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Treatment of pulp and paper mill effluent using low cost adsorbents: An overview

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Abstract
In the present review, the suitability of adsorption process using low cost adsorbent for the treatment of pulp and paper mill effluent has been discussed. It is clear that adsorption processes are appropriate for the removal of recalcitrant compounds such as surfactants and pesticides, among others biodegradable or non-biodegradable compound present in pulp and paper mills effluents. The importance of the adsorption is to improvement of the removal of various physico- chemical (biological oxygen demand (BOD), chemical oxygen demand (COD), colour, suspended solids, lignin), heavy metals (Cu, Cr, Fe, Zn, Ni and Mn etc) organochlorine compounds, all recalcitrant pollutant, reduce toxicity, enhance colour removal by using different cost effective adsorbents. The effective use of the different adsorbents developed from different adsorbent media such as activated carbon, agriculture by product and industrial wastes and sludge as adsorbents for the removal of different pollutants from the various processes and operations of pulp and paper mill as potential alternatives to different treatment process and received widespread attention. Adsorption necessity is a novel treatment option to improve the efficiency of removal within the discharge limits of wastewaters into the receiving bodies without causing any damage of the environment. However, still there is a need to find out the practical usefulness of such low cost adsorbent at industrial scale with the special reference to metals.

Keywords: Adsorption, Adsorbent, Agriculture waste materials, Pulp and paper mill effluent

INTRODUCTION

Industrialization has resulted in the formation of huge amount of waste products, which are released into the environment in the form of wastewater leading to environmental pollution and deterioration. The amounts of industrial wastes that cause the pollution interference with the best usage of the receiving water have initiated with the turn of the twentieth century (Nemerow 2007). The increasing public awareness of the fate of pollutants and stringent regulations recognized by the several authorities and agencies are pushing the industry to treat effluents to the required compliance level before discharging in to the environment (D'Souza *et al*., 2006).The environmental pollution by pulp and paper industry are not limited by wastewater generation, solid wastes including sludge generating from wastewater treatment plants and air emissions and other problems. Their disposal and treatment are essential for the environmental safety. The significant solid wastes such as saw dust, lime mud, lime slaker grits, green liquor dregs, boiler and furnace ash, scrubber sludge, wood processing residuals and wastewater treatment sludge are generated from different mills. These solid wastes have content the high amount of organic materials, partitioning of chlorinated organics, pathogens, ash and trace amount of metallic ions and their disposal cause the environmental pollution (Monte *et al*., 2009; Azimvand and Mirshokraie, 2016).
The importance of water quality preservation and improvement is essential for human life and protection to the environment. The scientists, academicians and governmental organizations are very serious on the pollution of water capitals globally. The surface and ground waters at many places of the world are contaminated and not fit for drinking purpose. By 2050, the worldwide population is

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Table 1. Potential pollutants from pulp and paper mills (Ali and Sreekrishnan, 2001).

Types of pollutants	Typical example and sources
Gases	Malodorous gases e.g. H_2S and mercaptain from Kraft pulping and recovery processes Oxides of sulfer e.g. SO_2 and SO_3 from recovery furnaces and lime kilns.
Effluents	Suspended solids including bark particles fiber pigments, dirt from debarking.
	Dissolved colloidal organics e.g. hemicellulose, sugars, sizing agents
	Chromotophores mainly lignin compounds
	Chlorinated compounds from bleach plants
	Dissolved organics, e.g. NaOH,Na_2,Na_2SO_2
	Tharmal loads
Particulates	Fly ash from coal fired power boilers
	Char from bark burners
Solid wastes	Sludges from primary and secondary treatment and recovery section
	Solids such as grit, bark and other mill solid wastes

Table 2. Physical-chemical properties of wastewater of paper mill along with WHO permissible limits (1995), for the release of treated wastewater to irrigation waterway (Farhan *et al.,* 2013).

Parameters	Characteristics of wastewater	Characteristics after treatment	WHO permissible limits
pH	6.39	6.9	6.8 - 8.5
Color	Dark brown	Colorless	
Temperature (o C)	30	Ambient temperature	20
Total solids (mg/l)	6810	>100	655
Total dissolved solids (mg/l)	3080	> 70	450
Total suspended solids (mg/l)	3720	> 32	200
Chemical oxygen demand (mg/l)	1640	> 147.6	300

expected to reach up to 9.3 billion (United Nations, 2011) and the world may be under great fresh water inadequacy in near future. Therefore, the removal of toxic organic and inorganic pollutants from wastewater is important in the present scenario (Ali *et al.,* 2012).Pollutants which are released during several process of pulp and paper production can be reduced by adopting several internal treatment methods, improvements, especially in combination with management measure of the industry (Kamali and Khodaparast, 2015).

The characteristics of the effluents to be treated depends on the type of wood, the nature of process, the quantity of water that the mill is able to circulate, the technology used and the selected management practices. Depending on the raw material and the process involved in pulp and paper mill, untreated waste effluents can have high biochemical oxygen demand (BOD), chemical oxygen demand (COD) (1640 mg/l), solids (6810 mg/l) dissolved solids (3080 mg/l) suspended solids (mainly fibers) (3720 mg/l), fatty acids, tannins, resin acids, lignin, and its derivatives. Certain pollutants are naturally arising and others are xenobiotics, which are formed during the pulp and paper manufacturing process (Area and Valade, 1998; Ali and Sreekrishnan, 2001 and Farhan *et al.,* 2013).

According to Ahmad and Hameed (2010); El-Ashtoukhy *et al*. 2008; Gong *et al.* (2008), various methods have been employed for waste water treatment, activated carbon adsorption was found to be superior for wastewater treatment compared to other physical and chemical techniques, as they possess inherent limitations such as high cost, formation of hazardous by-products and intensive energy requirements, activated carbon has emerged as a potential alternative to conventional physiochemical technologies in waste-treatment facilities. Adsorption process is very effective method in terms of cost, flexibility, simplicity of design, and ease of operation compared to other techniques. It does not result in the formation of harmful substances (Rafatullah *et al.*

Table 3. Alternative feedstock's proposed for the preparation of activated carbon (Mohanand Pittman, 2007) .

Bones	Lampblack
Bagasse	Leather waste
Bark	Municipal waste
Beat-sugar sludge	Molasses
Blood	Nut shells
Blue dust	News paper
Coal	Oil shale
Coffee beans	Olive stone
Coconut shell	Petroleum acid sludge
Coconut coir	Pulp mill waste
Cereals	Palm tree cobs
Carbohydrates	Petroleum cobs
Cottonseed hulls	Petroleum coke
Cottonseed hulls	Potassium ferrocyanide residue
Corn cobs	Rubber waste
Distillery waste	Rice hulls
Fertilizer waste slurry	Refinery waste
Fertilizer waste slurry	Scrap tires
Graphite	Sunflower seeds
Human hairs	Tea leaves
Lignin	Wheat straw
Jute stick	Wood

Table 4. Adsorption capacities qm (mg/g) of different agricultural wastes for the contaminants.

Adsorbent	Adsorbate	Maximum adsorption capacity (mg/g)	Reference
Carrot residues	Cr(III) Cu(II) Zn(II)	45.09 32.74 29.61	Nasernejad *et al.* (2005)
Orange peel	Cu(II) Zn(II)	0.15 0.18	Pehlivan *et al.* (2006)
Banana peel	Basic blue 9	20.8	Annadurai *et al*. (2002)
Rice husk	Acid yellow 36	86.9	Malik (2003)
Straw	Basic blue 9	19.82	Kannan and Sundaram (2001)
Banana stem	Pb(II)	91.74	Noeline *et al.* (2005)
Guava (Psidiumguajava) leaf Powder	Methylene blue	185.2	Ponnusami *et al.*(2008)
Peanut hull	Methylene blue	68.03	Gong*et al.* (2005)
Lemon peel	Methylene blue	29	Kumar and Porkodi (2006)
Peanut hull	Reactive dye	55.5	Tanyildizi (2011)
Maize cob	2,4-Dichlorophenol	17.94 mg/g	
Wheat straw	Cd(II)	14.56	Dang*et al.* (2009)
Sunflower stalk	Pb(II)	182.90	Jalali and Aboulghazi. (2013)

Table 5. Adsorption capacities of different industrial wastes as adsorbents for the removal of various pollutants from water (Bhatnagar and Sillanpaa, 2010; Ahmaruzzaman, 2011).

S. N.	Adsorbent	Adsorbate	Adsorption capacity (mg/g)	Reference
1	Fly ash	Phenol, 3-chlorophenol and 2,4-dichlorophenol	67, 20 and 22	Akgerman, M. Zard-koohi,1996
2	Solid waste from leather industry	As(V) and Cr(VI)	26 and 133	Oliveira *et al*., 2008
3	Biogas residual slurry	Cr(III)	7.8	Namasivayam, R.T. Yamuna,1999
4	Red mud	Cu^{2+}, Zn^{2+}, Ni^{2+} and Cd^{2+}	19.72, 12.59, 10.95 and 10.57	López*et al.,,*1998
5	Boron industry waste	Basic yellow 28 and basic red 46	75.00 and 74.73	Olgun and Atar, 2009
6	Activated slag	Cu, Ni	30.0, 29.35	Gupta, 1998
7	Sludge	Pb^{2+}	39.3	Lister and Line, 2001
8	Lignin	Cr^{3+}	17.97	Guo*et al.,* 2008
9	Tea industry waste	Fe^{3+}	24	Ahluwalia SS, Goyal, 2005

2010). Various low cost adsorbents such as fruit wastes, coconut shell, scrap tyres, bark and other tannin-rich materials, sawdust and other wood type materials, rice husk, petroleum wastes, fertilizer wastes, fly ash, sugar mill wastes blast furnace slag, chitosan and seafood processing wastes, seaweed and algae, peat moss, clays, red mud, zeolites, sediment and soil, ore minerals etc. These adsorbents have been found to remove of various organic pollutants ranging from 80 to 99.9%(Ali *et al.,* 2012).The aim of this review is to provide the information regarding the use of low cost adsorbent such as agricultural waste and industrial waste materials as adsorbents for the removal of numerous toxic contaminants and heavy metals from paper mill wastewater. The review also presents and discusses various factors affecting the adsorption process.

Pulp and paper mill: During the twentieth century the population of the world has increased three folded, while the utilization of water has increased six folded of the population. Globally, irrigation is the largest user of water, followed by industrial use and domestic use (Cosgrove and Rijsberman, 2000). Meeting the increased demand for water, whilst keeping the fresh water eco systems of the world intact, is considered to be one of the biggest challenges of this century. One solution that has been discussed is to increase the reuse of water (Postel, 2000).

The pulp and paper industry is a growing sector that requires a large amount of water and energy. The main environmental issues are emissions to water and air, waste build-up and energy consumption (Suntio *et al.,* 1988). The manufacturing of Pulp and paper with unit production capacities greater than 100 metric tons per day. As per the Ministry of Environment and Forest (MoEF), Government of India, the pulp and paper sector is in the "Red Category" list of 17 industries having a high polluting potential. Pulp and paper production is a major industry in India with a total capacity of over 3 million tons per annum (CPCB, 2001).

Pulp and paper industries is considered as one of the most polluter industry in the world and are categorized by the occurrence of colour and suspended solids, organic and inorganic compounds, bad smell, high concentration of nutrients that cause eutrophication of receiving waters bodies, and high toxicity overall.Such effluents have the potential to adversely affect the receiving aquatic environment through, for instance, slime production by microorganisms such as *Sphaerotilus* sp. These factories commonly produce considerable amounts of wastewaters, especially from virgin raw materials processing and formation of scum, as well as toxicity to the exposed communities, thermal impacts, colour problems, and aesthetical issues (Pellegrin *et al.,* 1999; Thompson *et al.*, 2001; Lacorte *et al.,* 2003; Pokhrel and Viraraghavan, 2004; Sumathi and Hung, 2006).

The pulp and paper mill produces large quantities of wastewater which needs suitable treatment prior to discharge in to the environment; otherwise it represents a significant consequence on the environment and health. The major challenging concern is the insistent dark brown colour due to lignin content and its derivatives, such as chloro lignin present in the effluent discharged from the pulp bleaching process (Prasongsuk *et al*., 2009).The potential recalcitrant compound which are released during the treatment of pulp and paper mill effluents that may the harmful effect on the environment and health (Table 1).

Characteristics of pulp and paper mill effluents: The pulp and paper mill effluent influenced by the type of manufacturing method used in the process and the extent of reuse of water recycled in industry. Effluent from kraft pulping is enormously polluted and categorized by parameters such as colour, adsorbable organic halides (AOX) and associated recalcitrant organic compounds. The major complications of pulp and paper mill effluent is the brown coloured effluent due to lignin and its derivatives. This effluent comes out from pulping, bleaching and chemical recovery phases. The brown colour of the effluent may increase water temperature and decrease photosynthesis, both of which may lead to reduce the concentration of dissolved oxygen of the water bodies (Bajpai *et al.,* 1993; Ragunathan and Swaminathan, 2004) (Table 2).

Low cost adsorbants and their applications: Adsorption is one of the physico-chemical treatment technique found to be very effective for wastewater treatment because of its simplicity and cost effectiveness. Adsorbent can be measured as low-cost if it is rich in nature, needs slightly processing and is a by-product of waste substantial from waste industry. The common adsorbents include activated carbon, molecular sieves, polymeric adsorbents, and some other low-cost materials are inexpensive as they have no or very low economic value. With the convenience and cheapness of different adsorbent materials such as industrial by-products, agricultural wastes and other natural waste materialsused as low-cost adsorbenthas become popular now days. In adsorption process, thermodynamic and kinetic aspects should be elaborated to know more details about its concert and mechanisms. Except for adsorption capacity, kinetic concert of a given adsorbent is also the great importance for the pilot application (Qiu *et al.* 2009). In the process of adsorption, the gas liquid solute gathers on the adsorbent surface which forms a molecular film of the adsorbent. For the removal of metal ions and other recalcitrant compound from the industrial effluents, activated charcoal, synthetic resins, agricultural waste and industrial waste used as adsorbent followed by physical, biological, and chemical methods (Yadanaparthi *et al*., 2009, Kwon *et al.,* 2010 and Gottipati *et al*., 2012).

Activated carbon: Activated carbon is the commonly preferred adsorbent; its widespread use is restricted due to high cost. The preparation and regeneration of low cost activated carbon in place of commercially available adsorbents. Several works has focused on various natural solid supports, which are able to remove pollutants from contaminated water using low cost activated carbon. Activated carbon is a crude form of graphite highly porous with variable pore sizes, noticeable cracks and crevices. Active carbon is prepared from coconut shells, wood char, lignin, petroleum coke, bone-char, peat, sawdust, carbon black, rice hulls, sugar, peach pits, fish, fertilizer waste, waste rubber tire, etc. (Mohan and Pittman, 2007) (Table 3).

Activated carbon has been shown to be an effective adsorbent for the color removal from the wastewater. Activated carbon in industrial treatments is more applicable with other technologies for example with Dissolved air flotation(DAF). DAF is used to remove the carbon from solution. Adsorption media cost and sludge production are factors limiting the application of this technology to the pulp paper industry. The carbon is activated usually by heat or chemical treatment; the adsorption is often so strong that the carbon has to regenerated or even discarded. (Diez *et al.* 1998 and Wingate. 2002). By chemical activation, activated carbon was prepared from paper mill sludge by using K_2CO_3. The results showed that the surface area at the experimental conditions is achieved to 908 m^2 g^{-1}. In addition to these advantages of preparation, the porous properties of activated carbon are outstanding because of the well-developed micro-pore. The adsorption isotherm data were fitted to two adsorption isotherm models and found to closely fit the Freundlich model with R^2 equal 0.99 at pH 8, indicating a multilayer of adsorption. Paper sludge is a promising low cost precursor for the production of activated carbon and have the maximum retention capacity of the produced activated carbon was 280 mg/g. (Nasr *et al.,* 2017).

Agriculture waste materials: The utilization of agricultural waste material scan be seen as having a great potential to be developed and used as a low cost sorbent for the treatment of effluents from the various industries. The possibility of these materials could be beneficial not only for the environment in resolving the solid waste disposal problem, but also the economy useful for the wastewater treatment (Upadhye and Yamgar, 2016).Various agricultural waste materials as adsorbents are being studied for the removal of different dyes from aqueous solutions at different operating conditions (Table 4).Several agriculture waste materials are accessible in environment in which adsorption properties have been reported e.g rice husk, saw dust, tea and coffee waste, orange peel peanut shells, activated carbon, dry tree leaves and barks (Asma *et al.,* 2005; Ferda and Selen, 2012; Kishore *et al*., 2008; Nuria *et al.,* 2010).

The efficiency of sawdust in the removal of Cu^{2+} and Zn^{2+} ions was conducted by Sciban *et al.* (2006). The adsorption capacities were compared with the untreated sawdusts, poplar and firwood were treated with NaOH (fibre-swelling agent) and Na_2CO_3 solutions. After treating with NaOH, increase in adsorption capacity was observed to be increase to 2.5 times for Cu^{2+} and 15 times for Zn^{2+}. The adsorption capacities shown by Langmuir model were 6.92 mg g^{-1} (poplar sawdust) and 12.70 mg g^{-1} (fir sawdust) for Cu^{2+}, and 15.83 mg g^{-1} (poplar sawdust) and 13.41 mg g_1 for Zn^{2+} (fir sawdust), respectively. Parab*et al.* (2006) observed the maximum adsorption capacity of coir pith was 12.82, 11.56 and 15.95 mg/g for cobalt, chromium and nickel, respectively. Optimum pH values for maximum metal ion adsorption were found as 4.3 for cobalt, 3.3 for chromium and 5.3 for nickel (Table 4).

By using Crude Pongamia Leaf Powder (CPLP) and modified with nitric acid Pongamia Leaf Powder (APLP) shows removal of Cr(VI) at pH 2.0 and contact time 165 minutes. APLP was found to be more efficient than CPLP with an initial Cr(VI) concentration of 5mg/l (Shivamani and Prince, 2008).Theivarasu and Mylsamy (2010)was studied the adsorption of Rhodamine-B dye using char prepared from the coconut shell with concentrated sulfuric acid at ratio of 1:1 (w/v). The activation was executed by heating in a muffle furnace at 550 °C for 7 hours, followed by washing and drying. The treated coconut shell char was found 41.67 mg/g adsorption capacity. Kulkarni and Kaware (2015) have examined the removal of metal ions in a packed bed adsorption column using rice husk as cadmium sorbent. It was observed that increase in the initial cadmium concentration from 10 mg/l to 50 mg/l decreased exhaustion time from 830 min to 570 min and break point time decreased from 330 min to 120 min. The adsorption efficiency was obtained as 69% for optimum conditions operating conditions (bed height, initial metal ion concentration, flow rate, and pH values) were found to be 50 cm, 30 mg/l, 60 ml/min, and 6, respectively (Table 4).

Industrial wastes and sludge as adsorbents: Widespread industrial activities produce huge amount of solid waste materials as by-products. Some of this material is being put to use while others find no proper utilization and are discarded in the different places. The industrial waste material is available without any cost and causes major disposal problem to the environment. If these industrial solid wastes could be used as low-cost adsorbents, the volume of waste materials and pollution load could be decreased at reasonable cost (Bhatnagar, M. Sillanpa, 2010). Various industrial wastes as adsorbent have been discovered with or without treatment as adsorbents for the removal of pollutants from wastewater. Mendez*et al.* (2009) studied the % reduction of $Cu^{2+}$from water using adsorbent generated from paper industry waste (de-inking paper sludge and sludge from virgin pulp mill). Experimental results revealed that the adsorbent prepared from the virgin pulp is of microporous nature whereas the deinked sludge based adsorbent is microporous in nature. Adsorbent materials remained then used for Cu^{2+} reduction from water at acidic pH. Final pH noticeably improved after treatment of water with adsorbent materials probably due to their raised $CaCO_3$content. In common, highest Cu^{2+}removal were attained using adsorbent prepared from de-inking paper sludge (Table 5).

Paper industry also produces black liquor, a waste product originated from paper industry, was examined for the adsorption of Pb^{2+} and Zn^{2+} by Srivastava *et al*. (1994). An adsorption capacity of 1865 and 95 mg/g was reported for Pb^{2+} and Zn^{2+}, respectively, at 40 ◦C. The adsorption of phenols on paper mills sludges was studied by Calce *et al.* (2002). The retention capacity of paper mill sludges was observed in the order: 2-nitrophenol = 4-nitrophenol_2-chlorophenol < phenol < 4-chlorophenol≤3–chlorophenol< 2,4-dichlorophenol < 3,4-dichlorophenol = 2,4,5-trichlorophenol < 3,5-dichlorophenol.Papermill sludge was also examined for the removal of orange Gdye (an anionic dye) from aqueous solutions (Bhatnagar *et al.,* 2007).

Aksu and Yener (1999) investigated the potential of fly ash as a alternate for activated carbon for phenol adsorption. At initial phenol concentration of 100 mg/L, the maximum phenol adsorption efficiency was obtained 27.9 mg/g for fly ash and 108.0 mg/g for granular activated carbon.

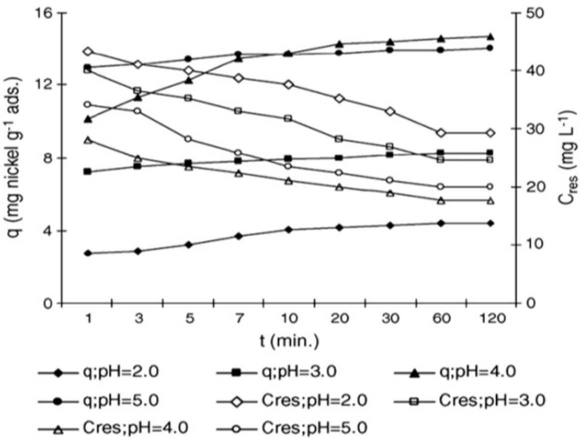

Fig. 1. *Effect of pH on Ni(II) uptake and residual concentration by waste tea (adsorbentdosage=10 g/L, agitating rate=360 rpm, C0=100 mg L−1) (Malkoc E, Nuhoglu, 2005).*

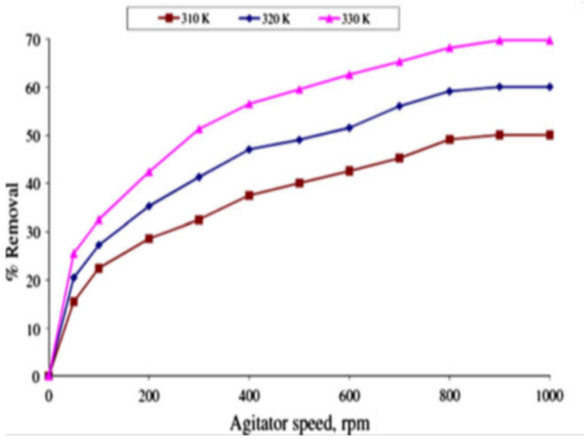

Fig. 2. *Effect of agitation speed with various temperatures (Patel and Vashi, 2010).*

The effectiveness of fly ash in removal of phosphate from aqueous solution and its related mechanism was studied by Lu *et al.*(2009). Results disclosed that 68–96% phosphate removal in the first 5min by fly ash. This is due to fast kinetics and high removal on behalf of precipitation, then a slower and longer removal due to adsorption.

Dried activated sludge has been explored for the sorption of phenol, o-chlorophenol and p-chlorophenol from aqueous solutions 1998. The maximum sorption capacity of dried activated sludge was found to be 86.1 mg/g for phenol, 102.4 mg/g for o-chlorophenol and 116.3 mg/g for p-chlorophenol at 100 mg/L initial pollutant concentration. Selvaraj *et al. (*2003) examined the potential of distillery sludge for Cr(VI) removal and described the Langmuir adsorption capacity of 5.7 mg/g for Cr(VI). Li et al. (2004) used wine processing waste sludge as an effective adsorbent for Cr(III) removal. Adsorption dynamics had been fruitfully described by the Lagergren model and intra particle diffusion model. The sewage treatment plant biosolids (sludge) was used as adsorbent in removing basic dyes, Basic blue 3,Basic red 22 and Basic black 9 from aqueous solutions (ZahangirAlam, 2004). Batch mode adsorption results are explored by varying different condition like contact time, initial dye concentration, adsorbent dose, agitation rate, temperature and pH. The results indicated that the adsorption capacity of basic dyes was higher (22-24 mg/g) with the lower values of the temperature (25-30°C), adsorbent dosage(0.5-0.75% w/v), higher values of the initial pH (8-9)and agitation rate (150-200 rpm). The equilibrium in the solution was observed within 2 h of operation (Table 5).

Factors affecting the adsorption

Effect of pH: The pH is the essential to the adsorption process and has precious not only adsorption capacity but also surface charge of the adsorbent, the degree of ionization of the chemistry of the dye solution. Methylene blue (MB) has a pK value of 3.8 with the cationic properties of the dye and (Kim*et al*. 2013).The H+ ions combine with the-ve charged adsorbent surface (oxides of aluminum, calcium, silicon, iron, etc.) and neutralize them (Devi et al., 2008)and thereby reduce the hindrance to the diffusion of organic molecules. The lower adsorption at higher pH might be possible due to the presence of OH- ions. These OH- ions cause hindrance to the diffusion of organic (contributing to COD) ions (Das & Patnaik, 2001).

According to the mechanism and the discussion of pH effect, the adsorption will lead to a decrease in pH as equivalent H+ will be released along with the adsorption. This is the case for most metal adsorption, but there is always exception. Some metals existing as negative species in solution, such as hexavalent chromium, may release hydroxide (OH−) instead of proton (H+) when they are adsorbed by waste materials, and therefore result in an increase in pH. Fig. 1 indicated that the effect of pH on Ni(II) uptake and residual concentration by waste tea (Malkoc E, Nuhoglu, 2005). Hydrogen ion work as a spanning ligand between the adsorbent wall and the dye molecule (Aksu and Tezer, 2005).At high pH,decreases the positive charge at the solution interface and the adsorbent surface seems negatively charged (Özcan *et al.* 2007).

Effect of agitation speed and contact time: According to Farhan *et al.*(2013) reported that at lower time concentrations (1-4hrs), *Cedrusdeodara* shows very sharp decrease in color ranging from 56% to 100%. *Dalbergiasissoo* has almost constant rate of reaction. Whereas Eucalyptus spp., show less reduction from 1- 3hrs but reduction shoots up sharply at 4hrs time duration, further increase from 4hrs to 6hrs does not significantly increase color reduction. Gupta *et al.,* (2011) also attained equilibrium of 60 min for Acid Blue 113 dye uptake .The decrease in dye reduc-

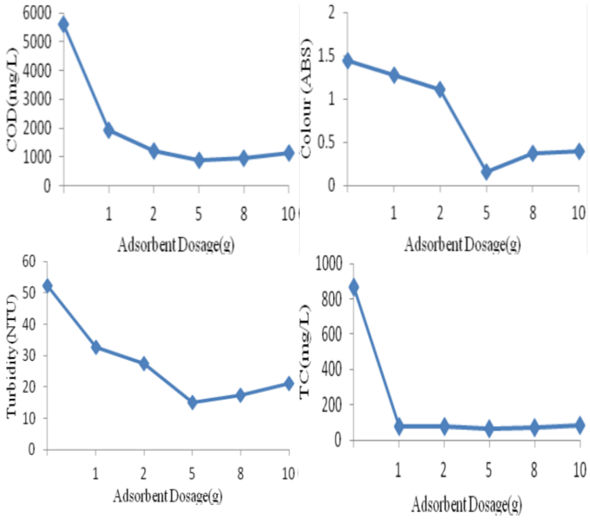

Fig.3. *Effect of Adsorbent Dosage on COD, colour, turbidity and total carbon removal from the paper mill effluent (Shivayogimath and Joshi, 2015).*

tion with time might be due to accumulation of dye molecules around the activated carbon. There will be no more reduction in COD, after the equilibrium point has reached. Infect at equilibrium the rate of adsorption of pollutant onto the activated carbon is almost equal to the rate of desorption (Mohan *et al.,* 2008).

The effect of contact time carried out by using the adsorbent–adsorbate solution with fixed adsorbent dose and initial dye concentration for different time intervals and shaken until equilibrium. Generallyt he rate of removal of dye increases with an increase in contact time to a certain extended period.Further increase in contact time does not increase the reduction of dye due to deposition of dyes on the available adsorption site on adsorbent material (Ansari and Mosayebzadeh, 2010). Patel and Vashi (2010) described that the percentage of crystal violet adsorption increased up to 15.5–50.0,20.5–60.0 and 25.5–69.7 % respectively with increasing the agitation speed at 50–900 rpm and different temperatures (310, 320 and 330 K), but then gradually approaches a constant value after 900 rpm (Fig.2).

Effect of adsorbent dose: The three activated carbons differ significantly in their COD capacity. This decrease in COD uptake capacity with increase in dose of activated carbon may be due to the formation of clusters of carbon particles resulting in decreased surface area (Nagda, 2006). At low concentration, the interaction between solute and solvent increase, so the solute has low tendency towards activated carbon and high affinity towards solvent (Nadeem *et al.,* 2006).Sharma and Uma (2010) conducted the experiment by taking different adsorbent doses (0.40–0.60 g) in 50 mL of dye solution in order to find the effect of different doses of rice husk cultivated carbon on the removal of methylene blue. The results showed that the removal increased from 86.75to 99.83 % with increasing adsorbent dose from 0.40 to0.60 g (fig. 3).

According to Uddin *et al.,*(2017) reported that the removal of dye was increased from 82 to 99% with an increase in adsorbent dose from 0.1 to 0.8 g. The increase in the dye removal percentage with adsorbent dosages could be attributed to increase in the surface area of adsorbent. Adsorption capacity was decreased from 164 to 25 mg/g when the adsorbent dose was increased from 0.1 to 0.8 g. The reduction in adsorption capacity qe (mg/g) with increasing adsorbent dose was due to the split in the flux or the concentration gradient between solute concentration in the solution and in the surface of the adsorbent.

Effect of particle size: Decrease inparticle size would lead to increase in surface area and then increase in the adsorption capacity at the outer surface of the waste materials. Besides adsorption at the outer surface of the waste material there is also adsorption of intra-particle diffusion from the outer surface into the pores of the material. Due to various factors, such as diffusional path length or mass transfer resistance, contact time and blockage of some diffusional path, most of the internal surface of the particle may not be utilized for adsorption; consequently the adsorption efficiency may become low (Ahmaruzzaman, 2011). Kulkarni *et al*., (2013) observed that as the particle size decreases from mesh number 36 to 72, the percentage removal was increases. This is because of the increase in the surface area available for adsorption. The removal percent remains constant for the particle sizes finer than 72 mesh no. particles. The adsorbate is not able to utilize the extra surface area due to lack of contact with adsorbent .

Amarasinghe and Williams (2007) was studied the effect of particle size of tea waste on the adsorption capacity of heavy metals. The results showed that for tea waste particles of mean size 1250, 925and 575 μm, the removal of Cu ion were 41%, 53% and 57% respectively. Batch adsorption experiments were also carried out to examine the effect of particle size of bagasse fly ash on the removal of Pb and Cr (VI) from their aqueous solution. The different particle sizes, 100–150, 200–250, and 300–350 μm at pH 6.0 for lead and 5.0 for chromium, with an adsorbent dose of 10 g/L, shaking time 80 min for lead and 60 min for chromium at 30 °C. The adsorption of lead and chromium was found to be 99.9, 95.0, and 88.0%, respectively using the above mentioned sizes (Gupta and Ali, 2004).

Conclusion

The pulp and paper industry is a complex activity which involves many different processes and products. Pollutants from the pulp and paper mill effluents are highly colored, mainly due to the presence of chromophoric compounds from wood extractives, lignin derivatives and organochlorine compounds, all recalcitrant. In this review various, low cost adsorbent such as agriculture waste product, activated carbon and industrial waste and sludge as adsorbent has been presented and also discussed the factor affecting the adsorption process. The use of these low-cost adsorbents suggests that these are the relatively cheap , easily available, renewable and show highly affinity for the removal of recalcitrant compounds from effluents of pulp and paper mills and the effluents from the various processes and operations of these industries. Adsorption is an innovative treatment option to improve the efficiency and increasing the possibility of discharge those wastewaters into the receiving bodies without causing any damage. Further research is to be focussed to make the process economically strong at industrial scale with the special reference to recovery of the metals and other recalcitrant compounds from the industrial wastewater.

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