany). All solutions were prepared with deionized water. Standard metal stock solution was prepared by dissolving given amounts of pure Cu(NO₃)₂·3H₂O. *L. vulgaris* shell was treated with diluted nitric acid and after that by sodium hydroxide,

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producing a basic *L. vulgaris* biosorbent (LVB). Xanthation was carried out by treatment of LVB with carbon disulfide in alkaline medium. This synthesis and characterization are discussed in previous research (Kostić et al., 2014).

2.2. Batch biosorption experiments

Batch sorption experiments were conducted in ultrasonic bath (Sonic, Serbia; total nominal power: 50 W) that operates at 40 kHz frequency (The experimental setup is shown in supplementary Fig. S1). The temperature in the ultrasonic bath was held at 20 °C (± 0.2 °C) by recirculating water from a thermostated bath Julabo F12-ED (Refrigerated/Heating Circulator, Germany). Sorption experiments were performed in 400 cm³ glass vessel, placed in ultrasonic bath with mechanical stirrer immersed in (200 rpm). Working standard solutions were prepared just before use by the appropriate dilution of the stock solutions (1.00 g dm⁻³). All experiments were performed with 4.0 g dm⁻³ quantity of dried biosorbent suspended in 250 cm³ of Cu(II) solutions in a concentration range between 10 and 400 mg dm⁻³ of Cu(II) ions at initial pH 5.0 \pm 0.2. At pH value 5.0 removal efficiency achieved maximum level, which was explained in previous research (Kostić et al., 2013). Samples (4.0 cm³) were taken before mixing the sorbent solution and Cu(II) ion bearing solution and at pre-determined time intervals (0, 1, 5, 10, 20, 40, 90, 120 and 180 min) for the residual Cu(II) ion concentration in the solution. The concentrations of residual Cu(II) ions in the solution were determined by an atomic adsorption spectrometer Analyst AA 300 (Perkin Elmer, USA). The amount of metal sorbed q_t (mg g⁻¹) (Meitei and Prasad, 2014) and the removal efficiency (RE) (Li et al., 2013) of metal ions was calculated using the Eqs. (1) and (2), respectively.

$$q_t = \frac{(c_0 - c_t)V}{m} \quad (1)$$

$$\text{RE}(\%) = \frac{c_0 - c_t}{c_0} \times 100 \quad (2)$$

where, c_0 and c_t are the initial and final concentrations of the metal ion in solution (mg dm⁻³), V is the solution volume (dm³) and m is the mass of the sorbent (g).

All the experiments were conducted in triplicate and average values were used in the data analysis using OriginPro 9.0 software (OriginLab Corporation, USA).

3. Results and discussion

3.1. Contact time

To investigate the effect of contact time on Cu(II) ions sorption, experiments were conducted at initial metal concentration of 50 mg dm⁻³ in the presence and absence of ultrasound, the acoustic power used was 50.0 W. In the presence of ultrasound the experimental results show high sorbent efficiency in removing Cu(II) ions: sorption equilibrium was obtained in 20 min of sorbent-sorbate contact, when 97.5% of total Cu(II) ions were removed (Fig. 1). After 180 min, initial concentration of Cu(II) ions decreases and removal efficiency was 98.8%. In the presence of ultrasound a shorter time is required to reach equilibrium. In the silent sorption, equilibrium was attained after about 50 min of contact time and RE was 80.1%. (Kostić et al., 2013). The shorter time to reach equilibrium and the faster rate of removal in the presence of ultrasound and mechanical stirring were attributed to the strong convective currents occurring within the reactor. These effects associated with the hydrodynamic phenomena due to cavitation and mechanical stirring are responsible for the perfect mixing of the vessel content (Hamdaoui et al., 2005).

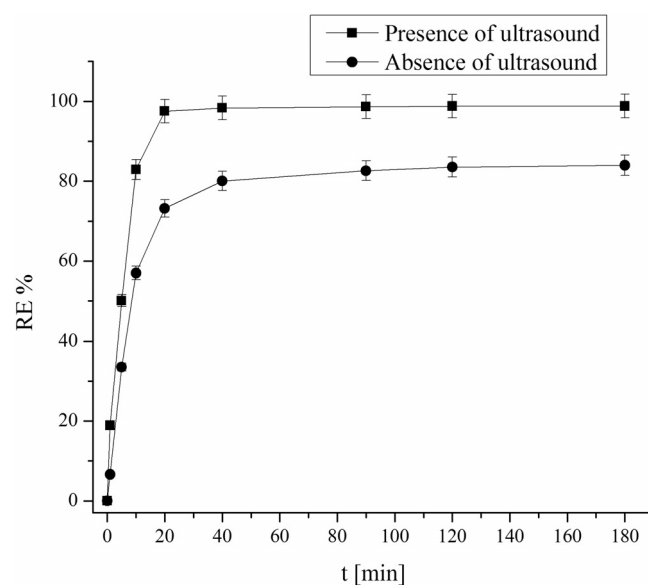


Fig. 1. Effects of the contact time in the presence and absence of ultrasound on the biosorption of Cu(II) ions on xLVB.

3.2. Kinetic study

The reaction kinetic models (the pseudo first-order and pseudo second-order model) were applied to study the reaction kinetics of the sorption process, whereas the intraparticle diffusion model and Boyd's were further tested to determine the diffusion limitations of the sorption of Cu(II) ions onto xLVB.

The original equations for pseudo first-order, pseudo second-order, intraparticle diffusion and Boyd's model are presented in detail in paper by Ho and McKay (1998); Yao et al. (2010); Hossain et al. (2012).

The determination coefficients (r^2) for the pseudo first-order kinetic model (Table 1) obtained for all the studied acoustic power were relatively low. Also, it was found that the calculated q_e values did not agree with the experimental q_e values. This suggests that the sorption of copper did not follow pseudo-first-order kinetics. The experimental data were observed to fit well to the pseudo-second order model. The determination coefficients for the pseudo-second order equation obtained at all the studied acoustic powers were high (from 0.9871 to 0.9970). Also, the q_e^{cal} values are in good agreement with the experimental data. In addition, it can be seen in Table 1. that with an increase in acoustic power, the rate constant of sorption (k_2) increases. In this case of Cu(II) ions sorption from aqueous solution, ultrasound increased significantly the sorption capacity and increase of k_2 was very high 0.0108 and 0.0235 g mg⁻¹ min⁻¹ for silent and ultrasound-assisted sorption, respectively. Based on the obtained data, the primary benefit of sonication is an increase in the speed of sorption, especially in the initial period. The values of the sorption capacity for sorption of Cu(II) ions by xLVB increased from 11.46 to 12.92 mg g⁻¹ while the acoustic power increased from 0 to 50.0 W. As can be seen from Fig. 1, amount of sorbed Cu(II) ions increased from 10.49 to 12.35 mg dm⁻³ with the increase in acoustic power from 0 to 50, indicating significantly influence of ultrasound. This means that ultrasound has an impact on both the rate of reaching equilibrium and the sorption capacity. The removal of Cu(II) ions was higher in the presence of ultrasound than in its absence, due to the cavitation process which increased the diffusion process. In accordance with the pseudo second-order reaction mechanism, the overall rate of Cu(II) sorption processes appear to be controlled by the chemical processes. The mechanism of ion-exchange which is probably

Table 1
Comparison of the kinetic model parameters for the sorption of Cu(II) ion at 50.0 mg dm⁻³ initial concentration.

Acoustic power [W]	0	14.9	20.2	26.4	33.5	41.3	50.0
Pseudo-first order							
k_1	0.0882	0.0997	0.1083	0.0908	0.0975	0.1828	0.1525
q_e^{cal}	9.1908	10.0588	9.7047	9.0114	9.4974	12.5576	9.7682
r^2	0.9769	0.9961	0.9828	0.9192	0.9319	0.9899	0.9170
Pseudo-second order							
k_2	0.0108	0.0126	0.0118	0.0159	0.0175	0.0232	0.0235
q_e^{cal}	11.4639	11.6077	11.8652	12.1477	12.6072	12.6342	12.9216
r^2	0.9901	0.9954	0.9871	0.9970	0.9975	0.9973	0.9971
Intraparticle diffusion model							
K_{id1}	2.2114	2.2344	2.4373	2.5181	2.6101	2.7320	2.9014
C_1	-0.5521	-0.4255	-0.5893	-0.3270	-0.1633	0.0377	-0.0728
r^2	0.9667	0.9691	0.9496	0.9767	0.9860	0.9885	0.9705
Boyd model							
D_1 (0.8 mm)	5.731×10^{-5}	6.472×10^{-5}	7.032×10^{-5}	5.894×10^{-5}	6.329×10^{-5}	9.903×10^{-5}	1.187×10^{-4}
r^2	0.9769	0.9899	0.9170	0.9319	0.9192	0.9828	0.9961

involved is favored by the presence of the xanthate groups with strong ion-exchange potential (Kostić et al., 2013, 2014). The results of intraparticle diffusion model for all studied initial Cu(II) concentrations showed multi-linearity and two stages in sorption in the absence and in the presence of ultrasound (supplementary Fig. S2c). The values of C for ultrasound-assisted and the silent sorption are given in Table 1. The intercept was greater in the presence of ultrasound than in its absence and is proportional to the boundary layer thickness and depicting the higher ultrasound-assisted sorption capacity of the xLVB, compared to silent condition. The slope of the linear portion characterizes the rate parameter K_{i1} corresponding to the intraparticle diffusion. It was found that the rate constant increased from 2.211 to 2.901 with increasing of ultrasound power from 0 to 50.0 W, i.e., ultrasound enhances the mass transport in the pores. The sorption rate increases in the presence of ultrasonic field. Initial sorption rates in the presence of ultrasound is higher than in the absence of ultrasound, due to reduced mass transfer resistances. These results indicate that ultrasound enhances the mass transport in the pores. This behavior could be attributed to the induced turbulence and additional convective mass transport inside the pores caused by micro-jets (Hamdaoui et al., 2003). It can also be observed that the plots did not pass through the origin; this was indicative of some degree of boundary layer control and this further showed that the intraparticle diffusion was not the only rate-limiting step, but other processes might control the rate of sorption. This was further tested by determining the pore diffusion coefficient (D [cm² s⁻¹]) (Taty-Costodes et al., 2003). According to Singh et al. (2006), D value in the order of 10^{-11} cm² s⁻¹ is indicative of pore particle diffusion as the rate determining step. In this investigation, the values of D were 4.29×10^{-5} and 2.47×10^{-5} cm² s⁻¹ in the presence and absence of ultrasound, respectively. As these amounts are larger than 10^{-11} cm² s⁻¹, then the pore diffusion in the sorption process is not the only rate-limiting step and some other steps may be involved (Singh et al., 2006). Plots of Bt versus t are shown in supplementary Fig. S2d. If the plot is linear and passes through the origin then pore-diffusion controls the rate of mass transfer. If the plot is nonlinear or linear but does not pass through the origin, then it is concluded that film-diffusion or chemical reaction control the sorption rate. From supplementary Fig. S2d the plot was neither linear nor passed through the origin. This could indicate that film diffusion (mass transfer) is the rate-limiting biosorption process for Cu(II) ions. Therefore, it is confirmed that the first linear segment represents film-diffusion (or chemical reaction), and that the following linear segments represent pore-diffusion and equilibrium (supplementary Fig. S2c). If film diffusion was to be the rate-determining step, the value of D_1 should be in the range of 10^{-5} – 10^{-8} cm² s⁻¹, while, if particle diffusion was

the rate-determining step, the D_1 value should be in the range of 10^{-11} – 10^{-13} cm² s⁻¹ (Gasser et al., 2006). The effective diffusion coefficients, as estimated from supplementary Fig. S2d and shown in Table 1, confirming that the slowest step in copper sorption on xLVB was film diffusion. Supplementary Fig. S2c and Table 1 shows the increase of the pore diffusion coefficient D_1 with increasing acoustic power, i.e., ultrasound enhances the mass transport in the pores.

3.3. Sorption isotherms

Sorption isotherms show the distribution of solute between the liquid and solid phase equilibrium conditions. To determine the maximum sorption capacity of the biomass, isotherms studies were conducted with varying initial Cu(II) ions concentration from 10 to 400 mg dm⁻³ at biomass dose of 4.0 g dm⁻³. Sorption process is described by four widely used isotherms: Langmuir, Freundlich and Temkin model. The original equations are presented in detail in paper by Gorgievski et al. (2013) and Wang et al. (2015).

The isotherm models were fitted with experimental data, and the value of the isotherms constants are listed in Table 2. The sorption characteristic of Cu(II) ions on the functionalized xLVB biosorbent most closely follows Langmuir isotherm model. Higher r^2 value for the Langmuir isotherm model than for the Freundlich isotherm model might be due to homogeneous distribution of active sites on biosorbent surface without interaction between sorbed ions. The R_L values were found to be less than 1 and greater than 0 for all experiments carried out at different initial concentrations. These results show that Cu(II) ions sorption on xLVB is favorable. From Table 2, q_{max} in the presence of ultrasound was obtained as 33.34 mg g⁻¹. Without the assistance of ultrasound, maximum loading capacities are decreased and amounts

Table 2
Equilibrium model parameters for sorption of Cu(II) ions onto xLVB in the presence of ultrasound (acoustic power 50.0 W).

Equilibrium model	Parameter	Value
Langmuir isotherm	K_L (dm ³ mg ⁻¹)	0.4798
	q_{max} (mg g ⁻¹)	33.3444
	R_{L400}	0.0052
	r^2	0.9990
Freundlich isotherm	K_F (dm ³ g ⁻¹)	9.9254
	n	3.8139
	r^2	0.9481
Temkin isotherm	b (J mol ⁻¹)	734.32
	K_t (dm ³ mg)	93.2090
	r^2	0.9637

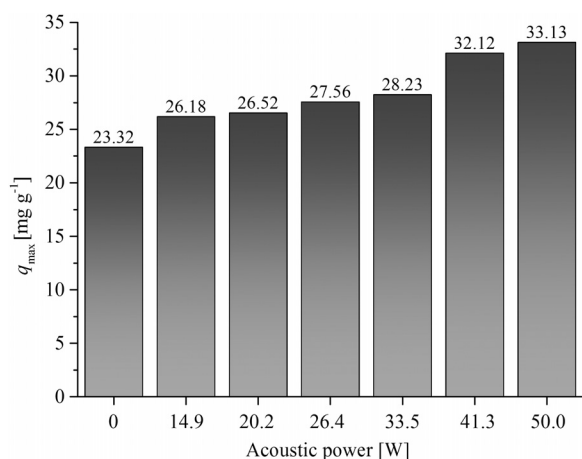


Fig. 2. Effect of acoustic power on the sorption of copper by xLVB.

23.32 mg g^{-1} for Cu(II) ions. This was attributed to cavitation effects which increased capability of the porous particle structure for Cu(II) ions sorption and/or the appearance of new sites of sorption by disruption of sorbent particles. Also, Entezari and Soltani (2008) have found that the maximum loading capacities from Saffron significantly increased under sonication conditions. The coefficients of determination for Freundlich isotherm were low (Table 2), showing a bad linearity, so that Freundlich model could not be applied for describing the ultrasound-assisted sorption of Cu(II) onto xLVB. The coefficients of determination were high showing a good linearity. Temkin constant, B , related to heat of sorption for the Cu(II) ions was 0.7 kJ mol^{-1} indicating that interactions between the sorbate and sorbent are physisorption. Value for q_{\max} estimated by Langmuir model is 33.34 mg g^{-1} . This value is higher than q_{\max} obtained in similar conditions involving Saffron 30.60 mg g^{-1} (Entezari and Soltani, 2008), Banana Peel 20.37 mg g^{-1} (Hossain et al., 2012) and Chestnut shell 12.56 mg g^{-1} (Yao et al., 2010). However, q_{\max} in this paper is lower than ones obtained in experiments with *Chlorella vulgaris* 58.82 mg g^{-1} (Abu-Al-Rub et al., 2006) and Bone char 47.62 mg g^{-1} (Cheung et al., 2000).

3.4. Effect of acoustic power

Study of the effects of acoustic power was conducted with varying initial Cu(II) ions concentration from 10 to 400 mg dm^{-3} at biomass dose of 4.0 g dm^{-3} . It was observed that the sorption efficiency increases with the increase in acoustic power from 0 to 50.0 W, i.e., 1.5 times (Fig. 2). The stronger the acoustic power is, the greater is the intensity of ultrasonic field, which led to the improvement of microstreaming, microturbulence, shock waves and microjets and to enhancement of mass transfer in the system and thus increased the amount of copper sorption. This indicates that the xLVB can absorb acoustic energy and this sorption can lead to an increase of sorption. Therefore, these effects of ultrasound (increasing of mass transfer, new sites and sorption of acoustic energy by xLVB) could be reasons for the enhancement of sorption at higher intensity (Entezari and Soltani, 2008).

4. Conclusion

The effect of ultrasound has a dual dimension compared with that of silent sorption: increases the rate of the process and increases the sorption capacity. The highest value of r^2 was obtained from the Langmuir model. The maximum sorption capacity of the sorbent for Cu(II) ions, in the presence of ultrasound is 1.5 times greater than that in the absence of ultrasound. Under

the influence of ultrasound Cu(II) ions are sorbed deeper into the cavities and pores of biosorbent compared to silent sorption. The removal of Cu(II) ions was faster and more efficient in the presence of ultrasound due to the cavitation process which facilitated the mass transfer and increased the surface area of the sorbent. The kinetics of the sorption process was found to follow the pseudo-second order kinetic model. The diffusive models also properly correlate with the obtained results, which prove the significance of diffusion both at the boundary layer and in the pores of the sorbent. As the intraparticle diffusion model and Boyd kinetic model fits well the kinetic data, sorption of Cu(II) ions on xLVB is also diffusion controlled. The mechanism of biosorption includes both, intraparticle diffusion and chemical reaction.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.ecoleng.2016.01.063>.

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