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Review

Brown marine macroalgae as natural cation exchangers for toxic metal removal from industrial wastewaters: A review



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ABSTRACT

The discharge of inadequately treated or untreated industrial wastewaters has greatly contributed to the release of contaminants into the environment, including toxic metals. Toxic metals are persistent and bioaccumulative, being their removal from wastewaters prior to release into water bodies of great concern. Literature reports the use of brown marine macroalgae for toxic metals removal from aqueous solutions as an economic and eco-friendly technique, even when applied to diluted solutions. Minor attention has been given to the application of this technique in the treatment of real wastewaters, which present a complex composition that can compromise the biosorption performance. Therefore, the main goal of this comprehensive review is to critically outline studies that: (i) applied brown marine macroalgae as natural cation exchanger for toxic metals removal from real and complex matrices; (ii) optimised the biosorption process in a fixed-bed column, which was further scaled-up to pilot plants. An overview of toxic metals sources, chemistry and toxicity, which are relevant aspects to understand and develop treatment techniques, is initially presented. The problem of water resources pollution by toxic metals and more specifically the participation of metal finishing industries in the environmental contamination are issues also covered. The current and potential decontamination methods are presented including a discussion of their advantages and drawbacks. The literature on biosorption was reviewed in detail, considering especially the ion exchange properties of cell wall constituents, such as alginate and fucoidan, and their role in metal sequestration. Besides that, a detailed description of biosorption process design, especially in continuous mode, and the application of mechanistic models is addressed.

1. Introduction

The rapid urbanisation and industrialisation in the last decades followed by population increase have caused serious economic, social, environmental and political problems in almost all the world, resulting in adverse effects on the life quality. Among these effects, the pollution of freshwater bodies with consequent deterioration in water quality can be cited. The discharge of inadequately treated or untreated sewage and industrial wastewaters has greatly contributed to toxic compounds released into the environment, including toxic metals (Jern, 2006; Vilar et al., 2008c).

A wide range of treatment methods, such as chemical precipitation, coagulation/flocculation, electrochemical processes, membrane filtration, ion exchange, adsorption and others have been applied for the removal of toxic metals from water. However, most of these techniques could be not effective enough for low metal concentrations or might be very expensive because of huge investment and/or operational costs, high chemical reagent and energy requirements, as well as the proper disposal of sludge (Abdolali et al., 2016; Syukor et al., 2016).

The application of the biosorption technique for toxic metal ions removal from aqueous solution has been demonstrated to be an economic and eco-friendly technique, even when applied to diluted

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Nomenclature	
a	empirical parameter of Yan model
A_c	cross-sectional area of the column (cm ²)
a_p	specific area of the thin plate particles (1/cm)
C_{bp}	breakthrough concentration (mg/L)
C_E	feed concentration of species j (mg/L or mmol/L or mEq/L)
C_{out}	concentration of species j at the column outlet (mg/L or mmol/L or mEq/L)
C_H	concentration of protons in solution (mol/L)
C_j	concentration of species j in the liquid phase at time t (mg/L or mmol/L or mEq/L)
$C_{j,0}$	initial concentration of species j in the liquid phase (mg/L or mmol/L or mEq/L)
$C_{j,e}$	equilibrium concentration of species j in the liquid phase (mg/L or mmol/L or mEq/L)
D	column inner diameter (cm)
d	adsorbent dose (g/L)
D_{az}	axial dispersion coefficient (cm ² /s)
$D_{h,j}$	homogeneous diffusion coefficient inside the particle (cm ² /s)
d_p	equivalent particle diameter (mm)
f	swelling factor
f_{LUB}	fraction of unused bed (%)
f_{LUSE}	fraction of used bed (%)
K	equilibrium constant for linear model (L/g)
$K_{H,i}^I$	average value of the affinity distribution for the protons
$K_{D,i}^H$	selectivity coefficient between divalent ions ($D = Mg^{2+}$, Ca^{2+} or Cu^{2+}) in the particle and H^+ ion in solution
$K_{D,i}^{Cu}$	selectivity coefficient between divalent ions ($D = Mg^{2+}$, Ca^{2+} or Cu^{2+}) in the particle and Cu^{2+} ion in solution
$K_{M,i}^H$	selectivity coefficient between monovalent ions ($M = Na^+$ or K^+) in the particle and H^+ ion in solution
$K_{M,i}^{Cu}$	selectivity coefficient between monovalent ions ($M = Na^+$ or K^+) in the particle and Cu^{2+} ion in solution
K_L	equilibrium constant for Langmuir multicomponent model (L/mEq)
$k_{p,j}$	mass transfer coefficient for intraparticle diffusion of species j (cm/s)
K_1	Thomas model rate constant (L × Eq/day)
L	bed height (cm)
L_{MTZ}	length of the MTZ (cm)
L_{UB}	length of the unused bed (cm)
L_{USE}	length of the used bed (cm)
m	algae dry weight (g)
$m_{H,i}$	width of their peak in the Sips distribution
n	number of components
Pe	Peclet number
pK_a	dissociation constant
Q	feed flow rate (mL/min)
q	uptake capacity (mg/L or mmol/L or mEq/L)
q_b	operating capacity until achieving the breakthrough point (mg/g or mEq/g algae)
q_E	total capacity (mg/g or mEq/g algae)
q_j	concentration of species j in the solid phase at time t (mg/g or mmol/g or mEq/g algae)
$q_{j,e}$	equilibrium concentration of species j in the solid phase (mg/g or mmol/g or mEq/g algae)
q_{max}	total capacity from batch experiments (mg/g or mmol/g or mEq/g algae)
$Q_{max,i}$	maximum concentration of each functional group (mmol/g)
Q_s	specific flow rate (BV/h)
q_T	total concentration of exchangeable binding sites (mg/g or mmol/g or mEq/g algae)
q_j	average concentration of species j in the solid phase at time t (mg/g or mmol/g or mEq/g algae)
r	wastewater treatment ratio (m ³ /kg)
R	radius of the spherical particle (cm)
RE	removal efficiency (%)
R_p	half of the thickness of the particle (cm)
t	time (min or h)
t_{bp}	time at breakthrough point (min or h)
t_e	exhaustion time (min or h)
t_{MTZ}	time of MTZ (min or h)
t_{st}	stoichiometric time (min or h)
u	superficial fluid velocity or linear flow rate (cm/s)
u_i	interstitial fluid velocity (cm/s)
u_{sh}	shock wave velocity (cm/min)
V	volume of the solution (L)
V_b	volume of wastewater purified until breakthrough point (m ³)
V_c	column volume (mL or L)
V_r	resin volume (mL or L)
W	mass of resin for the scale-up (kg)
z	bed axial position (cm)
<i>Greek letters</i>	
ε	bed porosity
ξ	mass column capacity factor
ρ_p	particle density (g/L, dry basis)
τ	residence time (min)
$\tau_{d,j}$	time constant for diffusion of species j into the particle (s)

solutions. This is due to the high quality of effluent generated, the use of inexpensive biosorbents, the minimization of chemicals consumption and hazardous sludge production, the possibility of biosorbent re-utilisation, the capacity to operate under a broad range of service conditions and, the possibility of metal recovery (Davis et al., 2003; Kratochvil and Volesky, 1998; Zhao et al., 2011). However, biosorption efficiency depends on the type of biomass and the wastewater composition (Vijayaraghavan and Balasubramanian, 2015). An ideal biosorbent should be largely available in nature or as part of an industrial waste material, easily collected and, with minor or no treatment required (Gadd, 2009; Vieira and Volesky, 2000). The application of dead marine macroalgae as biosorbent for toxic metal ions removal from aqueous solutions has been shown as very promising, once the biomass is abundantly available, present high metal removal efficiency, potential for regeneration and metal recovery and, the biosorption process

can be easily operated and controlled using mathematical models (Davis et al., 2003; Hannachi et al., 2015; Vijayaraghavan and Balasubramanian, 2015). Toxic metal uptake ability of dead marine macroalgae can be attributed to the presence of several negative charge functional groups present on the cell walls surface, such as hydroxyl, carboxyl, sulphhydryl, sulphate and amino groups (Davis et al., 2003; Kloareg and Quatrano, 1988). The presence of different polysaccharides in marine macroalgae cell walls is the reason why diverse groups of seaweed have different metal removal ability. The higher ion exchange capacity of brown marine macroalgae over other types of seaweeds may be explained by the presence of a high content of alginate (Misurcová, 2012). Alginate comprises between 10 and 40% of the dry weight of brown marine macroalgae, following by fucoidan that constitutes 5–20% and lastly by cellulose that only accounts for 1–8% (Rioux and Turgeon, 2015). K^+ , Na^+ , Ca^{2+} and Mg^{2+} , ions that exist in high

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