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Valorization of Fruit Peel Wastes as Biosorbent for Heavy Metals Removal -A review

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ABSTRACT: The presence of heavy metals into natural waters represents a serious problem for human health. A cost-effective alternative for dealing with heavy metal contaminated effluents is biosorption, a process based on the use of biological materials. The valorisation of agricultural wastes to treat wastewater represents a promising circular economy approach. In this work, the use of fruit peel wastes as biosorbent has been evaluated through literature review. The most relevant aspects of biosorption mechanisms, factors influencing the technology efficiency and techniques used to characterize the biomass are presented. Physical and chemical pre-treatments to overcome limitations associated with the use of fruit peel as biosorbent, including low adsorption capacity and release of soluble organic compounds, have been reported. Furthermore, immobilization of biomass to deal with potential limitations of the use of fruit peel wastes in fixed bed columns to improve mechanical strength have been discussed as a significant improvement for biosorption processes.

KEYWORDS: Biosorption, circular economy, continuous processes, fruit peel wastes.

I. INTRODUCTION

The increasing presence of heavy metals in water due to anthropogenic activities, including industrial, agricultural, and domestic run-off, is an alarming cause of the water deterioration [1]. The direct effect of metal content, even at low concentration, in human health through drinking water resources requires the development of technologies for the removal of heavy metals in effluents. Conventional techniques, such as chemical treatment, ion-exchange, chelation, solvent extraction and electrolytic methods have been widely assessed as solutions. Nevertheless, many economic and technical reasons limit the success of these techniques which have been reported to be ineffective at low metal concentrations[2]. Therefore, research efforts are focused on the development of new methods not only for the removal but also for the recovery of metals contained in water. These alternatives should overcome typical limitations of traditional methods, such as high operational costs associated with high energy consumption, incomplete metal recovery, production of high amount of secondary sludge and low removal efficiency[3,4]. Biosorption has been proposed as an emerging and low-cost alternative to the existing technologies for the removal of heavy metals from aqueous effluents. This technique is based on the use of biological materials as adsorbents[5]. A complex combination of mechanisms, including adsorption, absorption, ion exchange, surface complexation and precipitation, are involved in biosorption processes[6].

A high variety of biological materials has been proposed as biosorbent due to their affinity for organic and inorganic pollutants. These could be categorized as: bacteria (gram-positive, gram-negative bacteria and cyanobacteria), fungi (molds, mushroom and yeast), algae (micro- and macro-algae, brown and red seaweed), industrial (wastewater sludges, food wastes, etc.), agricultural (rice straws, wheat bran, fruit wastes, etc.) and natural residues (plant residues, tree barks, etc.)[7]. The use of low-cost waste biomass is a key factor to be considered in the application of biosorption. The valorisation of a waste to be applied as a biosorbent for wastewater treatment represents a promising circular economy strategy.

The consumption of fruit generates a significant amount of fruit peel wastes from food processing and agriculture industries which are usually disposed at landfills. According to the Food and Agriculture Organization of the United Nations (FAO) (Fig.1), the global production of fresh fruit in the world was approximately $3.95 \cdot 10^7$ tonnes in 2019[8]. Moreover, as the global population increased, the fresh fruit production is expected to increase dramatically which entails the growth of the fruit peel waste. The management of this solid waste, which represents an environmental concern, should be addressed. Therefore, the use of these fruit peels as a biomass resource to produce useful

biosorbents has been reported as a promising alternative to other commercial adsorbents materials[9]. The present paper aims to analyse the use of fruit peel as biosorbent to remove metals ions from aqueous effluents.

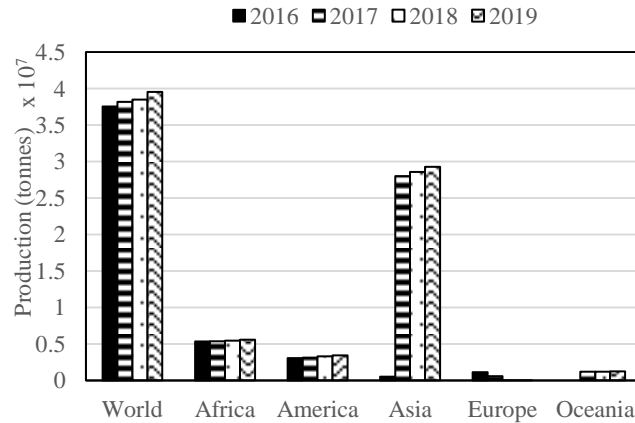


Fig. 1. Global fresh fruit production during 2016, 2017, 2018 and 2019. Source: Food and Agriculture Organization of the United Nations (FAO)[8].

II. BIOSORPTION-AN OVERVIEW

The biosorption of heavy metals on fruit peels is based on the attraction of ionic species by the solid phase through several mechanisms. The complexity of biological biomass makes difficult to determine the specific mechanisms involved in the metal removal from biosorbents. The process can be described as a single mechanism or as a combination of several mechanisms, such as chemisorption, precipitation, microprecipitation, adsorption, adsorption on surface and pores, chelation complexation and ion exchange. The binding mechanisms of metals onto fruit peels is highly influenced by numerous factors, including properties of the sorbates, operating parameters, and physical and chemical properties of the biosorbent. Several techniques are applied to a better knowledge about biosorption mechanisms (Table 1)[9–11].

Table 1. An overview on biosorption mechanisms, relevant factors influencing processes and techniques used to describe biosorption mechanisms.

Mechanisms					
Chemisorption	Precipitation/microprecipitation	Adsorption on surface and pores		Chelating complexation	Ion exchange
Factors influencing biosorption mechanisms					
Chemical properties of metal ions		Properties of biosorbent		Process parameters	
<ul style="list-style-type: none"> • Ionic radius • Oxidation state • Molecular weight • Hydrated radius, etc. 		<ul style="list-style-type: none"> • Structure of the biomass surface • Functional groups • Particle size, etc. 		<ul style="list-style-type: none"> • Temperature • Flow rate • Solid to liquid ratio • Initial concentration of metals • pH, etc. 	
Characterization to identify mechanism of biosorption					
Fourier Transformed Infrared Spectroscopy (FTIR)	X-ray Photoelectron Spectroscopy (XPS)	Scanning Electron Microscopy Energy dispersive X-Ray (SEM-EDX)	Titration	X-Ray diffraction analysis (XRD)	Thermogravimetric analysis (TGA)

III. FRUIT PEELS

The chemical composition of fruit peels consists basically of cellulose, hemicellulose, lignin, lipids, proteins, sugar, polysaccharides, and pigments. The presence of functional groups contained in the peel wastes are mainly hydroxyl, carboxyl, sulfhydryl, phosphate and amino. The important role of the functional groups in metal uptake have been widely evaluated using spectroscopy techniques. The importance of different groups involved in metal binding depends, among other parameters, on pH. According to Choynacka et al., the most relevant functional groups for different pH values are: pH 2-5: carboxyl group, pH 5-9: carboxyl and phosphate group, pH 9-12: carboxyl, phosphate and hydroxyl (or amine group)[12]. The exchange of protons or light metal cations bound to functional groups with metal cations

contained in an aqueous effluent has a great relevance on the biosorption process. Experimental FTIR results of several studies focused on lemon [13], banana [14], durian[15], grapefruit [16], passion fruit [17] and orange [18] peels, showed the importance of identifying functional groups located in the surface before and after metal loaded to elucidate the mechanisms of biosorption. The morphological changes of the biomass surface combined with information about the concentration of metals on the surface is usually evaluated through SEM-EDX. For adsorption processes, the surface area is a key factor. However, comparing fruit peel wastes with other commercial adsorbents, such as activated carbon, the surface area of this biomass waste is very low which is a characteristic of carbonaceous materials [19]. The BET surface area reported from different studies is ranged from 2 [20] to 119 m² g⁻¹ [21]. However, a low surface does not represent a drawback for biosorption processes since, as previously described, biosorption involves a variety of mechanisms which are not limited by surface area. Regarding the importance of oxidation state of metals bound on fruit peel, the XPS analysis is proposed to obtain valuable information about biosorption phenomena. The reduction and oxidation of metals could be described with this technique which provides relevant information for a better understanding of metal binding. With the aim of evaluating the thermal stability of the fruit peel as biosorbent, TGA analysis are carried out. The thermal decomposition behaviour of biomass allows us to associate mass loss at specific temperatures with degradation of different compounds present in fruit peel wastes [22].

The revalorization of fruit peel as biosorbent presents some limitations, including low adsorption capacity and release of soluble organic compounds. Some methods have been proposed to overcome these limitations. As previously described, the main physicochemical interactions involved in biosorption processes are ionic exchange or complex formation between metal ions and functional groups of the fruit peel surface. Hence, the improvement of these functional groups entails the enhancement of the biosorption efficiency. Physical and chemical pre-treatments can be proposed to modify the biosorbent surface (Fig 2). Heat treatment, autoclaving, lyophilization, boiling and cutting have been reported as physical methods to increase biosorption capacities. Chemical methods consist of exposing the biosorbent surface to dissolutions of acids, alkali, organic solvents, and other reagents. The modification of raw biomass to enhance their surface reactivity have been reported to be a promising solution of limitations arising in the use of fruit peel as biosorbent. However, as these treatments entail an increase in the operational costs, the economic viability should be carefully evaluated to keep the process costs down. The improvement of adsorption capacity for several fruit peel waste reported in literature is shown in Table 2.

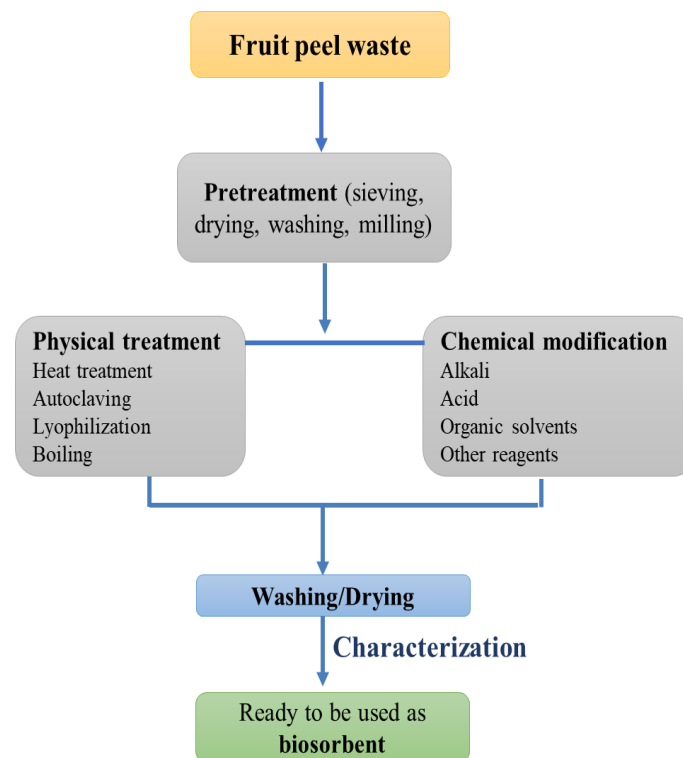


Fig. 2. Fruit peel waste treatments to be applied as a biosorbent.



Table 2. Enhancement of adsorption capacity of fruit peel waste after pre-treatment with different modifiers

Fruit peel waste	Treatment strategy (Chemical method)	Metal	Ratio of adsorption capacities after and before modification	Reference
Lemon	NaOH	Ni	2.62	[13]
Orange	NaOH+CaCl ₂	Cu	79.2	[23], [24]
		Pb	4.2	
		Cd	4.6	
		Ni	16.6	
Banana	NaOH	Pb	19.2	[25]
		Cu	2.4	
Jackfruit	EDTA	Ni	1.4	[26]
		Cr	1.3	
	Sodium dodecyl sulphate	Ni	1.2	
		Cr	1.2	
	NaOH	Ni	1.2	
		Cr	1.3	
	HNO ₃	Ni	1.2	
		Cr	1.2	

IV. BATCH AND COLUMN STUDIES

Biosorption processes are frequently evaluated by batch and continuous modes at laboratory scale. Batch mode, the simplest adsorptive system, is applied to obtain fundamental information to design continuous flow systems which are the most used for industrial applications [13–16,23,27]. Batch experiments are performed to optimize experimental conditions, such as pH, temperature, ionic strength, biosorbent dosage, particle size, initial solution concentration and agitation rate. However, these experiments are not carried out at realistic conditions and do not provide information about biosorption mechanisms[28]. Therefore, column biosorption studies are required to assess the feasibility of a process for industrial application.

The adsorption capacity represents a fundamental property to evaluate the feasibility of a biomass material. This property could be described by biosorption isotherms which are obtained from batch experiments and allows to compare different materials. Table 3 summarizes results of adsorption capacity of several toxic metals on fruit peel wastes. In general, this material presents promising adsorption capacity values for different toxic metals. The selection of suitable biomass would depend not only on the pollutant contained in the water effluent but also on the availability of fruit peel waste.

It should be noted that industrial effluents usually contain a combination of contaminants instead of one single metal. Several factors, including the number of metals, metal combination and their concentration directly affect the biosorption process. Therefore, studies dealing with real mixture of metals effluents are required to describe mechanisms of sorption for designing real applications[29]. Torab-Mostaedi et al. evaluated the binary sorption of Ni and Cd on grapefruit peel obtaining promising results for the treatment of industrial wastewater[16]. Lugo-Lugo et al. proposed the valorisation of orange peel wastes for the biosorption in binary systems containing Fe and Cr. Results of sorbent capacity and adsorption effectiveness showed a significant reduction for both metals in binary systems. However, it was concluded that Cr(III) ions can be removed in the presence of Fe(III) ions which represents a promising result for the use of orange peel as biosorbent [18].

Although batch adsorption systems could be suitable at laboratory scale and for small volumes of wastewater, packed columns are the most practical option for large-scale operations. In fact, according to Shanmugaprakash and Sivahumar, batch experiments are only useful to describe equilibrium and kinetics [30]. Hence, experiments in a continuous reactor should be performed to optimize and predict the performance of fruit peel waste as biosorbent to remove metal ions. Fixed-bed reactor has been widely proposed for wastewater treatment since the contact of biomass with wastewater provides concentration gradients which allows adsorption. The optimization of sorption phenomena in a fixed column is not straightforward due to the influence of relevant aspects, including axial dispersion, intraparticle diffusion

resistance, sorption kinetics and external mass transfer. Hence, mathematical models are required to predict the continuous mode operation and to optimize experimental conditions for continuous mode operation.

Table 3. Adsorption capacity of toxic metals from different fruit peel waste.

Metal	Biomass	Adsorption capacity (mg g ⁻¹)	Reference
As	Pomegranate	As (III) 5.57	[31]
		As (V) 4.50	
Cd	Mango	68.92	[32]
	Orange	63.35	[23,24]
	Ananas	18.21	[33]
	Banana	35.52	[34]
	Grapefruit	42.09	[16]
	Coconut	285.7	[35]
Cr	Banana	6.17	[36]
	Citrus limetta	250	[37]
	Pomelo	0.57	[38]
	Orange	7.14	[39]
	Coconut	8.73	[40]
Cu	Banana	2.433	[25]
	Orange	32.26	[41]
	Pineapple	19.42	[41]
	Mexerica mandarin	131.5	[42]
Hg	Orange	10012	[43]
	Mango	746.62	
	Lime	840.33	
	Banana	0.75	[27]
Ni	Lemon	3.94	[13]
	Orange	9.82	[23,24]
	Pomegranate	52	[44]
	Grapefruit	46.13	[16]
	Lansiumdomesticum	10.10	[45]
Pb	Orange	113.50	[23,24]
	Banana	4.64	[25]
	Citrus	658.9	[46]
	Ponkan	112.1	[21]
	Mango	99.05	[32]
	Muskmelon	167.83	[47]
Zn	Durian	36.73	[15]
	Orange	25	[48]

Chatterjee and Schiewer proposed the use of citrus peels as biosorbent to remove cadmium ions from aqueous solution in a fixed bed column [49]. From breakthrough studies, parameters such as inlet metal concentration, flow rate and bed height were evaluated to optimize the potential valorisation of citrus peel for cadmium removal. Chao et al. studied the use of citrus maxima peel as biosorbents for Cd, Cu, Ni and Pb ions in a fixed-bed column. Experimental results suggested that carboxylic acid groups act as exchangeable cation and complexation sites for removing metal ions. The prediction of adsorption breakthrough curves allowed them to examine the effect of flow rate and pH on the adsorption capacity. The methodology followed in this study offers a description of continuous column behaviour which is required to scale up fixed-bed columns for industrial applications [17]. In general, few studies dealing with the use of fruit peel wastes in fixed bed columns can be found in literature. This fact can be associated with some limitations of this material, such as poor rigidity and clogging and disintegration problems. The immobilization of biomass could overcome these shortcomings by providing rigidity, mechanical strength, and ideal size. Due to their low cost and ease of preparation, the most commonly used immobilization matrices are alginate and agar. Experimental results indicate that the immobilization entails higher capacities and higher diffusion coefficients which significantly improve the sorption process [49].

The regeneration and reuse of biosorbents are key processes for a successful implementation of biosorption as technology.

Desorption mechanisms could be described in a similar way to those of adsorption processes. The regeneration processes could be divided into physical (heating or microwaving) and chemical (addition of acids and other reagents) methods. In general, metal ions have been reported to desorb more than 95% from fruit peel wastes. After 5–6 adsorption/desorption cycles, the combustion of fruit peel biomass allows for taking advantage of its heating value [9,13]

V. CONCLUSION

The use of fruit peel wastes as biosorbent is a promising alternative to remove and recover heavy metal pollutants in wastewater effluents. The valorisation of fruit peel wastes has been reported to be feasible not only for being economic, renewable and eco-friendly but also for the high adsorption capacity of this abundant solid waste. The evaluation of biosorption mechanisms and influencing factors through characterization techniques and batch experiments are required for understanding biosorption phenomena. However, further column biosorption studies are required to optimize and design processes at large scales. Furthermore, additional research dealing with real industrial effluents with a combination of contaminants (not only metals but also organic pollutants) are necessary to demonstrate the potential of biosorption on an industrial scale.

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