Brazilian Journal
of Chemical
Engineering

ISSN 0104-6632
Printed in Brazil
www.abeq.org.br/bjche

Vol. 32, No. 01, pp. 1 - 12, January - March, 2015
dx.doi.org/10.1590/0104-6632.20150321s00003020

PREPARING AGRICULTURAL RESIDUE BASED ADSORBENTS FOR REMOVAL OF DYES FROM EFFLUENTS - A REVIEW

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(Submitted: October 9, 2013 ; Accepted: February 11, 2014)

Abstract - Industries engaged in dyeing operation generate coloured effluent due to the presence of spent dyes. Adsorption is among the various treatment processes employed for removal of dyes from effluents. Activated carbon is mostly used as an adsorbent in the treatment process. Attempts have been made by researchers to use non-conventional, low-cost, naturally-occurring biomass as adsorbents, including fruit peals, seeds, leaves, bark, sawdust, straw, ash, sludge and others that are abundantly available. The literature indicates that the dye adsorption capacities of these non-conventional biomasses largely depend on the methods of processing and the types of dyes. This review highlights methods used to prepare the adsorbents, along with their adsorption capacity for removal of different dyes from effluents.
Keywords: Adsorption; Dye removal; Agricultural residue; Low-cost adsorbent.

INTRODUCTION

Annual production of dyes is to the tune of $7x10^5$ tonnes worldwide (Allen and Koumanova, 2005). Dyes produced are classified as acid dyes, basic dyes, direct dyes, disperse dyes, reactive dyes and sulfur dyes according to their chemical constituents and application (Zollinger, 1991). Textile dyeing industries, pulp and paper industries and tanneries are the main consumers of dyes and are also responsible for discharge of voluminous coloured effluents. About 10 to 15% of the dyes consumed in the dyeing processes are disposed in the effluent (USEPA, 1997). In order to address the colour problems, industries are using various treatment processes including adsorption. Activated carbon is the most used adsorbent in adsorption columns. There have been attempts by researchers to explore the adsorption potential of non-conventional, naturally-occurring agricultural residues in dye removal from effluents. In India alone more than 400 million tonnes of agricultural residue is generated annually (Raghuvanshi *et al*., 2004), which includes rice husk, bagasse, stalk, coir pith etc. Exploring application of the agricultural residues for use as adsorbents can provide suitable alternatives for the removal of spent dyes from industrial effluents.

REVIEW OF ADSORBENTS

Fruit Peels

Babu *et al*. (2011) prepared three activated carbons from fruit peels, namely Citrus documana (NCDC), Citrus medica (NCMC) and Citrus aurantifolia (NCAC). The peels were dried, crushed and washed. After drying, the peels were carbonized at 500 °C in a nitrogen flow and subjected to oxidation with 1N HNO_3 solution. These carbons were washed

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to remove acid and dried at 150 °C for use as adsorbents for removal of Reactive red 2 dye from effluent. The study reported the adsorption capacities of NCDC, NCMC and NCAC as 0.608 mg/g, 0.580 mg/g and 0.566 mg/g respectively at an initial dye concentration of 20 mg/g and adsorbent dose of 30 g/L. the adsorption potential of mosambi peel was studied by Ladhe *et al*. (2011) using Erichrome black T dye as adsorbate. The adsorbent was cleaned, ground to obtain powder (180-300 μm) and dried. Then the powder was treated with concentrated sulfuric acid in a weight ratio of 1:1 for 24 hours, followed by washing with $NaHCO_3$ solution and distilled water and drying. They observed Erichrome black T dye removal efficiency was 90% at a dye concentration of 50 mg/L and adsorbent dose of 0.004 g/cc. Parvathi and Maruthavanan (2010) performed adsorption studies using tapioca peel for removal of Megeta MB dye. They found that higher percentage removals were observed at solution pH 7 and the equilibrium was reached within 120 minutes of contact time. Jackfruit peel (0.84 mm in size) was used as adsorbent by Jayarajan *et al*. (2011) to remove Rhodamine dye. They reported a maximum colour removal of 25.3% at an adsorbent dose of 3.0 g/L and dye concentration of 100 mg/L. In a study by Velmurugan *et al*. (2011), orange peel, neem leaves and banana peel were dried at 105 °C for 48 hours and ground to powder (600 μm particle size). The study reported that the adsorption capacities of orange peel for different dyes were in the order of Methyl orange > Methylene blue > Rhodamine B > Congo red > Methylene violet > Amido black 10B. An orange peel (*Citrus sinensis* L.) based adsorbent was also prepared and employed for the removal of Remazol brilliant blue dye from synthetic dye effluent by Mafra *et al*. (2013). They found that the equilibrium was reached in 15 hours of contact time with dye concentration 30-250 mg/L. The adsorption capacity of the orange peel adsorbent decreased with increase in temperature. The equilibrium data were reasonably described by the Langmuir and Freundlich isotherms. The authors reported that the adsorption capacities of orange peel adsorbent were 11.62 mg/g, 10.70 mg/g, 8.61 mg/g, 6.39 mg/g and 5.54 mg/g at 20 °C, 30 °C, 40 °C, 50 °C and 60 °C, respectively. In another study, grape juice waste was used by Ansari *et al*. (2011) to evaluate adsorption of Methylene blue dye from aqueous solution. They reported that maximum adsorption of Methylene blue dye was achieved at pH 10. Ong *et al*. (2010) studied adsorption of Methylene blue dye in a packed bed of durian peel powder (1 mm sieve size). The study established that durian peel is a potentially useful and attractive adsorbent for removed of Methylene blue from aqueous solution. They reported that a flow rate of 15 mL/min showed an early breakthrough time.

Sugarcane Bagasse

Azhar *et al*. (2005) studied the removal of Methyl red dye using treated sugarcane bagasse and compared the results with those obtained using powered activated carbon. As per the study, one portion of ground bagasse with particle size between -80 to +230 mesh was treated with 1% formaldehyde in w/v ration of 1:5 at 50 °C for 4 hours followed by activation at 80 °C for 24 hours. The other portion of the bagasse was treated with sulfuric acid and heated in a muffle furnace for 24 hours at 150 ^{o}C, followed by soaking in 1% sodium bicarbonate solution overnight. The study reported adsorption efficiencies of the different adsorbents in the order of powdered activated carbon > bagasse treated with formaldehyde > bagasse treated with sulfuric acid. Untreated, formaldehyde-treated and sulfuric acid-treated sugarcane bagasse powders were used by Abdullah *et al*. (2005) for removal of Ethylene red dye from aqueous solution. The method of preparing formaldehyde-treated bagasse powder and sulfuric acid-treated sugarcane bagasse powder remained the same as that used by Azhar *et al*. (2005). The results reported indicated that sulfuric acid-treated sugarcane bagasse gave better performance as compared to the formaldehyde-treated sugarcane bagasse. Amin (2008) studied the removal of Reactive orange dye from aqueous solutions by activated carbons prepared from sugarcane bagasse. The bagasse powder was sieved to an average size of 0.05 mm. One portion of bagasse was carbonized in the absence of oxygen at 600 °C for 1 hour. The second portion of bagasse powder was soaked in $ZnCl_2$ solution (50% concentration) and the third portion of bagasse powder in H_3PO_4 solution (28% concentration) for 24 hours. After decantation, the samples were pyrolyzed in a muffle furnace in the absence of air at 600 °C for 1 hour. The study reported that the adsorption capacities of physically carbonized, $ZnCl_2$-treated and H_3PO_4-treated bagasse powder samples were observed as 3.48 mg/g, 2.83 mg/g and 1.8 mg/g, respectively.

Adsorption studies were carried out using bagasse fly ash as an adsorbent and reactive dyes as adsorbate by Rachakornkij *et al*. (2004). The fly ash collected from a sugar mill was dried at 110 °C and sieved through a sieve size of 150 μm. The study reported monolayer adsorption capacity of 16.42 mg/g, 32.468 mg/g and 18.282 mg/g with Remazol Black B

dye, Remazol brilliant blue R dye and Remazol brilliant red dye, respectively. Reghuvanshi *et al*. (2004) treated sugarcane bagasse with concentrated sulfuric acid at 150-160 °C for 36 hours. The carbonized material so obtained was washed, dried and ground to 0.33 mm size. The processed bagasse and raw bagasse were used as adsorbents for the removal of Methylene blue from solution. The average percentage of dye removal with processed bagasse was found to be 18% more than that obtained with raw bagasse. The work done by Wong *et al*. (2009) with natural and quartenary ammonium chloride treated sugarcane bagasse shown that the optimum pH for the removal of Basic blue 3 dye and Reactive orange 16 dye is between 6 to 8. For their study, quartenisation of bagasse was carried out by soaking in NaOH solution for 30 minutes. The adsorbent was then mixed with quartenary ammonium chloride (65% w/w) in water. The mixture was heated at 60-70 °C for 4 hours. It was then rinsed with distilled water and suspended in dilute HCl solution at pH 3 for 30 minutes. Then the adsorbent was washed with distilled water and dried at 50 °C in an oven overnight for use as modified adsorbent. The maximum adsorption capacities were reported as 37.59 mg/g and 34.48 mg/g for Basic blue 3 dye and Reactive orange 16 dye, respectively.

Ho *et al*. (2005) studied adsorption of three dyes, namely Basic violet 10, Basic violet 1 and Basic green 4, from aqueous solution onto sugarcane dust (particle size 351-589 μm). The Langmuir monolayer equilibrium capacities for these dyes were 50.4 mg/g, 20.6 mg/g and 13.9 mg/g for Basic violet 1, Basic green 4 and Basic violet 10, respectively. Bagasse ash has been used as a low cost adsorbent by Khandelwal and Gaikwad (2011) for investigating removal of Orange II dye. They reported that the percent adsorption of dye increased with decreasing flow rate from 2 L/hour to 1 L/hour, by increasing bed height from 15 cm to 45 cm, by decreasing the initial dye concentration from 150 mg/L to 100 mg/L and by increasing the column diameter from 2.54 cm to 3.50 cm. Kausik *et al*. (2008) used powdered sugarcane bagasse, coconut coir pith, cow dung and eucalyptus bark treated with 20% sodium hydroxide solution. The adsorbents were aminated with a mixture of 10% amine and water at 70 °C and finally treated with acid for protonation. The adsorbents were used for treating Reactive blue 171, Reactive yellow 84 and Reactive red 141 dyes. The study reported that, among the above four adsorbents, bagasse had a slightly higher efficiency with 20-26% dye removal.

In another study, Ashoka and Inamdar (2010) used formaldehyde-treated bagasse and acid-treated bagasse for adsorption of Methyl red dye. The bagasse powder (0.1456 mm particle size) was reacted with 1% formaldehyde (w/v ratio of 1:5) at 50 °C for 4 hours followed by washing and drying at 80 °C. Acid treatment involved treating bagasse with sulfuric acid (in the ratio of 1:1) and then heating in a muffle furnace for 24 hours at 150 °C and soaking in sodium bicarbonate solution overnight to remove acid, followed by washing and drying at 150 °C for 24 hours. As per the study, the dye removal efficiency of acid-treated bagasse was higher than that of formaldehyde-treated bagasse. Using a similar treatment method for bagasse, Mahesh *et al*. (2010) observed that the Crystal violet dye removal efficiency of formaldehyde-treated bagasse was more than that of acid-treated bagasse.

Husk

Ong *et al*. (2007) ground rice hull to pass through a 1 mm sieve and used it as natural rice hull (NRH). Ethylenediamine (EDA) modified rice hull was also prepared by treating natural rice hull with ethylenediamine in a ratio of 1.0 g rice hull to 0.02 mole of EDA in a well-stirred water bath at 80 °C for 2 hours to enable it to function as a sorbent for removal of Basic blue 3 and Reactive orange 16 dyes. They observed adsorption capacities, calculated from the Langmuir isotherm, of 14.68 mg/g and 6.24 mg/g for Basic blue 3 dye and Reactive orange 16 dye respectively. Sharma and Janveja (2008) conducted a study on the removal of Congo red dye from the effluent of a textile industry using rice husk carbon activated by steam. The study reported that a dose of 0.08 g/L of rice husk carbon removed 10 to 99% of dye from aqueous solution with an initial dye concentration of 25 ppm within contact times from 20 to 200 minutes.

Sawdust

Experiments for removal of Ethylene blue dye using saw dust (420-85 μm) were performed by Gong *et al*. (2008). The activation process included treating sawdust with 240 mL of dioxane, 24 mL of 20% NaOH and 40 ml of epichlorohydrin for 5 hours at 65 °C. The reaction product was filtered, washed and dried. They observed Langmuir adsorption capacities of untreated and treated sawdust as 87.7 mg/g and 188.7 mg/g, respectively. Beach wood sawdust was used as an adsorbent by Izadyar and Rahimi (2007) to treat Direct orange 26, Acid orange 7 and Acid green 20 dyes. The adsorbent with a sawdust particle size of 150-250 μm had monolayer adsorption ca-

pacity of 2.78 mg/g, 5.06 mg/g and 7.81 mg/g for Direct orange 26 dye, Acid orange 7 dye and Acid green 20 dye, respectively. El-latif *et al*. (2010) performed a study of removal of Methylene blue dye using oak sawdust. The sawdust was treated with 0.1N NaOH solution and immobilized on alginate biopolymer for use as an adsorbent. The study revealed an adsorption capacity of sawdust of 38.46 mg/g. The kinetics followed closely a Pseudo-second order model.

Sludge

Activated sludge was dried at 105 °C to a constant weight and sieved to < 205 μm for use as an adsorbent in a study for removal of Rhodamine-B dye (Ju *et al*., 2006). The results indicated that the adsorption capacity of activated sludge increased with decreasing initial pH and temperature. The Langmuir monolayer adsorption capacities were 5.121 mg/g, 4.847 mg/g, 4.456 mg/g and 3.725 mg/g at temperature of 5 °C, 15 °C, 25 °C, and 45 °C, respectively. Reddy *et al*. (2006) conducted a study of reactive dye removal from dyeing unit effluent using sewage sludge-derived activated carbons by pyrolysis. The pyrolysis was carried out in a muffle furnace in the absence of oxygen. Subsequently, 10 g of pyrolysed sludge was impregnated into 25 mL of activating agent (3M $ZnCl_2$ solution) for 24 hours at room temperature, followed by washing with distilled water and drying at 105 °C for 24 hours. The Langmuir monolayer adsorption capacity of the sludge-derived carbon was 33.5 mg/g. The adsorption potential of granular sludge (from a pilot scale reactor treating waste water) with Acid orange 7 dye was investigated by Mendez-Paz *et al*. (2005). A dye removal efficiency of 92% was achieved in continuous treatment mode with dye loading rate of 590 mg/L.day. Won *et al*. (2006) studied the adsorption potential of protonated fermentation waste (*Corynebacterium glutamicum*) with Reactive black 5 dye. For that study, the protonated biomass was prepared by treating the biomass with 1N HNO_3 solution, followed by washing with deionized distilled water and drying. They observed that, in the range of pH 1 to 3, removal of dye was 100%. The maximum dye uptakes using the Langmuir isotherm were 169.5 mg/g and 185.2 mg/L at 20 °C and 40 °C, respectively. The study further showed that the uptake of dye was not significantly affected by the concentration of salt in solution. Won *et al*. (2006a) studied different sludges from a water treatment plant, sewage treatment plant, anaerobic digestion and land fill for the removal of Reactive orange 16 dye. The sludges were treated with 1 M HNO_3 solution for 24 hours and dried at 60 °C to use these sludges as adsorbents. The maximum adsorption capacities using the Langmuir equation were 159.0 mg/g, 114.7 mg/g, 86.8 mg/g 47.0 mg/g for land fill sludge, sewage sludge, anaerobic sludge and water treatment plant sludge, respectively at pH 2.

Organisms

Kim *et al*. (2004) studied the adsorption of Reactive orange 16 dye using dead cell of brewery yeast. The yeast was washed with deionized water and dried at 80 °C. The dried biomass was ground to an average size of 112.5 μm. An adsorption capacity of 0.604 mg/g, 0.090 mg/g and 0.50 mg/g were observed at solution pHs of 3, 7 and 10, respectively. Studies were conducted by Singh and Rastogi (2004) using baker's yeast cells as an adsorbent for adsorption of various dyes. For preparation of the adsorbent, baker's yeast (2 g) was suspended in saline (6 mL). The yeast was re-suspended in 6 ml of 0.1 M Acetate buffer with pH 4.6. The sediment was further suspended in acetate buffer to obtain a ca. 33% yeast suspension (v/v). Then 3 mL of yeast suspension was added to 1 mL of ferrofluid, and incubated at room temperature for 1 hour. After this treatment, the majority of the yeast cells were modified. The modified yeast cells were heated in boiling water and then washed with saline and stored at 4 °C for use as adsorbent. The study reported that the adsorption capacities for Acridine orange dye, Aniline blue dye, Crystal violet dye, Malachite green dye and Safranine O dye were 82.8 mg/g, 430.2 mg/g 85.9 mg/g, 19.6 mg/g and 90.3 mg/g, respectively. The decolourization potential of fungus was performed by Akar *et al*. (2009) using Reactive red 198 dye. A maximum dye adsorption capacity of 1.03×10^{-4} mol/g was observed at pH 2 and adsorbent dose of 2.0 g/L. They observed an increase in the adsorption capacity with temperature, indicating that the adsorption process is endothermic. Fu and Viraghavan (2003) studied the dye removal potential of immobilized fungal biomass (*Aspergillus niger*) with four dyes, namely Acid blue 29, Basic blue 9, Congo red and Disperse red 1. The adsorption capacities observed were 64.7 mg/g for Acid blue 29 dye, 8.9 mg/g for Basic blue 9 dye, 1.1 mg/g for Congo red dye and 0.1 mg/g for Disperse red 1 dye.

Grains

Jaikumar and Ramamurthi (2009) studied the adsorption of Acid yellow 17 dye by an adsorbent

prepared from spent brewery grains. The spent brewery grains were suspended in 0.13 M sulphuric acid solution (20 g of grain per 100 mL of solution) for one hour. The grains were washed, dried and ground for use as adsorbent. They observed the highest adsorption capacity at pH 2 with an initial dye concentration of 150 mg/L, dose of adsorbent 0.5 g/L and contact time of 40 minutes.

Coconut

Theivarasu and Mylsamy (2010) conducted an adsorption study of Rhodamine-B dye on char prepared by treating the coconut shell with concentrated sulfuric acid at ratio of 1:1 (w/v). The activation was performed by heating in a muffle furnace at 550 °C for 7 hours, followed by washing and drying. The adsorption capacity of the treated coconut shell char was reported as 41.67 mg/g. For removal of Coomassie brilliant blue dye on coir pith as adsorbent, Prasad *et al*. (2008) conducted studies including the effects of time, initial dye concentration and dose of adsorbent. Treatment of coir pith includes dipping the coir pith in a one molar solution of HCl, washing with distilled water and drying in an oven at 55 °C. The study reported a maximum adsorption capacity of 31.84 mg/g and the adsorption capacity for the system was 6.43 mg/g.

Palm Shell

Rajavel *et al*. (2003) evaluated the removal efficiency of Dark green PLS dye from textile industry wastewater using carbons prepared from palm nut shell, cashew nut shell and broom stick. The carbons were prepared by treating 4 parts of each material with 2 parts of concentrated sulfuric acid and heating at 140-170 °C for 24 hours. The resultant materials were filtered, washed with water, dried at 105-110 °C and sieved to an average particle diameter of 0.5 mm for use as adsorbents. The adsorption capacities of palm nut shell carbon, cashew nut shell carbon and broom stick carbon were reported as 0.84 mg/g, 1 mg/g and 0.63 mg/g, respectively. The adsorption study by Rusly and Ibrahim (2010) involved palm shell activated carbon and Reactive red 3 BS dye. They observed that, upon increasing adsorbent dose and agitation, the efficiency of dye removal increased. The study reported that, at the optimal condition, the dye removal efficiency reached more than 90% and the adsorption capacity was more than 7 mg/g. Batch adsorption experiments were carried out by Sreelatha and Padmaja (2008) for removal of Methylene blue and Rhodamine 6G dyes from solution using palm shell powder. The sorption capacity was dependent on the operating parameters and the process was pH dependent above pH 4.0. The adsorption capacities were reported to be 121.5 mg/g and 105 mg/g for Methylene blue dye and Rhodomine 6G dye, respectively.

Leaves

Gulmohor leaves were ground, washed and dried to use as adsorbent in an adsorption study by Ponnusami *et al*. (2009). The results indicated that the equilibrium dye removal capacity of gulmohar leaves with Methylene blue dye varies from 132.40 mg/g to 34.76 mg/g with adsorbent dose of 0.5 g to 2.5 g/L and a dye concentration of 100 mg/l. The monolayer adsorption capacities of gulmohar leaf powder were observed 120 mg/g, 178 mg/g and 253 mg/g at temperature of 293 K, 303 K and 313K, respectively. Singh and Rastogi (2004) used tea leaves as adsorbent for removal of Malachite green and Methylene blue dyes. The dried leaf powder was impregnated with H_3PO_4 (50% w/v) in the ratio of 2:1 (w/v) and carbonized at 300 °C. The carbons were washed and dried at 100 °C and sieved to 170-200 mesh size. The adsorption data fitted well the Langmuir isotherm with monolayer adsorption capacities of 444.44 mg/g and 454.5 mg/g for Malachite green dye and Methylene blue dye, respectively at 25 °C. The column study indicated break through capacities of 300 mg/g and 275 mg/g for Malachite green dye and Methylene blue dye respectively. Hamissa *et al*. (2008) conducted a study on adsorptive removal of Alpacide yellow dye from aqueous solution on fibres extracted from agave leaves. The agave leaves were subjected to a salt hydrolysis at 80 °C for 8 hours. The extracted fibres were washed to remove the parenchyma and cut to 4 cm size and dried at 70 °C for use as adsorbent. They observed maximum adsorption capacity of agave leaves of 16.97 mg/g, 15.79 mg/g and 21.41 mg/g at 20 °C, 30 °C and 50 °C, respectively. Neem (Azadirachta indica) leaf powder was used for treatment of Fast green dye (C.I. 42053) in a study conducted by Tahir *et al*. (2008). The neem leaves were powdered and washed with distilled water and then dried at 60 °C for use as an adsorbent. The study indicated that the maximum adsorption capacity was 92.6% with 1 g/30 mL of adsorbent dose and $5x10^{-4}$ mol/dm^3 strength solution of Fast green dye. Hema and Arivoli (2007) studied adsorption of dyes onto acid activated pandanus leaves. The activation method involved carbonization with concentrated sulfuric acid in a ratio of 1:1 (w/v) with heating at 400 °C for 12 hours in a

furnace. The resulting carbon was washed with distilled water and then dried at 100 °C for 4 hours for use as adsorbent. The adsorption capacities were 21.491 mg/g, 20.267 mg/g, 20.069 mg/g and 18.928 mg/g at 30 °C, 40 °C, 50 °C and 60 °C, respectively in the case of Congo red dye. With Malachite green dye, the observed adsorption capacities were 9.737 mg/g, 9.624 mg/g 9.633 mg/g and 9.569 mg/g at 30 °C, 40 °C, 50 °C and 60 °C, respectively.

Waste tendu (Diospyros melanoxylon) leaf cuttings were processed and used as an adsorbent for removal of Crystal violet dye by Nanda and Ghole (2008). Tendu leaf cuttings were powdered and sieved with 80 mesh (called TLR). Carbons were prepared by treating 5 parts of TLR with 3 parts of concentrated sulfuric acid at 120-130 °C for 24 hours. The carbonized mass was freed from acid by soaking in 1% solution of sodium bicarbonate, followed by drying and sieving through 80 mesh sieve for use as adsorbent (called TLR-CM). One portion of LTR was treated with 5 parts of 2N sulfuric acid for 24 hours. Then the material was washed, dried, powdered and used as adsorbent (called TLR-2N). The study reported adsorption capacities of 67.57 mg/g, 42.92 mg/g and 22.47 mg/g for TLR-2N, TLR and TLR-CM, respectively. El-Zawahry and Kamel (2004) used water hyacinths (*Eichhornia crassipes*) powder as adsorbent for removal of acid and reactive dyes from aqueous solution. The samples were cut to smaller size, air dried and ground and sieved to 0.147-1.5 mm size. The powder was subjected to a chemical scouring treatment (boiling with 20 g/L sodium hydroxide for 2 hours at 120-130 °C in a 20:1 ratio). The material was filtered, washed and air dried for use as adsorbent. The study reported that the higher nitrogen percent of hyacinths showed higher adsorption capacities. Vijayaraghavan and Yun (2008) studied adsorption of Reactive black 5 dye using seaweed (Luminaries sp.). The adsorbent was ground to an average size of 0.4-0.6 mm. Protonation of adsorbent involved treatment with 10 g/L of 0.1 M HCl solution followed by washing and drying. The maximum dye uptake of 101.5 mg/g was observed at pH 1.0 and 40 °C.

Ncibi *et al*. (2007) studied adsorption of Reactive red 228 on sea grass leaf sheaths with variables including temperature, pH and chemical pre-treatment, and observed maximum colour removal at pH 5.0. Pre-treatment of the absorbent with phosphoric acid and nitric acid solution increased the adsorption efficiency up to 80%. Aquatic plant (*Hydrilla verticillata*) biomass was experimented for removal of Malachite green dye from solution by Rajeshkannan *et al*. (2010) and the study showed an adsorption capacity of 91.97 mg/g at pH 8.0. Purai and Rattan (2009) used ash prepared from cow dung and commercial activated carbon. The ash was prepared in muffle furnaces at 500 °C. The study revealed that 4.98 mg/g and 4.67 mg/g of dye were adsorbed dye adsorbed by cow dung ash and activated carbon, respectively, at pH 3.33. The authors noted that at pH 8.76, the adsorption was the least for both the adsorbents.

Tree Bark

Patil *et al*. (2011) carried out adsorption studies of Methylene blue dye using teak tree bark with various process parameters. The maximum adsorption of Methylene blue dye was 333.33 mg/g. The study revealed an increase in dye adsorption efficiency with increasing pH, increasing temperature and decreased particle size of adsorbent.

Straw

Abdualhamid and Asil (2011) conducted adsorption studies for removal of Methylene blue dye using barley, wheat and oat straws as adsorbents. The straws were cut into pieces of 1 cm size, washed and dried at 65 °C overnight. One portion of each straw was subjected to soaking by immersing in water at room temperature for 20 days and then dried at 60 °C overnight for use as adsorbent. In the study, the maximum dye adsorption capacity for the straws before soaking followed the order: barley > oat > wheat, with values of 27.72 mg/g, 17.54 mg/g and 8.34 mg/g, respectively. The maximum dye removal capacity of straws after soaking in water was found in the order of oat > barley > wheat, with values of 50.00 mg/g, 22.22 mg/g and 11.11 mg/g.

Seeds

Esterified natural papaya seeds were used by Nasuha *et al*. (2011) for adsorption of Methylene blue and Congo red dyes from effluent. Esterification was carried out by treating the adsorbent with methanol and HCl followed by washing and drying. Data for adsorption of Methylene blue dye fit well to the Langmuir isotherm and maximum adsorption capacities of 250.0 mg/g and 200 mg/g were observed for esterified adsorbent and natural adsorbent, respectively. Santhi *et al*. (2010) studied the adsorption potential of *Annona squamosa* seed with adsorbates, namely Methylene blue dye, Methylene red dye and Malachite green dye. Carbon was prepared by treating the mass with H_2SO_4 for 12 hours. After washing, the mass was treated with 2% $NaHCO_3$

solution to remove remaining acid, followed by drying and sieving to 125-250 μm size. Dye adsorption capacities of 8.52 mg/g, 40.48 mg/g and 25.91 mg/l were observed for Methylene blue dye, Methylene red dye and Malachite green dye, respectively. Decolourization studies of Acid orange dye 7 by charcoal prepared from coffee grounds were performed by Nakamura *et al*. (2003). For preparing the adsorbent, extracted residues of coffee beans was dried to reduce the moisture content by 50% and the same was carbonized in a furnace at 800 °C, 1000 °C and 1200 °C and sieved through 10-20 mesh. The carbons were washed with distilled water and dried at 110 °C for use as adsorbents. The studies indicated that the equilibrium adsorption of Acid orange 7 dye was higher for charcoal carbonized at higher temperatures.

Other Biomasses

Habib *et al*. (2006) performed adsorption studies using tuberose sticks as adsorbent for removal of Methylene blue dye. The dried tuberose sticks were cut into small pieces, powdered and then sieved with a 425 μm sieve for use as adsorbent. The maximum dye removal of 80% was achieved at pH 11, adsorbent dose of 1 g/L and dye concentration in solution of 40 mg/L. In another study by Ramakrishna and Nagarajan (2009), flame tree (Delonix regia) pods were used for preparing adsorbents. The flame tree pods were crushed into smaller pieces and soaked with concentrated sulfuric acid in a 1:1 ratio (weight of material to volume of acid) for 48 hours and activated at 160 °C for 6 hours. The carbon so prepared was washed with distilled water and dried at 105 °C for 2 hours to prepare the adsorbent. The data were reported to fit well to the Langmuir and Freundlich isotherms. The maximum adsorption capacity observed with Crystal violet dye was 16.70 mg/g. A granule prepared from leaf, fruits and twigs of *Muntingia calabura* was utilized by Santhi *et al*. (2009) for adsorption of Methylene blue, Methylene red, and Malachite green dyes. The maximum adsorption capacities were 20 mg/g, 58 mg/g and 32 mg/g for Methylene blue dye, Methylene red dye and Malachite green dye, respectively. Sivakumar and Palanisamy (2010) prepared an adsorbent by treating precursor wood with H_3PO_4 solution followed by activation at 800 °C. The adsorbent so processed had a surface area of 918 cm^2/g. The adsorbent was used to remove Acid blue 92, Basic blue 29, Reactive red 4, and Direct blue 53 dyes from aqueous solution. Mittal *et al*. (2007) investigated the removal of Tartrazine dye by using hen feathers. To remove the adhering organic matter, feathers were treated with hydrogen peroxide followed by washing and drying. The study reported dye removal efficiencies of 47%, 52% and 55% at 30 °C, 40 °C and 50 °C, respectively. Piccin *et al*. (2011) tested adsorption potential of chitosan for removal of commercial dye (FD&C Red n° 40) at different temperatures (298 to 338 K). The maximum adsorption capacity of chitosan was observed as 1065.8 μmol/g, 1061.4 μmol/g, 800.8 μmol/g, 508.5 μmol/g at 308 K, 318 K, 328 K and 338 K, respectively. The authors observed that the adsorption process was exothermic in nature. Adsorption studies of Reactive red 120 and Reactive black 5 dyes onto cotton fibre were performed by Gamal *et al*. (2010). They observed the monolayer adsorption capacities of 11.63 mg/g and 6.22 mg/g for Reactive red 120 dye and Reactive black 5 dye, respectively. Schimmel *et al*. (2010) studied the adsorption potential of commercial activated carbon for Turquoise blue QG reactive bye. The adsorption studies were conducted to obtain isotherm and kinetic data under different experimental conditions. They observed maximum dye removal at a pH of 2 and temperature of 30 °C. The equilibrium data were reasonably described by the Langmuir and Freundlich isotherms. The authors reported that the adsorption capacity of activated carbon was 140.14 mg/g. The kinetics followed closely a Pseudo-second order model. Table 1 show different agricultural residues used to prepare the adsorbents, along with adsorption capacities for removal of different dyes from effluents.

Table 1: Adsorption capacities of some agricultural residue based adsorbents for removal of different dyes from effluents.

Name of Adsorbent	Dye	Adsorption Capacity	Reference
Citrus documana	Reactive red 2	0.608 mg/g	Babu *et al*. (2011)
Citrus medica	Reactive red 2	0.580 mg/g	Babu *et al*. (2011)
Citrus aurantifolia	Reactive red 2	0.566 mg/g	Babu *et al*. (2011)
Orange peel (*Citrus sinensis* L.)	Remazol brilliant blue	11.62 mg/g (20 °C), 10.70 mg/g (30 °C), 8.61 mg/g (40 °C), 6.39 mg/g (50 °C), 5.54 mg/g (60 °C).	Mafra *et al*. (2013)

Continuation Table 1

Continuation Table 1

Name of Adsorbent	**Dye**	**Adsorption Capacity**	**Reference**
Mosambi peel	Erichrome black T	90% (Initial dye concentration 50 mg/L & adsorbent dose 4 g/L)	Ladhe *et al*. (2011)
Palm nut shell carbon	Dark green PLS	0.84 mg/g	Rajavel *et al*. (2003)
Cashew nut shell carbon	Dark green PLS	1 mg/g	Rajavel *et al*. (2003)
Broom stick carbon	Dark green PLS	0.63 mg/g	Rajavel *et al*. (2003)
Coconut shell char	Rhodamine-B	41.67 mg/g	Theivarasu and Mylsamy (2010)
Coir pith char	Coomassie brilliant	31.84 mg/g	Prasad *et al*. (2008)
Palm shell activated carbon	Reactive red 3 BS	7 mg/g	Rusly and Ibrahim (2010)
Palm shell powder	Methylene blue Rhodamine 6G	121.5 mg/g 105 mg/g	Sreelatha and Padmaja (2008)
Sugarcane bagasse	Reactive orange	3. 48 mg/g	Amin (2008)
Sugarcane bagasse ($ZnCl_2$ treated)	Reactive orange	2.83 mg/g	Amin (2008)
Sugarcane bagasse (H_3PO_4 treated)	Reactive orange	1.8 mg/g	Amin (2008)
Sugarcane **b**agasse fly ash	Remazol Black B Remazol brilliant blue R Remazol Brilliant red	16.42 mg/g 32.468 mg/g 18.282 mg/g	Rachakornkij *et al*. (2004)
Sugarcane bagasse	Basic blue 3 Reactive orange 16	37.59 mg/g 34.48 mg/g	Wong *et al*. (2009)
Sugarcane dust	Basic violet 1 Basic violet 10 Basic green 4	50.4 mg/g 13.9 mg/g 20.6 mg/g	Ho *et al*. (2005)
Rice hull	Basic blue 3 Reactive orange 16	14.68 mg/g 6.24 mg/g	Ong *et al*. (2007)
Rice husk carbon	Congo red	10 to 99% (Initial dye concentration 25 ppm & adsorbent dose 0.08 g/L)	Sharma and Janveja (2008)
Saw dust	Ethylene blue	87.7 mg/g (natural saw dust). 188.7 mg/g (treated saw dust)	Gong *et al*. (2008)
Beech wood saw dust	Direct orange 26 Acid green 20 Aid orange 7	2.78 mg/g 7.81 mg/g 5.06 mg/g	Izadyar and Rahimi (2007)
Activated sludge	Rhodamine-B	5.121 mg/g (5 °C), 4.847 mg/g (15 °C), 4.456 mg/g (25 °C), 3.725 mg/g (45 °C)	Ju et at. (2006)
Sewage sludge activated carbons	Reactive dye	33.5 mg/g	Reddy et at. (2006)
Granular activated sludge	Acid orange 7	92% (at dye loading rate of 590 mg/L.day	Mendez-Paz *et al*. (2005)
Fermentation waste (*Corynebacterium glutamicum*)	Reactive black 5	169.5 mg/g (20 °C), 185.2 mg/g (40 °C)	Won *et al*. (2006)
Sewage treatment plant sludge	Rhodamine B	5.121 mg/g (5 °C), 4.847 mg/g (15 °C), 4.456 mg/g (25 °C), 3.72 5 mg/g (45 °C)	Ju *et al*. (2006)
Water treatment plant sludge	Reactive orange 16	47.0 mg/g	Won *et al*. (2006a)
Sewage treatment plant sludge	Reactive orange 16	114.7 mg/g	Won *et al*. (2006a)
Anaerobic digestion sludge	Reactive orange 16	86.8 mg/g	Won *et al*. (2006a)
Land fill sludge	Reactive orange 16	159.0 mg/g	Won *et al*. (2006a)
Brewery yeast dead cell	Reactive orange 16	0.604 mg/g (pH 3), 0.090 mg/g (pH 7), 0.50 mg/g (pH 10)	Kim *et al*. (2004)
Baker's yeast cells	Acridine orange Aniline blue Malachite green Safranine O Crystal violet	82.8 mg/g, 430.2 mg/g 19.6 mg/g 90.3 mg/g 85.9 mg/g	Singh and Rastogi (2004)
Fungus	Reactive red 198	1.03×10^{-4} mol/g	Akar *et al*. (2009)
Fungal biomass (*Aspergillus niger*)	Acid blue 29 Disperse red 1 Congo red Basic blue 9	64.7 mg/g 0.1 mg/g 1.1 mg/g 8.9 mg/g	Fu and Viraghavan (2003)
Gulmohor leaves	Methylene blue	120 mg/g (293 K), 178 mg/g (303 K) and 253 mg/g (313K)	Ponnusami *et al*. (2009)

Continuation Table 1

Continuation Table 1

Name of Adsorbent	Dye	Adsorption Capacity	Reference
Used tea leaves carbon	Malachite green Methylene blue	444.44 mg/g 454.5 mg/g	Singh and Rastogi (2004)
Agave (Americana (L.*)* leaves fibres	Alpacide yellow	16.97 mg/g(20 °C), 15.79 mg/g (30 °C) and 21.41 mg/g (50 °C)	Hamissa *et al*. (2008)
Pandanus leaves	Congo red	21.491 mg/g (30 °C), 20.267 mg/g (40 °C), 20.069 mg/g (50 °C), 18.928 mg/g (60 °C)	Hema and Arivoli (2007)
Pandanus leaves	Malachite green	9.737 mg/g (30 °C), 9.624 mg/g (40 °C), 9.633 mg/g (50 °C), 9.569 mg/g (60 °C)	Hema and Arivoli (2007)
Tendu (*Diospyros melanoxylon*) leaf	Crystal violet	67.57- 22.47 mg/g depending on processing of adsorbent.	Nagda and Ghole (2008)
Tuberose sticks	Methylene blue	80% (at pH 11, adsorbent dose 1 g/L, 40 mg/L dye concentration)	Habib *et al*. (2006)
Flame tree (*Delonix regia*) pods	Crystal violet	16.70 mg/g	Ramakrishna and Nagarajan (2009)
Muntingia calabura Leaves	Methylene blue Methylene red Malachite green	20 mg/g 58 mg/g 32 mg/g	Santhi *et al*. (2009)
Water hyacinths (*Eichhornia crassipes*)	Acid and reactive dyes	Higher N_2 percent of hyacinths showed higher adsorption capacities	El-Zawahry and Kamel (2004)
Seaweed (Luminaries sp.)	Reactive black 5	101.5 mg/g	Vijayaraghavan and Yun (2008)
Sea grass leaf sheaths	Reactive red 228	80% dye removal efficiency at pH 5	Ncibi *et al*. (2007)
Aquatic plant (*Hydrilla verticillata*)	Malachite green	91.97 mg/g	Rajeshkannan *et al*. (2010)
Teak tree bark	Methyleme blue	333.33 mg/g	Patil *et al*. (2011)
Oak saw dust	Methylene blue	38.46 mg/g	El-latif et at. (2010)
Barley straw	Methylene blue	27.72 mg/g	Abdualhamid and Asil (2011)
Wheat straw	Methylene blue	17.54 mg/g	Abdualhamid and Asil (2011)
Oat straw	Methylene blue	8.34 mg/g	Abdualhamid and Asil (2011)
Papaya seeds	Methylene blue	250.0 mg/g (esterified adsorbent) 200 mg/g (natural adsorbent)	Nashuha *et al*. (2011)
Annona squamosa seeds	Methylene blue Methylene red Malachite green	8.52 mg/g 40.48 mg/g 25.91 mg/g	Santhi *et al*. (2010)
Hen feathers	Tartrazine (azo dye)	47% (30 °C), 52% (40 °C), 55% (50 °C)	Mittal *et al*. (2007)
Chitosan	FD&C Red n° 40 dye	1065.8 μmol/g (308 K), 1061.4 μmol/g (318 K), 800.8 μmol/g (328 K), 508.5 μmol/g (338 K)	Piccin *et al*. (2011)
Cotton fibres	Reactive red 120 Reactive black 5	11.63 mg/g 6.22 mg/g	Gamal *et al*. (2010)
Commercial activated carbon	Turquoise blue QG reactive bye	140.14 mg/g	Schimmel *et al*. (2010)

CONCLUSIONS

Agricultural residues are abundantly available. For using as adsorbents, the agricultural residues are required to be properly treated. The treatments employed by researchers involve physical and chemical processes such as washing, drying, size reduction, burning to produce ash, burning in the absence of oxygen to obtain char, carbonizing and specific treatment to effect chemical modifications. This literature review shows that it is possible to develop agricultural residues for use as adsorbents in colour removal from effluents. The adsorption capacity data reported in the literature indicate that dye removal through the use of agricultural residue is feasible. Although intensive studies have been undertaken on the lab scale with different processes parameters, there is a need to explore the adsorption potential of the agriculture residues through pilot plant studies to establish the treatment process at commercial level.

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