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Chapter 4 Biosorption of dyes Jindrayani Nyoo Putroa, Yi-Hsu Jub,c, Felycia Edi Soetaredjod, Shella Permatasari Santosod, Suryadi Ismadjid aDepartment of Chemical Engineering,

8**National Taiwan University of Science and Technology, Taipei City, Taiwan**; bGraduate **Institute of Applied Science and Technology, National Taiwan University of Science and Technology, Taipei City, Taiwan**; cTaiwan **Building Technology Center, National Taiwan University of Science and Technology, Taipei City, Taiwan**

; dDepartment

8**of Chemical Engineering, Widya Mandala Surabaya Catholic University, Surabaya, East Java, Indonesia** 1. Introduction Contamination **of**

the aquatic environment by dyestuffs causes severe prob- lems for humans and aquatic biota. Environmental problems caused by the pollution of dyestuffs become more complicated because most of the dyes discharged into the environment are synthetic dyes. Mostly, these synthetic dyes are designed to be resistant to degradation by microorganisms. Once the synthetic dyes release into the environment, it will resist biological degrada- tion for a long time. Therefore, a treatment process should be conducted before the wastewater containing dyes releases to the water environment. Currently, several processes

3**for the treatment of** the **wastewater containing dyes**

are available. In terms of effectiveness, the

3**adsorption process is the** most suitable **method for** treating **wastewater containing dyes**

, especially at low concentra- tions. The success of treating wastewater containing dyes using

2**the adsorption process is** highly **dependent on the** adsorption capacity **of the** adsorbent used. **The**

use of a variety of environmentally friendly and inexpensive adsor- bents to remove dyes from water or wastewater has been studied by thousands of researchers in the last few decades. Several current review articles discussed the removal of dyes from water or wastewater and are also available in the scientific journal publications (Shamsollahi and Partovinia, 2019; Zhou

34**et al., 2019**; Afroze **and** Sen, **2018**; Anastopoulos **et al**., 2017; Bhatnagar **et al**

., 2010, 2015; Anastopoulos; Kyzas, 2014). These environmentally friendly and inexpensive adsorbents include agricultural wastes and its modified forms, seeds and leaves of various plants, industrial by-products, different kinds of microorganisms (living or dry ones), and natural polymer composites. Green Chemistry and Water Remediation: Research and Applications. https://doi.org/10.1016/B978-0-12-817742-6.00004-9

. 99 The studies of dyes removal using biosorbents have been conducted for more than three decades; however, the biosorption study still very attractive for the scientists indicated by a large number of publications in this area in recent years. Newly modified biosorbents including multifunctional and composite biosorbents have been prepared

17**and used for the** color **removal from aqueous solutions**. Therefore, reviews about **the**

current progress of dyes biosorption are still required. This chapter summarizes and discusses the up-to-date progress of biosorption of dyes using newly modified biosorbents. Various aspects of dyes biosorption are given and comprehensively explained. 2. Classification of synthetic dyes Synthetic dyes are developed to fulfill the human needs of modern and biologically and physically resistant coloring agents. Usually, synthetic

13**dyes are classified according to their application, chemical structure**, ionic forms in **the**

solution, as well as its color (Zhou et al., 2019).

32**Classification of dyes based on** their **chemical structure** possibly is **the**

most appropriate way since it offers many advantages (Gregory, 1990). Based on this method of classification, dyes can be categorized as follow: l Azo dyes: CI disperse yellow 16, CI solvent yellow 14, disperse blue, reactive brown, CI acid black 1, CI direct green 26, etc. l Anthraquinone dyes: quanizarin, CI disperse red 15, CI disperse violet 1, disperse torquise, etc. l Benzodifurano dyes: disperse red. l Polycyclic aromatic carbonyl dyes (anthraquinonoid vat dyes): the blue, indanthrone, CI vat black, the green, the brown, etc. l Indigoid dyes: tyrian purple. l Polymethine and related dyes: cyanine, carbocyanine, azacarbocyanines, etc. l Styryl dyes: malononitrile, CI disperse yellow 31, dicyanovinyl dyes, tet- racyanoethylene, etc. l Di and tri-aryl carbonium and related dyes: diarylcarbonium dyes, mala- chite green, CI basic blue 9, etc. l Phthalocyanines dyes: CI direct blue 86, hemin, etc. l Quinophthalone dyes: CI solvent yellow 33, CI disperse yellow 54, etc. l Sulfur dyes: CI sulfur black 1. l Nitro and nitroso dyes: CI acid yellow 1, CI 10,305, CI disperse yellow 14, CI acid green 1, etc. l Miscellaneous dyes: stilbene dyes, formazan dyes. Details about properties, synthesis, and structures of synthetic dyes can be seen elsewhere (Gregory, 1990). 3. Agricultural and plant wastes as biosorbents A variety of agricultural and plant wastes have been employed as cheap ad- sorbents for the removal of various dyes from aqueous solutions as listed in Table 4.1. Some of the biomass has

3potential application for the industrial- scale wastewater treatment

process, while others possibly will end up only in the laboratory-scale application. The low adsorption capacity of agricultural and plant wastes materials is the main obstacle of using these alternative adsorbents for the industrial-scale treatment of wastewater containing dyes. TABLE 4.1 Adsorption of various dyes using agricultural and plant wastes. Agricultural waste Dyes Adsorption capacity, mg/g Reference Azolla filiculoides Basic orange 833

24at pH 7 and T ¼ 30 C Tan et al

. (2010) Brazil nutshells Methylene blue (MB) and indigo carmine (IC) 7.81 (MB), and 1.09 (IC) Brito et al. (2010) Chia seeds (Salvia hispanica) Reactive yellow B2R 70.95 at pH 2 and T ¼ 30o Da Silva and Pietrobelli (2019) Corn silk

14Reactive blue 19 (RB19) and reactive red 218 (RR218

) 71.6 (RB19) and 63.3 (RR218) at pH 2.0 and T ¼ 25 C Degermenci et al. (2019) Elaeagnus angustifolia L. fruits Methylene blue (MB), and Indigo Carmine (IC) 344.8 (MB) and 9.7 (IC) at room temperature Oymak and Eruygur (2019)

16Glossogyne tenuifolia leaves Congo red (CR) and malachite green

(MG) 13.4 (CR), and 210.9 (MG) at T ¼ 40 C Yang and Hong (2018) Mentha pulegium Direct Red 80 (DR80) and Acid Black 26 (AB26) 52.3 (DR80) and 46.3 (AB26) at pH 2.5 and T ¼ 25 C Mahmoodi et al. (2011) Moringa oleifera seed Reactive red 120 413.32 at T ¼ 50 C and pH 1.0 Celekli et al. (2019) Musa acuminate peel Emerald green 10.75 at T ¼ 25 C Rehman et al. (2019) Olive stone

26Alizarin Red S (ARS), and methylene blue (MB

) 16.10 (ARS), and 13.2 (MB) Albadarin and Mangwandy (2015) Continued TABLE 4.1 Adsorption of various dyes using agricultural and plant wastes.dcont'd Agricultural waste Dyes

18Adsorption capacity, mg/g Reference Orange bagasse Reactive blue 5G

28.9 at T ¼ 40 C Fiorentin et al. (2010)

25Palm date stones Basic violet 3 (BV3), and Basic red 2 (BR2

) 97.8 (BR2) and 117.3 (BV3) at pH 7.7 and T ¼ 35 C Wakkel et al. (2019) Palm kernel fiber Methyl violet 114.2 at T ¼ 66 C Ofomaja et al. (2011) Parthenium hysterophorus L Safranine 89.3 at pH 6 Shrivastava (

212010) Paulownia tomentosa stud leaf powder Acid Orange 52

10.5 at T ¼ 25 C Deniz and Saygideger (

21**2010) Pinus sylvestris L Reactive red 195**

6.69 at 20 C, and 7.38 at 50 C Aksakal and Ucun (2010) Pine tree leaves Basic red 46 71.94 at T ¼ 45 C Deniz and Karaman (2011) Princess tree leaves Basic red 46 43.10 at T ¼ 45 C Deniz and Saygideger (2011)

18**Rice husk Direct red 31** (DR31) and **Direct orange 26**

(DO26) 57.88 (DR31) at pH 2, and 36.14 (DO26) at pH 3, adsorption temperature was 30 C Safa and Bhatti (2011) Sapindus mukorossi dead leaves CI Reactive red 241 8.18 at T ¼ 30 C Javed et al. (2019) Sesame hull Methylene blue 359 at T ¼ 30 C Feng et al. (2011) Solanum tuberosum peel Emerald green 2.61 at T ¼ 25 C Rehman et al. (2019) Soybean hulls BFe4B reactive red 19 at pH 1 and T ¼ 45 C Módenes et al. (2019) Walnut shell Methylene blue 80.4 at T ¼ 45 C Liu et al. (2019) Water hyacinth leaves Amaranth dye 70 at pH ¼ 1.5 and T ¼ 50 C Guerrero-Coronilla et al. (2015) Agricultural and plant wastes biomass are categorized as lignocellulosic materials which are mainly composed of lignins, hemicellulose, and cellulose. Lignins are composed of benzene derivatives such as coumaryl, sinapyl, and coniferyl alcohols, and it has very complex and variable structures. Hemi- celluloses are sugar polymers that consist of C5 and C6 sugar monomers. The polymerization degree in hemicelluloses is lower than celluloses. While cel- luloses are polysaccharides which consist of a linear chain of b-1,4-linked D- glucose monomers. The composition of lignins, hemicellulose, and celluloses in the lignocellulosic materials

33**strongly depends on the type of** biomass, geographic condition, **and** also season. **The**

chemical composition variation of agricultural and plant wastes is another big challenge of using these ligno-cellulosic materials as the industrial biosorbents. Low Brunauere-Emmere-Teller (BET) surface area as well as small pore volume are the characteristics of lignocellulosic materials. With this poor pore structure, natural lignocellulosic materials usually possess small adsorption capacity toward a particular type of dyes (Aksakal and Ucun, 2010; Brito et al., 2010; Deniz and Saygideger, 2010, 2011; Fiorentin et al., 2010; Albadarin and Mangwandy, 2015; Yang and Hong, 2018; Javed

9**et al., 2019**; Módenes **et al., 2019**; Rehman **et al., 2019**

). Typical nitrogen sorption isotherm of agricultural waste material (cassava peel) is given in Fig. 4.1. However, in some cases, the excellent adsorption capacity was observed for some natural lignocellulosic materials (Tan et al., 2010; Celekli et al., 2019; 7 6 Volume adsorbed, cm3/g STP 5 4 3 2 1 Adsorption Desorption 0 0.0 0.2 0.4 0.6 0.8 1.0 Relative pressure, p/po FIG. 4.1 Sorption isotherms of nitrogen onto cassava peel waste. Oymak and Eruygur, 2019). The natural lignocellulosic materials have high adsorption capacity possible due to the excess presence of polar functional group-containing compounds (Lesmana et al., 2009). In the adsorption of dyes using lignocellulosic materials, particular surface functional groups of adsor- bent play an essential role. These functional groups are likely responsible for binding with dyes molecules during the adsorption process. Several polar and nonpolar functional groups which usually exist in the agricultural waste materials are as follow: O e H stretch (alcohols or phenols), N e H stretch (1 and 2

amines), C e H stretch (methylenes), C ¼ O stretch (carboxylic acids, saturated aliphatic esters, carboxyls), C ¼ C stretch (al- kynes), and C e H bend (aromatic compounds). The existence of surface functional groups on the agricultural waste usually is obtained using a Fourier Transform Infra-Red (FTIR) method. In the case of the adsorption

16**of reactive red 120 on Moringa oleifera seed**, the carbonyl, amines, **and**

amide groups play essential roles in the binding of the dye (Çelekli et al., 2019). 4. Thermal modification of agricultural wastes To increase the adsorption capacity of some agricultural waste toward dyes molecules, the modification at high temperatures is one of the best options. During the thermal treatment, the lignocellulosic materials will be decom- posed into some low molecular weight compounds (gases and tar) and leaving solid carbonaceous material known as biochar or the black product. The resulting biochar possesses a porous structure; however, in many cases, the direct use of this solid material as the adsorbent often gives unsatisfactory results. Activation using some chemicals (known as chemical activation) or oxidizing gases (physical activation) usually is conducted to increase

7**the adsorption capacity of the** biochar/**activated carbon (Table**

4.2). Thermal modification of lignocellulosic materials (physical or chemical) produces porous materials known as activated carbons or activated biochars. Thermal modification using chemicals as the activating agent offers several advantages than the physical one (Sudaryanto et al., 2006), such as l l Lower temperature, usually between 400 and 600 C. The single-step process, the activation, and the carbonization process take place in one process. l The chemical activation process produces a higher yield of solid than physical activation. The main drawback of the modification of lignocellulosic materials using chemical agents is often, it produces hazardous wastewater (depends on the chemical agents used), which creates another environmental problem. The physical activation is known as an environmentally friendly process since it only utilizes steam or carbon dioxide as the activating agent. TABLE 4.2 Thermal process of some agricultural and plant wastes. Agricultural waste Almond shell Coconut shell Corncob Corn stigmata Foxtail palm Mangosteen peel Olive tree pruning and coffee husk Peanut shell Pineapple crown leaf Pinecone Pinecone Prickly pear seeds and cactus Shiitake mushroom Thermal process Microwave-assisted process Thermochemical activation process using ZnCl2 at 700 C Steam activation at 892 C for 40 min Hydrothermal carbonization at 180 C followed by CO2 activation at 900 C Vacuum pyrolysis at 400 e700 C and activated using H2SO4, H3PO4, and KOH ZnCl2 activation at 600 C Chemical activation at 500 C (K2CO3 and KOH), and physical activation at 910 C (steam) Carbonized at 800 C in the presence of FeCl3 in CO2 environment Carbonization at 500 C followed by chemical activation (KOH) using microwave irradiation Pyrolized at 500 C and activated using ZnCl2 Microwave-induced ZnCl2 activation Activated using H3PO4 at the temperature of 450 C Activated using K2CO3 and carbonized at 800 C Application Methylene blue adsorption

4**Adsorption of methylene blue**, malachite green, **and** methyl **orange**

20**Methylene blue** adsorption **Methylene blue** adsorption **Methylene blue** adsorption **Methylene blue** adsorption **Methylene blue**

adsorption Malachite green adsorption Methyl violet adsorption Alizarin red S adsorption

4**Methylene blue** adsorption **Methylene blue** adsorption **Methylene blue**

adsorption Reference Du et al. (2016) Gupta and Khatri (2019) Yu

11**et al. (2019**) Mbarki **et al. (2019**) Dos Santos **et al**. (2019) Nasrullah **et al**. (2019) Mamani **et al**. (2019) Guo **et al**. (2018) Astuti **et al**. (2019) Bhomic **et al**. (2018) Ozhan **et al**. (2014) Ouhammou **et al**. (2019) Sun **et al**

. (2019) Continued TABLE 4.2 Thermal process of some agricultural and plant wastes.dcont'd Agricultural waste sunflower seed husk Walnut shell Water fern Thermal process ZnCl2

17**was used as** a **chemical activating agent**, and **the**

carbonization was conducted at 500 C using microwave irradiation as the heat source Activated with phosphoric acid and carbonized at 450 C Carbonized at 300 C, and activated using NaOH and KOH and followed by activation at 600 C under argon atmosphere for 3 h Application Methylene blue adsorption Malachite green adsorption Acid orange II adsorption Reference Baytar et al. (2018) Hajialigol and Masoum (2019) Emrooz et al. (2019) In the thermal activation process, several steps of processes occurred during the heating process that leads to the pore creation. The first step of the methods is the release of free moisture content and bound water from the structure of lignocellulosic material. The evaporation of both free and bound water occurs at a temperature of around 50e200 C. In the subsequent step of the processes, which occur at a temperature range of 200e300 C, the hemicellulose will disappear due to the decomposition. At higher temperatures (300e360 C), the cellulose decomposes into the smaller molecular weight of organic compounds, and the last part of organic substance in the lignocellu- losic material, lignin, will decompose at a temperature of 360e500 C. The degradation or decomposition of high molecular weight compounds into the smaller ones of the structure of lignocellulosic material will create space within the internal structure of the material. The creation of space in the in- ternal structure of materials will increase the active surface area of the ma- terials and later will enhance its adsorption performance. Chemical and physical activation processes were employed by Mamani et al. (2019) to prepare the activated carbons from the coffee husk and olive tree pruning. The impregnation of the biomasses was conducted at 130 C using potassium hydroxide (KOH) and potassium carbonate (K2CO3) as chemical activating agents, and the activation was performed at 800 C. For physical activation, the carbonization was conducted at 500 C and the gasi- fication at 910 C using superheated steam as the activating agent. The chemical activation produced the carbon with a surface area over 2500 m2/g, while for the physical process created carbon with BET surface area around 1500 m2/g. The resulting carbons could remove more than 99%

14**of methylene blue from aqueous solution**

. 5. Combination of agricultural wastes and other lignocellulosic materials with other materials to form composites To enhance the

14**adsorption capacity of the** activated carbon **derived from the** agricultural waste, **the**

combination with other materials was usually performed to produce new composite materials which have high adsorption performance or capacity. Naushad et al. (2019) developed arginine-modifiedeactivated carbon (AGDPA@AC) for removal of methylene blue from aqueous solution. The maximum uptake of methylene blue by the AGDPA@AC was achieved

22**at pH 8 and temperature of 25 C**

(219.9 mg/g). The time required to reach the equilibrium condition was 120 min. The combination of macroalgae Saccharina japonica (which is also known as kelp) with montmorillonite to form the composite of biocharebentonite was studied by Sewu et al. (2019). The composite

3**was utilized as the adsorbent for the removal of an**

32**anionic dye (Congo Red) and cationic dye**

(crystal violet). The incorporation of bentonite into biochar increased its mesopores structure, and it enhanced the adsorption performance of the composite material toward the Congo Red and crystal violet (at 30 C). 6. Algae as biosorbents Many research groups around the world have conducted

2**studies on** the **removal of dyes from** water or **wastewater using**

microalgae as well as macroalgae for many years. Their adsorption capability and its abundant availability as diverse groups of plant-like organisms which can be found easily as either in multi- cellular or unicellular forms make these organisms very attractive as the alternative low-cost biosorbents (Table 4.3). Sargassum dentifolium and Ulva fasciata (macroalgae species) have been tested for their adsorption performance to remove methylene blue from aqueous solution by Moghazy et al. (2019). There is high removal efficiency for both macroalgae due to the presence of various surface functional groups that possess high adsorption affinity toward methylene blue molecule. Different adsorption mechanisms

7**between the surface functional groups of**

macroalgae with methylene blue cation occurred during the adsorption pro- cess, such as ionic interaction, coordination, ion-dipole bond, and van der Waals interaction (Moghazy et al., 2019). Brown macroalgae Stoechospermum marginatum was utilized to recover

7**Acid Blue 25, Acid Orange 7, and Acid Black 1** (acid dyes) **from** the **aqueous**

TABLE 4.3 Adsorption of dyes using algae as biosorbents. Algae Chlorella pyrenoidosa Chlorella vulgaris Enteromorpha prolifera Fucus vesiculosus Laminaria japonica Dye Direct Red-31 Methylene blue Rhodamine B Methylene blue Supranol Red 3BW, Lanaset Red 2 GA, and Levafix Navy Blue EBNA Direct Fast Scarlet 4BS Methylene blue Methylene blue Parameters pH: 3e14; contact time: 0e180 min; initial dye concentration: 10e50 mg/L pH: 6; temperature: 28 C; time: 24 h; initial concentration of dye: 100 mg/L pH: 2e10; temperatures: 25, 35, 45 C; contact time: 5e240 min; initial dye concentration: 100 mg/L Modified with H3PO4 and ZnCl2; pH: 6; temperature: 28 C; adsorbent dose: 2 g/L pH: 3.85e11.40; temperature: 35e58 C

pH: 0.5e10; temperature: 30e50 C; time: 5e540 min Modified with H3PO4 and ZnCl2; pH: 6; temperature: 28 C; adsorbent dose: 2 g/L pH: 2e10; temperature: 25 C; time: 0 e60 min; adsorbent dose: 0.2e2.0 g/L Adsorption capacity, mg/g 30.53 101.75 63.14 212 256.4 (Supranol Red 3BW), 345 (Lanaset Red 2 GA), and 188.7 (Levafix Navy Blue EBNA) 318.87 1162.9 549.45 Reference Sinha

> 6**et al. (2016**) Lebron **et al**. (2018) Da Rosa **et al. (2018**) Lebron **et al**. (2019) Lim **et al**. (2010) Sun **et al**. (2019) Lebron **et al. (2019**) Shao **et al**

. (2017) 108

> 2**Green Chemistry and Water Remediation: Research and Applications**

Sargassum dentifolium Spirulina maxima Stoechospermum marginatum Ulothrix sp. Ulva fasciata

> 7**Methylene blue Methylene blue Methylene blue**

> 7**Acid Blue 25, Acid Orange 7, and Acid Black 1**

Methylene blue Methylene blue pH: 3e9; temperature: 20e50 C;

> 19**biosorbent dose: 0**.25e3 **g/L; contact time**: 0e90 **min**; initial concentration **of**

dye: 20e80 mg/L pH: 6; temperature: 28 C; time: 24 h;

> 33**initial concentration of dye: 100 mg/L**

Modified with H3PO4 and ZnCl2; pH: 6; temperature: 28 C; adsorbent dose: 2 g/L pH: 2e10; temperature 27 C; initial dye concentration: 10e50 mg/L pH: 1.3e10.4; temperature: 20e60 C;

> 19**biosorbent dose: 0**.5e5 **g/L; contact time**

: 0.5e12 h. pH: 3e9; temperature: 20e50 C;

> 19**biosorbent dose: 0**.25e3 **g/L; contact time**: 0e90 **min**; initial concentration **of**

dye: 20e450 mg/L 66.6 145.34 343.66 22.2 (

> 12**Acid Blue 25**); 6.73 (**Acid Orange 7), and** 6.57 (**Acid Black 1**

) 86.1 244 Moghazy

10et al. (2019) Lebron et al. (2018) Lebron et al. (2019) Daneshvar et al. (2012) Dogar et al. (2010) Moghazy et al. (2019) Biosorption of

dyes Chapter | 4 109 solution by Daneshvar et al. (2012). Various operation parameters (pH, tem- perature, initial concentration of acid dyes, the particle size of biosorbent, and

7contact time) influence the uptake of acid dyes by

Stoechospermum margin- atum.

7The maximum adsorption capacities of the

brown macroalgae were 6.57.6.73, and 22.

72 mg/g for Acid Black 1, Acid Orange 7, and Acid Blue

25. The low adsorption capacity of brown macroalgae Stoechospermum margin- atum toward acid dyes is due to the lack of internal micropore and mesopore structures in brown macroalgae. The adsorption of acid dyes solely occurred in the macropore structure and several surface functional groups available on the surface of the macroalgae. The FTIR analysis reveals that the amine and hydroxyl groups are mainly responsible for the uptake of the acid dyes. Ulothrix species is a filamentous green alga which is widely found in both marine and freshwater. This species of algae had been studied for its possi- bility as an alternative adsorbent for

12methylene blue removal from aqueous solution (Dogar et al., 2010). The adsorption capability of

the Ulothrix species toward methylene blue was strongly influenced by the

4pH of the solution and temperature of the system. The adsorption capacity of

this filamentous green algae

1increased with the increased of the pH

of the

1solution. The experimental adsorption results indicate that the

3temperature had a negative effect on the amount of methylene blue adsorbed by this biosorbent. The

amount of methylene blue adsorbed by the algae decreased with increasing temperature. This phenomenon suggests that physical adsorption is the primary mechanism that controls the adsorption of methylene blue on Ulothrix species. 7. Fungus/fungi as biosorbents Fungus/fungi have been utilized as the alternative low-cost biosorbents

13for the removal of dyes from the water environment by a large number of

research groups. The capability of fungal biomass in decolorizing of large numbers of dyes has been discussed by

23Sen et al. (2016). The combination of enzyme degradation and adsorption is the

primary mechanism

13in the decolorization of dyes by fungal

biomass (Sen

13et al., 2016). The ability of the fungus to degrade

complex synthetic dyes is mostly due to the availability of extracellular lig- ninolytic enzymes in the fungus itself. Several of the studies on the utilization of the fungus or fungi to decolorize dyes from the solutions are listed in Table 4.4. The modification of fungus with chemicals is usually conducted to increase its adsorption capacity. The modification of Aspergillus versicolor using CTAB was studied by Huang et al. (2016) to increase its adsorption capability toward Reactive Black 5. The highest removal efficiency of Reactive Black 5 (>98%) by surfactant-modified A. versicolor (1.5% CTAB) was achieved at pH 2.0 and

34contact time of 420 min. The high removal efficiency of

surfactant-modified A. versicolor is due to the increased active sites

18of surface functional groups as well as the

increased BET surface area. At an acidic pH TABLE 4.4 Adsorption of dyes using fungus/fungi as biosorbents. Fungus/fungi Agaricus bisporus Aspergillus lentulus FJ172995 Aspergillus versicolor Ceriporia lacerata P2 Coriolopsis sp. Cunninghamella elegans Dye Reactive Red 2

23Acid Navy Blue, Orange HF, Fast Red A, Acid Sulfone Blue, and Acid Magenta

Reactive Black 5 Crystal Violet

15**Crystal Violet, Methyl Violet, Cotton Blue, Malachite Green**

Acid Blue 62, Acid Red 266 and Acid Yellow 49 Parameters The fungal was modified by CTAB (cetyl trimethyl ammonium bromide). pH: 2.0e8.0,

28**temperature: 25, 35**, 45 **C; contact time**: 5e90 **min**

. pH: 4.0e10.

28**0; initial dye concentration**: 100e900 **mg/L**

; temperature: 30, 35, 40, 45 C The fungal was modified by CTAB; pH: 2.0e8.0; temperature: 30 C; adsorption time: 0e24 h pH: 2.0e10.0; initial concentration: 10e500 mg/L;

25**temperature: 20 C Initial dye concentration: 50**, 100, and 200 **mg/L**

;

28**temperature: 30 C Contact time**: 0e125 **min**

; temperature: 30 C; pH: 3.0e11.0 Adsorption capacity, mg/g 141.53 212.92 227.27 239.25 n.a 1035 Reference Akar and Divriklioglu (2010) Kaushik and Malik. (2010) Huang et al. (2016) Lin

15**et al. (2011**) Chen **and Ting** (2015a) Russo **et al**

. (2010) Continued Biosorption of dyes Chapter | 4 111 TABLE 4.4 Adsorption of dyes using fungus/fungi as biosorbents.dcont'd Adsorption capacity, Fungus/fungi Dye Parameters mg/g Reference Lenzites elegans Crystal Violet, Malachite Green, pH: 7.0; temperature: 30 C; time: 0e72 h. n.a Pandey et al. (2018) WDP2 Fuschsin Basic, and Brilliant Green Mucor circinelloides Congo Red Temperature: 25, 50, 75 C; initial concentration of dye: 150, 300, 600, 800, 1000 mg/L Penicillum restrictum Reactive Yellow 145 pH: 1e8; temperature: 20, 30, and 40 C Rhizopus arrhizus Methylene blue Modified with SDS (sodium dodecylsulfate) surfactant. pH: 2e12; SDS concentration: 0e20 mM; temperature: 25 C; initial concentration of MB: 25e1100 mg/L 169.49 Azin and Moghimi (2018) 116.5 Caner et al. (2011) 1666.6 Aksu et al. (2010) Biosorption of dyes Chapter | 4 113 (2.0), the protonation of several

26**functional groups such as** carboxyl, **hydroxyl**, amide, **and amine groups** in **the surface of**

surfactant-modified A. versicolor occurred. The surface of biosorbent became positively charged, and electro- static interaction between the surface of biosorbent and negatively charged (SO?3) ions of Reactive Black 5 occurred. This phenomenon significantly enhanced the uptake of the anionic dye from the aqueous solution (Huang et al., 2016). 8. Bacteria as biosorbents As microscale organisms, bacteria also have been widely

> 3**explored for its potential** application **as** low-cost **adsorbents for** decolorization of **wastewater**

containing dyes. Several recent studies on the utilization of bacteria as bio- sorbents for dyes removal from an aqueous environment are summarized in Table 4.5. The activity and adaptability of bacteria in the wastewater treatment system strongly influence the effectiveness of bacteria in the decolorization process (Meerbergen et al., 2018). Bacteria possess different decolorization mecha- nisms compared with other conventional biosorbents such as fungus, algae, or other lignocellulosic biomass. In the conventional biosorbents, the removal of color or decolorization process is mainly due to the adsorption process, so basically, the decolorization process using conventional biosorbents will be effective if the concentration of dye in the solution or wastewater is quite low. In the decolorization process using bacteria as the biosorbent, the main decolorization mechanism is degradation rather than adsorption; therefore, this process is likely very effective for wastewater containing a high concentration of dyes (Meerbergen et al., 2018). 9. Equilibrium studies Various adsorption isotherm equations which were initially

> 1**developed for the gas-phase adsorption** are employed **to represent the adsorption equilibria** data **of**

various dyes by different kinds of biosorbents. Those equations are either two-parameter models (Langmuir, Freundlich, DubininеRadushkevich (DR), Temkin, FloryeHuggins, BET) or three-parameter models (Sips, Toth, and RedlichePaterson). Among them,

> 3**Langmuir and Freundlich are the most widely used** isotherm **equations to** represent **the adsorption**

equilibria data due to their simplicity. Langmuir

> 1**equation has the form as follows**

: qe ¼ qmax1 þKLKCLeCe (4.1) Parameters qmax and KL represent the adsorption capacity of biosorbent toward specific dye pollutants, and KL is the adsorption affinity. In most cases, TABLE 4.5 Adsorption of dyes using bacteria as biosorbents. Bacteria Acinetobacter (ST16.16/164) Klebsiella (ST16.16/ 034) Penicillium simplicissimum Pseudomonas aeruginosa

> 22**Dyes Reactive Orange 16 and Reactive Green 19**

Reactive Orange 16 and Reactive Green 19 Methyl Violet, Crystal Violet, and Cotton Blue

16**Reactive Red 21, Reactive Orange 16, and Reactive Blue 19**

Notes The bacteria were

22**isolated from activated sludge** of two **wastewater treatment**

plants located in Flanders, Belgium. The influence of

15**carbon sources, pH, temperature, dye concentration, and**

concentration of salt addition on the decolorization capability of Acinetobacter was explored. The incubation was conducted for 3 days. The decolorization efficiency of Acinetobacter toward both of azo dyes was > 80%. The bacteria were isolated from activated sludge of two wastewater treatment plants located in Flanders, Belgium. The influence of

15**carbon sources, pH, temperature, dye concentration, and**

concentration of salt addition on the decolorization capability of Klebsiella was explored. The incubation was conducted for 3 days. The decolorization efficiency of Klebsiella toward both of azo dyes was > 80%. The bacteria were isolated from wastewater at Monash Uni Malaysia. The decolorization process was conducted at 30 C for 14 days with initial dyes concentration of 50, 100, and 200 mg/L. The removal efficiencies were 98% for Methyl Violet, 95% for Crystal Violet, and 82% for Cotton Blue. Jack fruit seed powder was employed as cosubstrate.

24**Response Surface Methodology (RSM**) was used **to** evaluate **the** influence **of** different process **parameters on the decolorization** performance **of**

Pseudomonas aeruginosa. The bacteria were capable to remove 97.7 0.3 % of Reactive Red 21, 98.9 0.3 % of Reactive Orange 16, and 92.6 0.4 % of Reactive Blue 19. Reference Meerbergen et al. (2018) Meerbergen et al. (2018) Chen and Ting (2015b) Mishra et al. (2019) Biosorption of dyes Chapter | 4 115 both of these parameters are temperature dependent. For physical adsorption (exothermic process), the values of parameters qmax and KL

4**decrease with the increase of** the **temperature** of **the** system. While **the**

opposite behavior is observed for chemical adsorption (endothermic process). Freundlich adsorption isotherm has the mathematical expression as follows: qe ¼ Kf Ce1=n (4.2) The parameters Kf and n represent Freundlich adsorption capacity and heterogeneity of the system, respectively. Usually, the

3**value of parameter n is between 1 and 10. The** higher the **value of** parameter **n**

, the more heteroge- neous the adsorption system. Although it does not have the saturation capacity in high concentration and

1**Henry's law at very low concentration, the**

Freundlich

1**equation could represent the adsorption** equilibria **data of**

many systems very well, as indicated in Table 4.6. In all cases, the adsorption ex- periments were conducted at low to moderate dye concentrations, and at these ranges of concentrations, the drawback of the Freundlich model does not exist; therefore, this adsorption equation could describe the experimental data well (Febrianto et al., 2009). Another adsorption isotherm model, which was initially developed for the adsorption of gas onto microporous adsorbents, DR equation, also widely employed to correlate the adsorption of dyes onto biosorbents (nonporous materials). The DR equation can be expressed in mathematic form as follows: 2 ! qe ¼ qmaxexp ? RTlnaðEcoe=csÞ (4.3) The parameter a is

1**an affinity coefficient** that **is proportional to the molar volume**

of the adsorbate. The theoretical value of a can be estimated based on the refractive index of the adsorbate (Ismadji and Bhatia, 2001). Symbol Cs indicates the solubility of the solute in the solvent. The parameter Eo repre- sents solid characteristic energy (usually compared to benzene, which is generally chosen as the reference compound). One of the unique features of the DR equation is in the characteristic curves.

4**If the DR equation** could **represent the experimental data, all of the**

1**adsorption data at different** tem- peratures when **plotted as the** logarithmic **amount adsorbed** (qe) **versus the square of potential energy**

(RTln(Ce/Cs)) should lie on the same straight line. However, in all of the studies which were employed, the DR equation failed to provide this characteristic curve, the judgment of the validity of the

1**model to represent the experimental** adsorption **data** is **only based on the value of**

the R2 (Daneshvar et al., 2012; Ari and Celik, 2013; Lebron et al., 2018; Deniz and TABLE 4.6 Adsorption equations

1**to represent the equilibria data of the adsorption**

of dye onto biosorbent. Biosorbent Dye Isotherms used Isotherm valid Aspergillus niger Acid Blue 161 and Langmuir and Freundlich Freundlich Aspergillus terreus Aspergillus versicolor Banana trunke activated

carbon Carpinus betulus sawdust Cashew nutshelle based carbons Ceriporia lacerata P2 Chlorella pyrenoidosa Chlorella vulgaris Corn silk Procion Red MX-5B

30**Acid Blue 161 and Procion Red MX-5B**

Reactive black 5 Methylene blue Basic red 46 Methylene blue Crystal violet Methylene blue Direct Red-31 Supranol Red 3BW Reactive Blue 19, and Reactive Red 218

5**Langmuir and Freundlich Langmuir and Freundlich Langmuir, Freundlich**

, DR, Temkin, Frumkin, Harkins-Jura, and Smith Langmuir, Freundlich, and DR

5**Langmuir and Freundlich Langmuir, Freundlich**, Koble-Corrigan, **and** Scatchard **Langmuir, Freundlich**

, DR, Temkin

5**Langmuir and Freundlich Langmuir and Freundlich Langmuir, Freundlich**, DR, Temkin, Halsey, **and** Harkins-Jura **Freundlich Langmuir Freundlich**

Freundlich and Langmuir Freundlich and Langmuir Koble-Corrigan

5**Langmuir Langmuir and Freundlich Langmuir and Freundlich** Freundlich **and** Temkin References Almeida **and** Corso (2019) Almeida **and**

Corso (2019) Huang et al. (2016) Danish

6**et al. (2018**) Deniz **and** Yildis (**2019**) Spagnoli **et al**. (2017) Lin **et al**. (2011) Lebron **et al. (2018**) Sinha **et al**. (2016) Lim **et al**. (2010) Degermenci **et al**

. (2019) Date palm Eucalyptus Furfural lignocellulosic waste Kefir grains Laminaria japonica Macroalga Stoechospermum marginatum Microalgae Scenedesmus Mucor circinelloides Neonectria radicicola Methylene blue

29**Methylene blue Methyl Orange and Rhodamine B**

Remazol navy Methylene blue

7**Acid Blue 25, Acid Orange 7, Acid Black 1**

Methylene blue Congo red Acid Orange 51, Reactive Red 75, and Direct Blue 86 Langmuir, Freundlich, and DR

> 5**Freundlich Langmuir**, Freundlich, **and** DR **Freundlich Langmuir and Freundlich Langmuir**, Freundlich, **and** Temkin **Langmuir** (Methyl orange), **and Freundlich**

(Rhodamine B) Temkin and

> 5**Langmuir Langmuir and** Sips **Langmuir** Langmuir, **Freundlich**, DR, **and** Temkin **Freundlich Langmuir** Langmuir Langmuir **and Freundlich** Freundlich **Langmuir, Freundlich**

, RedlichePeterson, Toth, and Temkin Freundlich and Temkin Esmaeili and Foroutan (2019) Esmaeili and Foroutan (2019) Chen

> 31**et al. (2019**) Erdogdular **and** Apar (**2019**) Shao **et al**. (2017) Daneshvar **et al**

. (2012) Afshariani and Roosta (2019)

> 31**Azin and Moghimi (2018**) Ghariani **et al. (2019**

) Biosorption of dyes Chapter | 4 Continued 117 TABLE 4.6 Adsorption equations

> 1**to represent the equilibria data of the adsorption**

of dye onto biosorbent.dcont'd Biosorbent Dye Isotherms used Isotherm valid References Neurospora sitophila Reactive blue 49 Langmuir and Freundlich Langmuir Akar and Celik (2011) cellseZeamays silk tissue biomass system Olive stone by- product Pomegranate peel Prunus dulcis leaves Pyracantha coccinea Rhizopus oligosporus Saccharomyces cerevisiae Sargassum dentifolium Alizarin red and methylene blue Malachite green Acid Blue 113 Orange G

> 30**Acid Blue 161 and Procion Red MX-5B**

Brilliant green and methylene blue Methylene blue

> 9**Langmuir, Freundlich**, RedlichePeterson, **and Temkin Langmuir, Freundlich**

, Temkin, DR, and RedlichePeterson

5**Langmuir and Freundlich Langmuir, Freundlich**, DR **Langmuir and Freundlich Langmuir and Freundlich Langmuir and Freundlich**

RedlichePeterson Langmuir Langmuir Langmuir Freundlich Freundlich Freundlich Albadarin and Mangwandi (2015) Gunduz and Bayrak (2017) Jain and Gogate (2019) Ari and Celik (2013) Almeida and Corso (2019) Ghaedi et al. (2013) Moghazy et al. (2019) Sargassum muticum Sesame hull Sesame straw Sour lemon sawdust soybean hulls Spirulina maxima Swede rape straw Trichoderma harzianum Ulva fasciata Walnut shell Walnut shell Water hyacinth

20**Methylene blue Methylene blue Methylene blue Methylene blue** Reactive Red 195 **Methylene blue Methylene blue** Reactive black B **Methylene blue**

Malachite green Methyl blue and Congo Red Amaranth anionic dye Langmuir, Freundlich, DR, Temkin

9**Langmuir, Freundlich, and Temkin** Langmuir **and** Freundlich **Langmuir, Freundlich**

, and DR Langmuir, Freundlich, Toth, Sips, Radkee Praustnitz, Temkin, and BET Langmuir, Freundlich, DR, Temkin

9**Langmuir, Freundlich, and Temkin Langmuir, Freundlich**

, and Temkin

5**Langmuir and Freundlich Langmuir**, Temkin, **Freundlich, and** Dubinine KaganereRadushkevich (DKR) **Langmuir** Langmuir, **Freundlich**

, Temkin, Halsey, DR, Sips, RedlichePeterson, Radke- Prausnitz, Toth DR and Temkin

5**Langmuir Langmuir Freundlich Langmuir**, BET, **and** Toth **Freundlich and** Temkin **Langmuir Freundlich** Freundlich **Langmuir**

Langmuir Langmuir, Sips, Redliche Petersen El Atouani

6**et al. (2019**) Feng **et al**. (2011) Feng **et al**. (2017) Esmaeili **and** Foroutan (**2019**) Módenes **et al**. (2019) Lebron **et al**. (2018) Feng **et al**. (2013) Karthik **et al. (2019**) Moghazy **et al**

. (2019) Hajialigol and Masoum (2019) Liu et al. (2019) Guerrero-Coronilla et al. (2015) Biosorption of dyes Chapter | 4 119 120

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FIG. 4.2 The characteristic curve of the adsorption of methyl violet onto jackfruit peel waste activated carbon. Yildis, 2019; Degermenci et al., 2019; El Atouani et al., 2019; Esmaeili and Foroutan, 2019). The characteristic curve of the adsorption of methyl violet onto jackfruit peel waste-activated carbon is given in Fig. 4.2. Based on Fig. 4.2, it is evident that the DR equation can represent the equilibria data of the adsorption of methyl violet onto jackfruit peel wastee activated carbon well. As mentioned previously that the

3**DR equation was developed for the adsorption**

of gas onto porous solids,

1**therefore, this model is** mostly applicable **to represent the adsorption** equilibria **data**

of various sub- stances onto porous materials such as activated carbons. Temkin equation is also a popular model

1**to represent the adsorption of** dyes onto **various**

biomass systems. Similar to the previous adsorption isotherm models, the Temkin model was also developed for the gas-phase adsorption (adsorption of H2 onto Pt electrodes). The mathematic expression of Temkin equation is as follows: qe ¼ RT lnðKT CeÞ (4.4) bT The parameter bT represents the heat of adsorption, while the binding energy of the adsorption at equilibrium condition is given by symbol KT. Temkin equation is also frequently used by many researchers to correlate the Biosorption of dyes Chapter | 4 121 biosorption of dyes onto biosorbents such as adsorption of methylene blue on banana trunkeactivated carbon (Danish et al., 2018), adsorption of methylene blue on Chlorella pyrenoidosa (Lebron et al., 2018), adsorption of acid dyes

12**onto brown** macroalgae **Stoechospermum marginatum (Daneshvar et al., 2012), adsorption**

14**of Reactive Blue 19, and Reactive Red 218**

on corn silk (Degermenci et al., 2019), adsorption of Remazol navy on kefir grains (Erdogdular and Apar, 2019), adsorption of Acid Orange 51, Reactive Red 75, and Direct Blue 86 on Neonectria radicicola (Ghariani et al., 2019), etc. In several systems, this equation gave better performance than other adsorption equations in correlating the experimental adsorption data of dyes onto bio- sorbents (Lebron et al., 2018; Degermenci et al., 2019; El Atouani et al., 2019; Ghariani et al., 2019). In general, the adsorption of dyes onto biosorbents involvs very complex phenomena; several factors such as the pH of the so- lution, solubility of dyes in water, and the surface chemistry of the biosorbents play crucial roles on the uptake of dye molecules by biosorbents. Since the development of the Temkin adsorption equation is only based on

straightfor- ward assumptions, the complex phenomena of dyes adsorption by the bio- sorbents cannot be captured by this equation, and therefore, in most

> 1**cases, this equation** fails **to represent the adsorption equilibria data**. Compared with **the** other two parameter **adsorption**

equations, the Halsey isotherm is seldom used

> 1**to represent the** equilibrium **data of the adsorption**

of dyes onto biosorbents. Halsey isotherm was derived according to the condensation of gases in a multilayer condition (the distance relatively far from the surface of the solid). The Halsey isotherm

> 27**equation can be written as follows: qe** ¼ KH 1=nH (**4**

.5) Ce where KH and nH are the parameters of the Halsey isotherm equation.

> 4**This equation** fails **to represent the adsorption of** Amaranth anionic dye **onto**

water hyacinth (Guerrero-Coronilla et al., 2015). Similar to Halsey isotherm equation, the BET isotherm is also rarely employed

> 1**to correlate the adsorption data of** dyes **onto** biosorbents. **The**

BET model for liquid-phase adsorption has the form as follows: qe ¼ qmax?Ce ? Cs ½1 þBCðBe ? 1ÞðCe=CsÞŠ (4.6) The development of the BET model used a similar kinetic concept of Langmuir isotherm (rate of adsorption ¼ rate of desorption). Unlike the Langmuir equation, the restriction that one adsorption site can be occupied by one molecule of adsorbate has been omitted in the BET model. The BET model has been successfully employed to correlate the adsorption data of Reactive Red 195 on the soybean hull (Módenes et al., 2019). In several cases, the available two-parameter adsorption equations could not

> 1**represent the adsorption equilibria data well, and for**

this reason, three-parameter adsorption isotherms were developed. Sips isotherm is the three-parameter model which

> 4**was developed to overcome the** drawback **of** the **Freundlich equation**; this **equation**

possesses saturation capacity at relatively high concentration of the adsorbate. qe qmax1 þ ðKsCeÞn ¼ ðKsCeÞn (4.7) Similar to Freundlich, the parameter n in the Sips equation also

4represents the heterogeneity of the adsorption system. For the homogeneous system, the parameter n

in Sips equation is close to unity, and it will become the Langmuir equation. As a three-parameter equation, the Sips equation is more superior in rep- resenting the adsorption equilibria than other two-parameter equations. Sips equation could represent the adsorption equilibria for many systems such as water hyacinthdAmaranth anionic dye (Guerrero-Coronilla et al., 2015), acid-treated coffee huskeMalachite Green (Murthy et al., 2019), Eugenia umbelliflora BergdMethylene Blue (Postai and Rodrigues, 2018), Anethum graveolensdMethylene Blue (Hamitouche et al., 2017), BeechwoodeDirect Red (Muntean et al., 2017), etc. Since the Sips equation also

1does not have a correct Henry's law at very low concentration

, therefore, this equation mostly

17fails to represent the adsorption data at low concentration. Toth equation

is another empirical three-parameter adsorption isotherm, this equation is able to represent many adsorption equilibria of heterogeneous systems. qe ¼ qmaxð1 þ ðKKTTCCeeÞnÞ1=n (4.8) Toth

1equation has a correct Henry law for very low concentration

. Being an empirical three-parameter equation, Toth should be able

3to describe the experimental data for most complex adsorption systems

. However,

1this equation failed to represent the adsorption equilibria of

particular systems such as water hyacinthdAmaranth anionic dye (Guerrero-Coronilla et al., 2015), Neonectria radicicoladAcid Orange 51, Neonectria radicicoladReactive Red 75, and Neonectria radicicoladDirect Blue 86 (Ghariani et al., 2019). While for the biosorption Reactive Red 195 onto soybean hull, Toth gave the best correlation compared with other available isotherm equations (Módenes et al., 2019). Table 4.6 indicates that RedlichePaterson isotherm is also a popular model

1to represent the equilibria data of the adsorption

of dyes on biosorbents. The RedlichePaterson

1isotherm has the form as follows

: qe ¼ 1 þKRaCReCeb (4.9) For several adsorption systems, the RedlichePaterson could fit the experimental data well, as indicated in Table 4.6. Eq. (4.9) combines both characteristics of Freundlich and Langmuir equations. When b is equal to 0, Eq. (4.9) reduces to Henry's law form, and if b is equal to 1, it becomes a Langmuir equation. 10. Kinetic studies

> 1**For the design of** efficient and effective **adsorption system**

, both equilibria and kinetic data are required. Many

> 1**kinetic models are available to represent the adsorption** kinetic **of**

dyes onto biosorbents. The

> 3**most widely used models to** describe **the** kinetics of **adsorption** of dyes **onto**

biosorbents are pseudoefirst- order and pseudoesecond-order equations. Both of those equations were developed according to the

> 1**concept of chemical reaction** that occurs **on the surface of**

adsorbent (Plazinski et al., 2009). In 1898, Lagergren proposed the pseudoefirst-order equation. This

> 3**empirical equation was developed to describe the adsorption of**

malonic and ocalic acids on the surface of the charcoal (Plazinski et al., 2009). The pseudoefirst-order equation has the following mathematical form: qt ¼ qeð1 ?expðk1tÞÞ (4.10) where qt is the

> 27**amount of dye adsorbed** by biosorbent **at time t**, while qe **and k1** are **the** parameters **of**

the pseudoefirst order which indicate the

> 4**amount adsorbed at equilibrium condition and time scaling factor**

. The parameters qe and k1 are usually obtained through the linear regression method. Since k1

> 4**is a time scaling factor, the** higher the value **of**

this parameter, the sooner the system reaches equilibrium (shorter time required) as illustrated in Fig. 4.3. Usually, the parameter k1 is dependent on the pH of the solution, tem- perature, as well as the initial concentration of the solute (Albadarin and Mangwandi, 2015; Akar and Celik, 2011; Akar and Divriklioglu, 2010). Pseudoesecond-order equation was proposed by Blanchard et al. (1984), which has the form as

follows: qt ¼ 1 þq2ekq2etk2t (4.11) Similar to the parameter k1 in pseudoefirst order, the parameter k2 is also a time scaling factor. The effect of k2 on fractional loading of dye on biosorbent is depicted in Fig. 4.4. The

1**higher the value of** k2, **the shorter** the time **required**

by the system

1**to reach the equilibrium condition. Similar to**

k1, k2 is also influenced by the adsorption condition such as initial concentration, pH of the solution, and temperature. In most adsorption systems, the pseudoesecond order gave better FIG. 4.3 The effect of k1 on fractional loading of dye on biosorbent. FIG. 4.4 The effect of k2 on fractional loading of dye on biosorbent.

1**representation of the experimental data** compared with **the**

pseudoefirst order (Da Silva

6**et al., 2019**; Liu **et al**., 2019; Módenes **et al**., 2019; Moghazy **et al**., 2019; Rehman **et al., 2019**; Lebron **et al., 2018**; Huang **et al**., 2016; Ghaedi **et al**

., 2013). The parameter qe in pseudoesecond order equation is less sen- sitive from the effect of the

1**random experimental error** (Plazinski **et al., 2009**

); therefore, this equation gives better prediction than the pseudoefirst-order equation. 11. Thermodynamic of adsorption of dyes on biosorbents The thermodynamic of adsorption is also one of the important information which is required for the design of the adsorption systems. Unfortunately, there is no single equipment that can be used to determine the thermodynamics of adsorption directly. The only option to obtain the thermodynamic of adsorp- tion is through the interpretation of the adsorption isotherms using Van't Hoff equation lnKD ¼ DSo R ? DH o RT (4.12) where KD is the linear sorption distribution coefficient, DSo and DHo are the standards of entropy and enthalpy change, respectively. In most cases, the value of KD was obtained through a simple relation KD ¼ qe/Ce which is not a correct one (Da Silva

11**et al., 2019**; Degermenci **et al., 2019**; Deniz **and** Yildiz, 2019; Danish **et al., 2018**; Daneshvar **et al**., 2012; Akar **and**

Celic, 2011; Caner et al., 2011; Akar and Divriklioglu, 2010). Through this simple relation, in one adsorption isotherm, there are many qe and Ce, and any single relation between qe and Ce will produce one value of KD, and it is not clear which value of KD should be chosen. The value of KD should be calculated based on the method of Khan and Singh (1987). Through a straight-line plot of ln(qe/ce) versus Ce, the value of KD will be obtained (see Fig. 4.5). Through

12**Van't Hoff** equation, **the** value **of the**

standard of entropy and enthalpy change can be obtained. Another valuable thermodynamic property of adsorption is Gibb's free energy change (DGo) which can be calculated through the following relation: DGo ¼ ? RTlnKD (4.13) If the value of DGo is negative, it means that the adsorption process is spontaneous and feasible in the term of thermodynamic (Daneshvar et al., 2012; Akar and Celic, 2011; Caner et al., 2011; Aksakal and Ucun, 2010; Brito

29**et al., 2010**). The **positive value of** DGo **indicates the** adsorption **of** dyes onto **the**

surface of biosorbents is not a spontaneous process; the thermodynamic equilibrium condition between adsorption and desorption processes could not be achieved (Degermenci et al., 2019; Guerrero-Coronilla et al., 2015). The exothermicity and endothermicity of the

21**adsorption process can be determined from** the value **of** DHo. If **the**

uptake of adsorbate by adsorbent FIG. 4.5 Plot for determination of the value of KD. increases with temperature, the adsorption process is endothermic which is indicated by the positive value of DHo (Degermenci

6**et al., 2019**; Guerrero- Coronilla **et al**., 2015; Daneshvar **et al**., 2012; Akar **and** Celic, **2011**

; Aksakal and Ucun, 2010). While the negative value of DHo suggests that the adsorption process is exothermic, in this case, the

3**temperature has a negative influence on the** amount adsorbed by **the**

adsorbent (Brito et al., 2010). The randomness of the adsorption system is measured by DSo. Positive value of DSo indicates that adsorbate molecules possess a high affinity toward the active sites of the adsorbent. The adsorption system becomes more random between both interfaces of solute and adsorbent as the entropy of the system increases (Degermenci et al., 2019; Daneshvar et al., 2012; Akar and Celic, 2011; Aksakal and Ucun, 2010). When the values of DSo is negative, the adsorption systems become more order (Guerrero-Coronilla et al., 2015; Brito et al., 2010). 12. Conclusions Various low-cost biosorbents have explored its potential application as in- dustrial adsorbents for the removal of dyes from aqueous solutions or waste- water. The performance of the biosorbents in removing dyes

4**is strongly** influenced **by the pH** and temperatures **of the solution**

. Among

4**various adsorption isotherm equations** employed **to correlate the experimental data**

, Langmuir and Freundlich could represent most of the adsorption systems very well. For kinetic models, the pseudoesecond order is the best choice

3**to represent the adsorption kinetic data. The thermodynamic** of adsorption **was**

obtained through the interpretation of the adsorption isotherms using Van't Hoff equation. From the results of the studies, it seems the

3**utilization of these low-cost** biosorbents in **industrial** scale **is still** far **from reality**

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