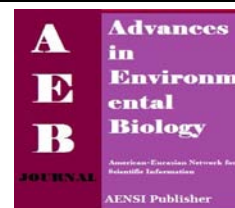




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## Sorptive Removal of Dyes from Aqueous Solution: A Review

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### ABSTRACT

In this article, adsorption as an effective method for removing dyes from wastewater has been reviewed. It is clear from the literature survey that adsorption can be achieved through different materials such as commercial activated carbon, naturally occurring materials, synthetic polymer, waste material and activated carbon from waste materials. However, there is need to study more on the effect of other pollutants on adsorption of dyes so as to reduce its environmental and health effects.

#### Keywords:

Activated carbon, Biosorption, Dyes, Pollutants, Synthetic polymer.

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## INTRODUCTION

Most industries especially textiles, paper, plastics, leather, food, cosmetics etc use dyes to colour their final product. Such extensive use of dyes poses problems in form of coloured wastewater which require pretreatment for colour prior to disposal into receiving water bodies [104]. This is because this dye-bearing wastewater impart toxicity to aquatic life and damage the aesthetic nature of receiving water bodies environment [26]. Dyes are synthetic aromatic water soluble, dispersible organic colourants, having potential application in various industries.

Dyes, once released into water bodies not only produce toxic amines by the reductive cleavage of azo linkages which causes severe effects on human beings through damaging the vital organs such as the brain, liver, kidneys and reproductive systems but also prevent photosynthetic activity in aquatic life by reducing light penetration [56,71]. The removal of colour from effluent is often more important than the removal of soluble colourless organics which normally contributes to the major BOD load. Visual pollution is a serious problem in water quality, it is not easy to accept red or brown rivers [12]. Many of the dyes used in industries are stable to light and oxidation and so they are considered as non-oxidizable substances because of their complex structure and large molecular size [26]. Due to all these reasons, removal of dyes from effluents becomes environmentally important. As a result of serious efforts of researchers all over the world, a number of methodologies with varying degrees of success have been developed. They are coagulation, foam floatation, filtration, ion exchange, sedimentation, solvent extraction, electrolysis, chemical oxidation, disinfection, chemical precipitation and membrane process. All these methods have shortcomings and limitations. For example, coagulation requires pH control and causes further problems of sludge disposal and ozonation does not minimize chemical oxygen demand.

They are also less effective especially for the removal of brightly coloured water-soluble reactive and acid dyes because these dyes show resistance to many chemicals, oxidizing agents and light [47]. Adsorption techniques appear as successful efficient alternatives for the removal of dyes in waste water [30]. Adsorption process has an edge over other methods due to its sludge free operation and complete removal of dyes even from dilute solutions. Biosorption process is a type of adsorption which utilizes not only plant materials but also a wide variety of micro-organisms in dead, pretreated and immobilized form as adsorbing agents. These materials are cheap to produce and carry wide range of binding sites for dye molecules [104,32,36]. In adsorption, the chemical properties of surface groups influence the adsorption equilibria to a large extent [12].

The term 'adsorption' was proposed by Bois-Reymond and it has been widely used for the removal of solutes from solutions and gas from air atmosphere. The extent of adsorption depends on the nature of adsorbent especially its porosity and surface areas. The important characteristics of a good adsorbents is its high porosity and

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consequently a larger surface area with more specific adsorption sites. A better adsorbent is the one with large surface area and which requires less time for adsorption equilibrium. Hence, one generally looks to adsorbents with high surface area and faster kinetics for the removal of dyes. In this study, various adsorption / biosorption methods of dye removal has been reviewed. The main aim of this review is to provide recent information concerning the sorptive removal of dyes.

The review;

- i. presents various adsorbents available for adsorption
- ii. discusses various mechanisms involved in the sorption process
- iii. presents adsorption capacities of various sorbents available

## 2.0. Category of adsorbents used in sorption process:

- 2.1 Activated carbon
- 2.2 Naturally occurring materials
- 2.3 Synthetic polymers
- 2.4 Agrowaste materials
- 2.5 Industrial waste
- 2.6 Activated carbon from agrowaste materials
- 2.7 Biosorbents

### 2.1. Activated Carbon:

This is the most popular and widely used adsorbent in effluent treatment. Charcoal is the source of commercial activated carbon and has been used to purify water as far back as 2000 B.C. It is produced by a process of raw materials dehydration and carbonization followed by activation. The material is dehydrated and carbonized by slowly heating in the absence of air. Activated carbon exists in two forms namely; (i). powdered activated carbon(PAC) and (ii). granular activated carbon (GAC). The granular form is preferable since it is more adaptable to continuous contraction and there is no need to separate the carbon from the bulk fluid.

### 2.2 Naturally Occurring Materials:

These are substances that have not been processed but are available in nature. They are abundant and cheap. Examples are clay, stone, soil, chitin, chitosan, gypsum, zeolite, bentonite etc.

### 2.3 Synthetic Polymer:

These are materials processed to suite the purpose of adsorption. For example, cyclodextrin is a polymer produced by crosslinking of starch with a coupling agent (e.g chitosan) to form water-insoluble networks [24].

### 2.4. Agrowaste Materials:

These are waste materials from forest and poultry industries. They are available in large quantities and may have potential as a sorbent due to their physico-chemical properties and low cost. They contain various organic compounds such as lignin, cellulose and hemicellulose with polyphenolic groups that might be useful for binding dyes through different mechanisms. Examples are sawdust, wood, hen feather, sheep wool, corn cob, rice husk, coir pith, rice bran, wheat bran etc [24].

### 2.5 Industrial Waste:

These are materials collected after a particular product processing. They are otherwise called spent materials or by-products. Examples are carbon nanotubes, fly ash, bagasse, ash, sludge etc.

### 2.6 Activated Carbon From Agrowaste Materials:

These are substances produced by carbonization of lignocellulosic materials. They are produced by placing the wastes in the furnace at a temperature of about 450°C to drive away all other component except carbon. Materials that can be used are coconut shell, rice husk, sawdust, coir pith, date pits, bamboo, cassava peel, nutshells etc

### 2.7 Biosorbents:

These are materials called biomass (dead or living). They are mainly micro-organisms which include algae, yeasts, bacteria and fungi. Microbial biomass is produced in fermentation process to synthesize valuable products such as enzymes but the by-products generated can be used in biosorption. Examples are *Aspergillus niger*, *Chlorella vulgaris*, *Rhizopus arrhizus*, *Spirodela polyrrhiza* etc.

### 3.0 Sorptive Removal of Dye by Various Adsorbents:

#### 3.1 Removal of dye By Commercial Activated Carbon:

Voudrias *et al.* (2002) studied the adsorption of three dyes namely Cibacron Blau F-R, Reactive blue 21 and Cibacron Rot F-B using commercial activated carbon. It was found that adsorption isotherm fitted Langmuir best indicating that the sorption was localized in a monolayer. Also, the equilibrium time was observed to be 72 hours and the percentage removal was higher at lower pH. Adsorption of basic red 14, reactive red 24 and acid blue 113 on granular activated carbon was investigated by Faria *et al.* [30]. It was observed that the adsorption data of both the reactive red and acid blue fitted both Langmuir and Freundlich models well while that of the basic red fitted Langmuir only.

Wang *et al.* [102] investigated the adsorption of methylene blue on activated carbon. It was found that higher pH of the solution results in higher adsorption capacity and the kinetic studies follows the pseudo-second order model. Yavuz and Aydin [107] studied the removal of direct dyes (direct yellow 50, direct red 80 and direct blue 71) from aqueous solution using commercial activated carbon. It was found that the equilibrium time was 40 minutes and the isotherm data fitted the Langmuir model. Powdered Activated Carbon was used for adsorption of Reactive Black 5 by Eren and Acar [29]. It was observed that as the initial dye concentration increased, adsorption efficiency decreased and that increase in adsorbent dose resulted in higher percentage removal of dye. Also, the adsorption data followed Langmuir isotherm with maximum adsorption capacity 58.823 mg/g and the kinetic data followed pseudo-second order model.

Decontamination of textile wastewater containing methylene blue, basic yellow and JK2R by Powdered Activated Carbon was investigated by Barka *et al* [14]. It was found that the equilibrium time for methylene blue, basic yellow and JK2R were 60, 90, 120 minutes respectively. Also, adsorption increases as the activated carbon dosage increases and the isotherm data followed Langmuir model with maximum adsorption capacities of 294.12, 322.6 and 222.2 mg/g for methylene blue, basic yellow and JK2R respectively. The effect of ionic strength on these dyes showed that adsorption of methylene blue and basic yellow increases as the ionic strength increases while that of JK2R decreases as the ionic strength increases; the effect of pH also followed the same trend.

Dincer *et al.* [26] investigated the comparison of activated carbon and bottom ash for removal of reactive dye (Vertigo Navy marine) from aqueous solution. The extent of dye removal increased with decreased initial concentration of the dye and also increased with increased contact time. It was found that the adsorption of the dye onto GAC occurred faster and reached higher equilibrium levels as compared to bottom ash. Also, the GAC had a higher adsorption capacity than bottom ash at lower and higher dye concentrations.

Yamin Yasin *et al.* [106] studied the adsorption of methylene blue onto KOH treated and untreated activated carbon. The adsorption experiment indicated that both adsorbents were effective in removing methylene blue from aqueous solution. The percentage removal increased with increase in contact time and attains equilibrium at about 120 minutes for treated activated carbon while the untreated one was 180 minutes. Also, the adsorption increased with increase in pH and increase in adsorption dosage. The adsorption data fitted Langmuir isotherm well and the treated activated carbon was more effective having shown great reduction of colour and lesser contact time.

Mohd Din and Hameed [60] investigated the adsorption of methyl violet onto acid modified activated carbon. The experimental data was tested by various 2-parameter isotherm models and the order of fitness was found to be Freundlich > Temkin > Langmuir > Dubinin-Radushkevich. The values of the calculated standard Gibbs free energy,  $\Delta G$ , enthalpy,  $\Delta H^0$  and entropy,  $\Delta S^0$  showed that the adsorption was spontaneous, entropy-driven and endothermic in nature. It was also found that the mechanism was not only limited to physical adsorption but chemisorption too in the complex aqueous phase due to high activation energy.

Attia *et al.* [12] studied the capacity of activated carbon in the removal of acid dyes subsequent to its thermal treatment. It was found that irrespective of the thermal treatment, there is decreasing capacity in the order of acid blue > Acid Red >> Acid yellow. This was ascribed to the different chemical nature of the three dyes with respect to substituent and / or branched side chains that increase bulkiness and reduce diffusion ability. Suteu and Bilha [93] studied the equilibrium and kinetics of the reactive dye (Brilliant Red HE-3B) adsorption by commercial activated carbon. The adsorption data was analysed by Freundlich and Langmuir models but the Langmuir model fitted best. The thermodynamic value indicated that the process was endothermic. The kinetic data followed the pseudo-second order model which indicated that the intra particle diffusion was the rate limiting factor.

#### 3.2 Removal of Dyes by Naturally Occurring Materials:

In 2004, Khan *et al* investigated the removal of basic dyes (methylene blue, malachite green and rhodamine B) onto akash kinari coal. The adsorption data fitted both Langmuir and Freundlich models but Freundlich fitted best.

The negative values of  $\Delta G^{\circ}$  and  $\Delta H^{\circ}$  indicated that the adsorption process was spontaneous and exothermic. Juang *et al.* [45] investigated the adsorption of dye (Amido Naphthol Red G) on modified montmorillonite and organoclay. It was found that adsorption increases slightly at low and high pH values. The adsorption data fitted Langmuir best and the kinetic data could be well described by pseudo-second order model. The low value of  $\Delta H^{\circ}$  indicated the characteristics of physical adsorption. Alkan *et al.* [7] studied the removal of Acid Yellow 49 from aqueous solution by adsorption on sepiolite (a clay mineral). It was observed that the adsorption equilibrium was attained in 1 hour. Also, the adsorbed amount of the dye increased with increasing ionic strength and temperature but decreased with increasing pH and the isotherm data were reasonably correlated by Freundlich isotherm. The order of heat of adsorption corresponds to a physical reaction.

Gypsum, a natural material, otherwise known as plaster of paris was used by Rauf *et al.* [79], to remove methylene blue from aqueous solution. The adsorption data was analyzed by Langmuir, Freundlich and Temkin isotherm models. The maximum monolayer adsorption capacity was found to be  $36 \text{ mg g}^{-1}$ . The kinetic data was found to obey pseudo-second order model. Shian and Pan studied the adsorption of basic dyes (Basic Green 5 and Basic violet 10) from aqueous solution by activated clay and montmorillonite. The adsorption data could be well described by Langmuir-Freundlich equation. The kinetic data fitted the pseudo-second order model and  $\Delta H$  indicated that the process is endothermic. Yavuz and Aydin [107] investigated the removal of direct dyes (direct yellow 50, direct red 80 and direct blue 71) from aqueous solution using raw kaolinite and montmorillonite. It was found that adsorption increased as pH increase and the equilibrium was attained in 40 minutes. The adsorption data was well described by Langmuir model. Methylene blue was adsorbed from aqueous solution onto clay (Jordarian Tripoli) by Al-zaydien [4]. It was observed that the amount of dye adsorbed varies with initial pH, adsorbent dose, adsorbate concentration and contact time. The equilibrium data were found to fit the Langmuir isotherm well indicating monolayer adsorption on a homogenous surface. The maximum adsorption capacity was found to be  $16.6 \text{ mg g}^{-1}$ . The equilibrium time was found to be 120 minutes and the kinetic data followed pseudo-second order model. It can also be described by intra particle diffusion. The free energy change indicated the spontaneous nature of the adsorbent and confirmed the affinity of the adsorbents for methylene blue..

Chamargore *et al.* [20] studied the removal of methylene red from aqueous solution by treated and untreated kotta powder. It was found that the adsorption increased as the pH increased and it also increased as adsorbent dose increased. Also, the treated kotta powder showed higher adsorption efficiency than the untreated. Smaranda *et al.* studied the sorption of congo red from aqueous solution onto soil. It was found that the amount of congo red adsorbed increased with increasing dye concentration, temperature and contact time. The kinetic data followed pseudo-second order kinetic model and the intra particle diffusion studies indicated that the adsorption mechanism was not exclusively controlled by the diffusion step. The adsorption data was best described by Freundlich isotherm which indicated the heterogeneity of the sorption sites on the soil particle and thermodynamic analysis indicated that the system was spontaneous and exothermic in nature.

### 3.3 Removal of Dyes by Biomaterials:

Walker and Weatherley [101] studied the biosorption of Tectilon Blue (TB4R) using three strains of bacteria namely; *Bacillus gordonae*, *Bacillus benzeovorans* and *Pseudomonas putida*. Up to 19% of the decolorisation was found to be caused by biosorption of the dye onto the biomass. The reaction rate was found to be intermediate between zero and first order at dye concentration of 200-1000 mg/L.

Kumar *et al.* [51] investigated the adsorption of malachite green onto *Pithophora sp.*, a fresh water algae. The algae was used raw and some were thermally activated at  $300^{\circ}\text{C}$  for 50 minutes. The equilibrium data were analyzed by Freundlich, Langmuir and Redlich-Peterson isotherm models but was very well represented by Redlich-Peterson isotherm. It was found that the thermally activated adsorbent possess a higher sorption capacity of  $117.647 \text{ mg/g}$  than the raw one with  $64.4 \text{ mg/g}$ . The kinetic data followed the pseudo-second order model closely and the negative value of free energy change indicated that the sorption process was spontaneous. Waranusantigul *et al.* [104] investigated the kinetics of methylene blue biosorption by *Spirodela polyrrhiza* (giant duckweed). It was found that as the amount of sorbent increased, the percentage of dye sorption increased and the optimum pH was reached at pH 3. The dye removal time was influenced by the initial dye concentration and the process followed the first-order rate kinetics.

Supaka *et al.* [92] studied the microbial decolourisation of reactive azo dyes (Remazol brilliant orange 3R, Remazol black B and Remazol Brilliant violet 5R) in a sequential anaerobic-aerobic system. It was observed that the colour removal yield with the more complex diazo dye (Remazol black B) was, however, much lower than that obtained with the monoazo dyes (Remazol Brilliant Orange 3R and Remazol Brilliant Violet 5R). Also, it was found that a longer retention time of anaerobic phase may be therefore required to enhance the rate of elimination of black dye, Hence, decolourisation of Remazol Brilliant Orange 3R, Remazol Black B and Remazol Brilliant Violet 5R by

a mixed bacterial culture are most likely due to complete breakdown of the reactive azo dyes to form aromatic amines. Akkaya and Ozer [3] investigated the biosorption of Acid Red 274 on *Dicranella varia* (a moss). It was observed that the biosorption data fitted better to Langmuir model. The monolayer adsorption capacity obtained was  $2000\text{mgg}^{-1}$ . It was observed that the equilibrium intake increased with increasing initial dye concentration and the optimum dose was 0.5g. The kinetics was well described by pseudo-second order kinetic model. The values obtained in the thermodynamics indicated that the process was exothermic in nature and was a reversible process.

El Nemr *et al.* [27] investigated the removal of chrysophenine (direct yellow 12) from aqueous solution using *Ulva lactuca* (a marine green algae). It was found that the adsorption data fitted the Freundlich model best. Also, the kinetic data was consistent with pseudo-second order model. The maximum adsorption capacity was  $80\text{mgg}^{-1}$ . Biological decolourisation of dye solution containing malachite green by *Cosmarium sp.* (microalgae) was studied by Daneshvar *et al.* [25]. It was observed that the optimum pH was 9 and that the decolourisation rate increased with the temperature rise. The biosorption of anionic textile dyes (Reactive Black 8, Reactive Brown 9, Reactive Green 19, Reactive Blue 38, Reactive Blue 3) by nonviable biomass of fungi and yeast (*Aspergillus niger*, *Aspergillus japonica*, *Rhizopus nigricans*, *Rhizopus arrhizus*, *Saccharomyces cerevisiae*) was demonstrated by Kumari and Abraham [50]. It was observed that *Rhizopus nigricans* adsorbed at higher percentage of 90-96% in 15 minutes at  $20^{\circ}\text{C}$  and pH 6.0. The adsorption data fitted Langmuir for all the biosorbents. The maximum uptake capacity for the dyes was in the range of 112-204 mg/g for all the biomass.

The sorption of three basic dyes (Astrazon Blue, Astrazon Red and Methylene Blue) onto *Caulerpa lentillifera* (green macroalgae) was investigated by Marungueng and Prasad [58]. The results obtained revealed that the algae exhibited greater sorption capacities than activated carbon for the three dyes. The isotherms followed Langmuir model which suggested that the sorption was monolayer. The kinetic data obeyed the pseudo-second order kinetic model and the adsorption could more rapidly sequester astrazon red when compared with activated carbon. Jirasripongpun *et al.* [43] studied the decolourization and degradation of Reactive red 195 by *Enterobacter sp.*, *Serratia sp.*, *Yersinia sp.* and *Erwinia sp.* *Enterobacter sp.* was observed to remove dye up to 90% within 2 days of incubation. Aravindhan *et al.* [10] investigated the removal of basic yellow dye from aqueous solution by sorption on *Caulerpa scalpelliformis* (a green algae). The sorption equilibrium studies demonstrated that the biosorption followed Freundlich isotherm model, which implied heterogeneous sorption phenomenon. The sorption kinetics followed pseudo-second order kinetic model and maximum uptake was  $27\text{mg/g}$ . Also, the thermodynamic parameters suggested that the sorption process was exothermic, spontaneous and shows decreased randomness at the solid-liquid interface.

Isken *et al.* [42] studied the biosorption of Reactive black 5 dye by *Penicillium restrictum* (a filamentous algae). It was observed that the maximum biosorption capacity was  $142.04\text{ mg/g}$  under the optimum conditions of pH 1, contact time of 75 minutes, biomass dose of 0.02 g at initial concentration of 200 ppm at  $35^{\circ}\text{C}$ . Furthermore, the biosorption process was found to obey a pseudo-second order model and increase in temperature caused an increase on the biosorption capacity of the adsorbent. Revankar and Lele [81] studied the decolourisation of recalcitrant dyes (Amaranth Reactive Orange 16, Cibacron Brilliant Red 3B-1, Acid Red 106, Orange II, Brilliant Blue R) by an indigenous strain of white rot fungus *Ganoderma sp.* It was observed that the maximum decolourisation of (96%) amaranth was achieved in 8 hours and rate of dye decolourisation by the *Ganoderma sp.* was very high compared to the most widely used strains (*Trametes versicolor* and *Phanerochaete chrysosporium*). *Corynebacterium glutamicum* (a fermentation waste) was utilized for biosorption of Reactive Black 5 from aqueous solution by Vijayaraghavan and Yun [98]. The effect of pretreatment on the biosorption capacity of *C. glutamicum* toward RB5 was initially studied using several chemical reagents such as HCl,  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$  etc. It was observed that among these reagents  $0.1\text{M HNO}_3$  gave the maximum enhancement of RB5 uptake with capacity of  $175\text{ mg/g}$  at pH 1. The isotherm fitted Langmuir best with maximum uptake of  $419\text{ mg/g}$ . The kinetic data followed pseudo-second order models. The thermodynamic parameters indicated that the system was a spontaneous and endothermic process.  $0.1\text{M NaOH}$  solution was used to successfully desorb almost all the dye molecules.

El-Sersy [28] investigated the bioremediation of Methylene Blue by *Bacillus thuringiensis* 4G1. It was found that percentage decolourisation was 98.23%. Tahir *et al.* [95] removed a basic dye (methylene blue) by two biosorbents *Ulva lactuca* and *Sargassum*. It was found that the optimum dose was 0.2 g of the adsorbents and the equilibrium time was 25 minutes at an optimum pH of 7.0. The percentage removal of dye was 96% and the thermodynamic parameters showed that the process was spontaneous and endothermic. Remenarova *et al.* [80] studied the sorption of cationic dyes (Malachite Green, Auramine O, Thioflavine T) from aqueous solutions by moss *Rhytidiadelphus squarrosus*. The results showed that the equilibrium was reached within 1-2 hours. The equilibrium data fitted the Freundlich isotherm data well. *Aspergillus niger* (a fungus) and *Spirogyra sp.* (a fresh algae) was used as a biosorbent for the removal of a reactive dye (Synazol) from its multicomponent wastewater by Khalaf [49]. The biosorbents were subjected to autoclaving and gamma irradiation. It was observed that the

percentage removal of dye was higher in the biosorbents pretreated with autoclave than that of gamma-irradiation and this was also higher than the raw biosorbents. The autoclaved biosorbents exhibited maximum dye removal of 88% and 85% respectively at pH of 3, temperature of 30°C, dosage of 8g and at a contact time of 18 hours.

Li and Jia [53] investigated the decolourisation and biosorption of congo red by system rice hull-*Schizophyllum* sp. F17 under solid state condition in a continuous flow packed-bed bioreactor. It was observed that the rice hull-white rot fungi has a maximum decolourisation of 89.71%. A brown algae, *Cystoseira barbatala* was used in the biosorption of Methylene Blue by Caparkaya and Cavas [19]. It was observed that the optimum dose was 0.1g and the adsorption increased as the pH increased. The adsorption data fitted the Langmuir model best with maximum adsorption capacity of 38.610 mg/g and the kinetic data followed pseudo-second order closely. The energy values indicated that the adsorption process was physical in nature. Thermodynamic parameters showed that the adsorption process was endothermic and spontaneous. A deuteromycete fungus, *Alternaria solari* was used to biodecolourised Acid Violet 19 in aqueous solution by Ali and Muhammad [6]. The results showed that the percentage decolorisation was 88.6% at a dye concentration of 30 mg/L within a period of four days. Youssef *et al.* [108] studied the decolorisation of Malachite Green, Crystal Violet, Carbol Fuchson and Methylene Blue by a filamentous fungus (*Acremonium kiliense*). The percentage decolourisation after 48 hours was 96.0%, 90.7%, 92.4% and 73.1% for Malachite Green, Methylene Blue, Crystal Violet and Carbol Fuchson respectively.

### 3.4 Removal of Dyes By Industrial Waste:

Albanis *et al.* [5] investigated the removal of dyes (Acid Orange, Acid Yellow 23, Disperse Blue 79, Basic Yellow 28 and Direct Yellow 28) from aqueous solution by adsorption on mixture of fly ash and soil. It was observed that the removal of dyes decreased with increase in dye solution concentration from 10 to 50 mg/L. The percentage removal was 33.8%, 59.4%, 84.2%, 98.2%, 60.3% for acid yellow 7, acid yellow 23, direct yellow 28, basic yellow 28 and disperse blue 79 respectively. Rachakornkij *et al.* [75] studied the removal of reactive dyes (Remazol Black B RBB, Remazol Brilliant Blue R RBBR and Remazol Brilliant Red F3B RBRF3B) from aqueous solution using bagasse fly ash (a waste from the sugar industry). It was observed that the removal efficiencies of RBB, RBBR, and RBRF3B were 58.48-98.03%, 46.15-93.47% and 46.30-94.60% respectively. Also, the initial pH did not affect the removal and the percentage removal decreased with increasing initial concentration of the dye but increased with increasing adsorbent dose. The optimum contact time was 240, 300 and 240 minutes for RBB, RBBR and RBRF3B respectively. The adsorption data can be described by Langmuir isotherm model.

Sewage treatment plant biosolids was used for the biosorption of three basic dyes (Basic Blue3; BD-Blue, Basic Red 22; BD-Red and Basic Black 9; BD-Black) by Alam. The results showed that the higher biosorption capacity (22-24 mgg<sup>-1</sup>) of the three dyes was observed with the variation of process factors such as contact time of 2 hours, pH of 8-9, adsorbent dosage of 0.5g, temperature of 25-30°C and agitation rate of 150-200 rpm. Also, it was observed that basic blue 3 was adsorbed well by the adsorbent compared to the two other dyes and the equilibrium data were slightly better fitted by Freundlich isotherm for BD-Blue and BD-Red while Langmuir fitted better for BD-Black. Bioadsorption of methylene blue dye on sugarcane bagasse was studied by Raghuvanshi *et al.* [76]. The adsorption potential was investigated in batch experiment in two different forms of the adsorbent i.e. raw and chemically activated. It was observed that the removal was better and more effective with chemically activated form in comparison to the raw form. The equilibrium data fitted the Freundlich isotherm well.

Saiful Azhar *et al.* [82] studied the removal of dye (methyl red) from aqueous solution on treated sugarcane bagasse. The adsorbent was treated with sulphuric acid and formaldehyde. It was observed that the adsorption efficiency was higher in sulphuric acid treated than formaldehyde treated adsorbent. Also, the initial pH of 6-10 favours the adsorption process. The use of jute processing wastes for treatment of waste water contaminated with dye was investigated by Banerjee and Dastidar [13]. On comparing the adsorption efficiency of the adsorbents with powdered activated carbon and granular activated carbon, it was observed that the maximum percentage removal of methylene blue by the adsorbent was 81.7% compared to that of PAC and GAC which was 61% and 40% respectively. The adsorption data conformed well to both Langmuir and Freundlich isotherm model. Liew Abdullah *et al.* [54] investigated the removal of azo dyes (methylene red) by sugarcane bagasse (treated and untreated). The adsorbent was treated with sulphuric acid and formaldehyde. The results showed that the adsorption of methyl red was dependent on the adsorbent dose and the dye concentration. It also showed that as the amount of the adsorbent increased, the percentage dye removed increased accordingly and higher adsorption percentages were observed at lower concentration of the dye. Furthermore, the sulphuric acid treated adsorbent showed a better performance compared to formaldehyde treated and raw adsorbent. The removal of composite reactive dye (Procion Brilliant Blue M-R, Procion Brilliant Red H-8G, Procion turquoise Blue M-G) by sewage sludge derived activated carbon was studied by Sreedhar Reddy *et al.* [94]. The results revealed that the adsorbed amount of dye increased as the adsorbent mass increased and the decrease in the pH of the solution leads to a significant increase in the adsorption

capacities of reactive dyes. The adsorbed amounts of reactive dyes increased with an increase in contact time and reached equilibrium in 120 minutes. The equilibrium data were analyzed using Langmuir and Freundlich isotherms but fitted Langmuir better.

The equilibrium and kinetics studies of adsorption of Reactive black 5 from aqueous solution using fly ash was done by Eren and Acar [29]. The adsorbent, fly ash, is a waste generated from thermal power plant. The adsorption data fitted Freundlich isotherm better, the kinetic data followed pseudo-second order model more closely and the monolayer adsorption capacity was 7.936 mg/g. Ju *et al.* [44] studied the biosorption characteristics of a reactive dye (Rhodamine B) onto dried activated sludge (from a municipal sewage treatment plant). In the experiment, three different pretreatment were given to the adsorbents viz sulfuric acid, hydrochloric acid and sodium hydroxide. It was observed that the adsorbent pretreated with sulfuric acid has the highest maximum adsorption capacity. The adsorption data was analyzed by both Freundlich and Langmuir isotherm model with Langmuir model showing the best agreement. The kinetic data obeyed pseudo second order kinetic model. Also, the adsorption capacity of the adsorbent increased with decreasing initial pH and temperature.

Bokbunja seed waste (from wine making industry) was utilized in adsorption of Reactive Blue 4 by Binupriya *et al* [18]. The waste was used to prepare carbon which was used for the adsorption process. Part of the carbon was treated with n-hexane. It was found that the hexane treated adsorbent was more efficient in removing the dye. Also, optimization of process parameters showed that pH 2 and contact time of 120 minutes was favourable for maximum adsorption. Langmuir isotherm described the equilibrium data well and the kinetic data showed a better fit to pseudo-first order model. The removal of basic (Basic blue 3) and reactive dye (Reactive orange 16) using quarternised sugarcane bagasse was investigated by Wong *et al* [105]. In the experiment, the natural adsorbent was compared with quarternized one in single and binary solution of the dye. It was observed that the percentage removal of the dye was higher in binary solutions than in single solution for both natural and quarternized adsorbent. The optimum pH for the sorption of the dye solutions was found to be in the range of 6-8. The study showed that the adsorption of the dyes fitted the pseudo second order model better. Furthermore, the sorption isotherm indicated that BB3 obeyed Freundlich isotherm in both binary and single system while RO 15 followed Langmuir in single and Freundlich in binary system. Also, the sorption of RO16 increased with increasing agitation rate while it has minimal effect on BB3 and the sorption of BB3 was favourable with increasing temperature while the sorption of RO16 was favourable at lower temperature. Conclusively, the optimum sorbent dosage for the dye sorption was 0.10g.

Shahryari *et al.* [86] studied the adsorption of methylene blue from aqueous solution onto carbon nanotubes (a waste product from petroleum industry). Experimental results showed that the amount of dye adsorbed increased with increasing initial concentration of dye, adsorbent dosage and temperature. The removal efficiency for adsorbent dose of 400 mg was more than 90% and was considered as optimum dosage. Also, the kinetic data were analyzed and it was found that pseudo-second order kinetic model was the most appropriate model describing the adsorption kinetics. The adsorption equilibrium was attained within 120 minutes. The equilibrium data was analyzed by Langmuir, Freundlich, Temkin and Sips isotherm models but the data were best described by Sips model with adsorption capacity of 132 mgg<sup>-1</sup>. The thermodynamic parameters indicated that the process was spontaneous ( $\Delta G$  is negative), endothermic in nature ( $\Delta H$  is positive) and showed increased randomness at the solid—solution interface ( $\Delta S$  is positive).

### 3.5 Removal of Dyes By Agrowaste Materials:

Kagbu *et al.* [46] evaluated the adsorption of dye residues of coloured textile effluent on yankassa sheep wool. It was found that increase in temperature of the effluent result in substantial increase in adsorption. It was also established that increase in pH of the effluent leads to decrease in the removal efficiency of the adsorbents. The use of rice husk (treated and untreated) for the adsorption of Direct Red 23 from aqueous solution by Abdelwahab *et al.* It was found that the adsorption was highly dependent on the contact time, adsorbent dose and dye concentration. The adsorption isotherm could well be described by Langmuir isotherm model with adsorption capacity of 2.415 mg/g for untreated and 4.35 mg/g for treated one. Kinetics for adsorption follows Lagergren first order kinetic model with film diffusion being the constitutive rate –controlling step.

The removal of acid dyes (Nylosane Blue, Erionyl yellow, Nylomine Red, Erionyl Red) from aqueous solutions using orange peel as a sorbent was investigated by Benaissa [17]. It was observed that the dyes sorption performance were strongly affected by parameters such as contact time, initial dye concentration and dyes type. The amount of dye sorbed by this material increased with the increase of these parameters at a specific time. The result also showed that the kinetics of these dyes sorption were well described by a pseudo-second order model. A good fitting of dyes sorption equilibrium data was obtained with Langmuir model in all the range of dyes

concentration studied. From the results, high maximum dyes sorption capacities were observed (65.88, 64.14, 62.07 and 40.72 mg/g for Nylosane Blue, Erionyl yellow, Nylomine Red, Erionyl Red respectively) using this material.

The adsorption of dyes (Methylene Blue and Basic Red 22) on prehydrolysed beech sawdust was studied by Batzias and Sidiras [16]. It was found that the adsorption data fitted the Freundlich isotherm well for both dyes and the kinetic data followed the Lagergren model. The monolayer adsorption capacity were found to be 9.78 and 20.16 mg/g for methylene blue and basic red 22 respectively. The uptake of cationic dyes (acridine orange, neutral red, methylene blue) from aqueous solution onto granular kohlrabi peel was investigated by Gong *et al* [35]. It was found that above the value of initial pH of 4, the three dyes studied could be removed effectively. The isotherm data fitted the Langmuir model in the case of Neutral Red with adsorption capacity of 112.36 mg/g and the Freundlich model for all the three dyes sorption. The processes followed the pseudo-first order closely.

Ncibi *et al.* [66] studied the kinetics and equilibrium of methylene blue adsorption by *Posidonia oceanica* fibres. The results showed that adsorption capacity was optimal using 6-9 solution pH range and by increasing the sorbent dose to 1g. The kinetic modelling study has shown that the experiment data were found to follow the pseudo-second order model suggesting a chemisorption process. The equilibrium sorption phenomena were found to be well described by both Langmuir and Redlich–Peterson models. The monolayer adsorption capacity was found to be 5.56mg/g. The adsorption removal of a reactive dye (Reactive red 228) using *Posidonica oceanica* leaf sheaths fibres was studied by Ncibi *et al* [67]. It was observed that the sorption capacity seems to be enhanced by increase in temperature and the maximum colour removal was observed at pH of 5. Pre-treating the adsorbent with  $H_3PO_4$  and  $HNO_3$  increased the adsorption efficiency up to 80%. Experimental kinetic data were found to follow Lagergren first order for raw adsorbents and pseudo second-order for the pre-treated ones. Equilibrium data were well represented by the Freundlich isotherm model for all the adsorption systems (both pre-treated and raw) and the thermodynamic study showed that the adsorption phenomenon was favourable, endothermic and spontaneous.

Kumar and Porkodi [50] studied the sorption of methylene blue using *Paspalum notatum*. It was observed that the amount of dye sorbed increased with increasing initial solution pH and the amount of dye uptake increased with increase in dye concentration and contact time. Also, the amount of dye sorbed decreased with increase in adsorbent dose and the equilibrium data was found to follow Langmuir isotherm suggesting the monolayer sorption of dye ions. Furthermore, the overall kinetics of methylene blue uptake by the adsorbent was found to followed pseudo-second order model and Boyd plot confirms the external mass transfer as the slowest step involved in the sorption process. The effect of initial dye concentration on the external mass transfer was represented in the form of dimensionless mass transfer number ( $Sh/Sc^{0.33}$ ).

Hen feathers was utilized for the adsorption of Indigo Carmine from simulated effluents by Mittal *et al* [59]. It was found that as the pH increased, adsorption decreased. The results revealed that the adsorbent exhibited almost 100% adsorption at lower concentration of  $1 \times 10^{-4}$  to  $3 \times 10^{-4} M$ , whereas at a dye concentration of  $10 \times 10^{-4} M$ , removal of 96.6, 91.9 and 87.9% of the dye takes place at 30, 40 and  $50^\circ C$  temperatures respectively. The equilibrium data was well described by Freundlich, kinetic data followed Lagergren first order model and it was controlled by film diffusion mechanisms at all the concentrations. The thermodynamic parameters indicated that there was spontaneity at higher temperatures, the process was endothermic in nature and there was good affinity of the dye towards the adsorbent material. Hamdaoui and Chiha [37] investigated the removal of methylene blue from aqueous solution by wheat bran. The results obtained suggested the sorbed amount increased with an increase in initial concentration of dye, solution pH and temperature. It was found that the dye recovery increased with an increase in sorbent dose and an increase of dye removal was obtained by a decrease of particle size and ionic strength in the aqueous phase. The sorption kinetics could be quite successfully fitted to a pseudo-second order equation with low value of activation energy suggesting that the sorption was an activated and physical process. Also, the applicability of both internal and external diffusion models showed that liquid-film and particle diffusion were effective sorption mechanisms.

The adsorption of dyes (Navy brown 3REL, direct red, procion red H8B, methylene blue, meta mega chrome yellow, lanasyn green 5GL, Solar violet and procion blue H5G) on sawdust phosphates was investigated by Prakash *et al* [73]. It was reported that percentage dye removal increased with decrease in pH. The equilibrium data fitted the Langmuir model best. The kinetics followed the Lagergren first order equation. Hashemian *et al.* [38] studied the sorption of acid red 138 from aqueous solution onto rice bran. It was observed that the dye sorption onto the adsorbent increased in the presence of inorganic salts and with a mesh size of 20 activated with saturated sodium chloride, it has higher adsorption capacity. Also, the optimum reaction time at a speed of 30 rpm was 60 minutes at initial pH of 2 and at room temperature, the dye was removed more effectively. The isotherm data fitted Freundlich model better. The kinetics and equilibrium studies of the sorption of Coomassie Brilliant Blue by Coir pith was studied by Naveen Prasad *et al* [65]. It was reported that the kinetic data followed the pseudo-second order model and the equilibrium data fitted Freundlich isotherm model better. The maximum adsorption capacity was found to be 31.847 mg/g and the results showed that the acid based adsorbent suited better for the removal of the dye.



Ofomaja and Ho [69] studied the effects of temperatures and pH on methylviolet biosorption by mansonia wood sawdust. The result showed that the sorption process was strongly dependent on solution pH and percentage dye removal became significant above pH 7, which was slightly higher than the  $pH_{pzc}$  of the adsorbent. Also, the equilibrium data fitted both the Langmuir and Redlich-Peterson isotherm model. The thermodynamic parameters result suggested that the process was spontaneous and endothermic. The removal of basic dyes (methylene blue and malachite green) from aqueous solution using agricultural by-products (wheat bran and rice bran) was investigated by Wang *et al* [103]. It was found that an increase in solution pH leads to a significant increase in the adsorption capacity. Also, the adsorbed amounts of the two dyes increased with an increase in contact time and reached equilibrium in 60-90 minutes and the removal efficiency and the adsorption capacity decreased with an increase in adsorbent dose. Furthermore, the adsorption capacity of both adsorbent towards the two dyes decreased with an increase in ionic strength and the system was particle-size dependent. The equilibrium data of methylene blue fitted better to Langmuir isotherm model while that of malachite green fitted Freundlich model. The kinetic data conformed better to pseudo-second order equation while the external diffusion was the rate controlling step of the initial fast adsorption ( $< 15$  minutes) and in the next stage the intraparticle diffusion dominated the mass transfer. Moreover, the thermodynamic analysis revealed that the adsorption behaviours of the two dyes could be considered as a spontaneous and physical process.

Nagda and Ghole [63] investigated the biosorption of congo red by hydrogen peroxide treated tendu (*Diospyros melanoxylon*) waste. Comparison was done on the extent of biosorption between untreated and treated forms of the adsorbents. It was reported that the adsorption occurred very fast initially and attained equilibrium within 60 minutes at  $pH = 6.2$  and the equilibrium attained faster after modification. The data fitted Freundlich isotherm model for treated ones while it fitted Langmuir for untreated ones and the maximum adsorption capacity was 46.95 and 134.4 mg/g for untreated and treated ones respectively. The kinetic data followed pseudo-second order rate equation. The removal of methylene blue from aqueous solutions using sawdust and sawdust coated with polypyrrole was studied by Ansari and Mosayebzadeh [9]. It was observed that comparing the data of untreated adsorbent with that of treated one, the adsorption by the former was not a pH dependent process at least from pH 4-10 and that there was higher sorption percentage for treated adsorbent at alkaline pH. It was also noted that the equilibrium isotherms i.e Langmuir and Freundlich were applicable with maximum monolayer adsorption capacity of 19.41 and 34.36 mg/g for untreated and treated adsorbent respectively. It was also concluded that the sorption isotherm using sawdust and polypyrrole sawdust followed the Freundlich and Langmuir models respectively and the dimensionless factor,  $R_L$  for both adsorption system revealed that the process was very favourable.

Annadurai *et al.* [8] prepared banana and orange peels as an adsorbent and used it to adsorb dyes (methyl orange, methylene blue, rhodamine B, congo red, methyl violet and amido black 10B). It was observed that an alkaline pH was favourable for the adsorption of dyes. Also, the Freundlich equation showed a somewhat better fit than does the Langmuir equation for adsorption of dyes using banana peel but the other way round for orange peels. Furthermore, it was found that the amount of dye adsorption increase with time and it remains constant after a contact time of about 65 minutes. The kinetic parameters followed Lagergren pseudo-first order equation and intra particle diffusion within the particle was identified to be rate limiting. Ho *et al.* [40] removed a basic dye (Basic Red 13) from aqueous solution using tree fern. It was found that the dye sorption capacity of adsorbent increased as the adsorbent particle size decreased. The isotherm data followed Langmuir with adsorption capacity of 408 mg/g. The thermodynamic parameters indicated that the system was spontaneous, endothermic and there was increase of randomness at the solid-liquid interface. Adsorption of water soluble organic dyes (Acridine Orange, Bismarck Brown, Crystal Violet, Malachite Green, Methyl Green, Nile Blue, Safranin O and Saturn Blue) on ferrofluid-modified sawdust was studied by Safarik *et al* [83]. The isotherm data followed four isotherm model namely Langmuir, Generalized Freundlich, Generalized Langmuir, Langmuir-Freundlich. The maximum adsorption capacities for the dyes ranged between 34 and 59 mg/g. Also, a change in pH value increased the adsorption of dye. Tendu leaves (an agrowaste) was utilized for removal of crystal violet from aqueous solution by Nagda and Ghole [62]. The adsorbent was used in raw form (TLR), dilute acid modified form (TLR-2N) and sulfuric acid carbonized form (TLR-CM). It was found that the adsorption data could be well described by Langmuir adsorption isotherm for the adsorbents and the kinetic data followed pseudo-second order kinetic. Interestingly, milder acid treatment of adsorbents (TLR-2N) enhanced adsorption whereas drastic acid carbonization of adsorbent (TLR-CM) resulted in reduced adsorption. The maximum adsorption capacities for crystal violet for TLR-2N, TLR and TLR-CM are 67.57, 42.92 and 22.47 mg/g respectively.

Tahir *et al.* [95] investigated the remediation of azo dyes (malachite green and methylene blue) by using household used black tea as an adsorbent. The adsorbent was used in raw form and impregnated form. It was observed that the adsorption of dye increased as the amount of adsorbent increased up to 0.7g for malachite green and 1.0g for methylene blue and the decreases. The adsorption percentage removal increased with increase in time

and reaches equilibrium in 10 minutes for malachite green and 25 minutes for methylene blue, then the values decreased and attains a constant value. Also, the data showed that there was decrease in the amount of the dyes adsorption with increase in temperature. The thermodynamic parameter showed that the process was exothermic and spontaneous. Furthermore, the result showed that the raw form of the adsorbent showed better adsorption capacity compared to its impregnated form.

Ong *et al.* [70] studied the equilibrium and continuous flow studies on the sorption of congo red using ethylenediamine modified rice hulls. It was observed that the equilibrium sorption was achieved in 240 minutes and the sorption kinetics obeyed pseudo second-order kinetics. The isotherm study showed that equilibrium sorption data conform to Langmuir isotherm model with maximum sorption capacity of  $26.4 \text{ mgg}^{-1}$ . The removal of methyl orange from solution using yam leaf fibres was studied by Vinoth *et al* [99]. The result revealed that the optimum pH was 3, equilibrium time was 45 minutes and the dosage of adsorbent increased as the adsorption increased. Also, as the initial dye concentration increased, adsorption also increased. The equilibrium data were best represented by Freundlich model and the kinetic data followed pseudo-second order model closely. An acidic dye (sunset yellow FCF) and a basic dye (malachite green) was removed by adsorption on chemically treated mangrove bark by Seey and Kassim [85]. It was observed that the optimum pH for malachite green and sunset yellow was 8 and 2 respectively. Also, the removal percentage of dye increased as the adsorbent amount increased and then becomes constant. The result from the contact time effect revealed that 4 hours of agitation time is sufficient to reach equilibrium when 100 mg/L of dyes concentration was employed and it was found that an increase in the dye concentration caused a decrease in the percentage of dye removal. The isotherm data fitted the Langmuir isotherm model best and the kinetic data followed pseudo-second order model closely.

Prasad and Santhi [74] studied the adsorption of cationic dyes (crystal violet and rhodamine B) from aqueous solution onto *Acacia nilotica* leaves as an eco-friendly adsorbent. The result showed that the high surface area of the adsorbent can be responsible for its use in the removal of dyes from aqueous solution. The equilibrium adsorption was practically achieved in 120 minutes. The highest removal of dyes was obtained at pH 6. The removal efficiency increased with increase in agitation time and initial dye concentration. The equilibrium data were best described by Langmuir isotherm model with maximum monolayer capacity of 33 and  $37 \text{ mgg}^{-1}$  for crystal violet and rhodamine B respectively. The adsorption kinetics can be successfully fitted to pseudo-second order kinetic model. The result of the intraparticle diffusion model suggested that intraparticle diffusion might not be the only rate controlling step. Desorption study revealed that the recovery of the dye from adsorption was possible.

### 3.6 Removal of Dyes By Synthetic Polymer:

Chiou *et al.* [23] investigated the adsorption behaviour of two anionic dyes AAVN and RB4 in acid solutions on chemically cross-linked chitosan beads. It was found that the adsorption capacities had very large values of  $3362 \text{ mgg}^{-1}$  for AAVN and  $3823 \text{ mgg}^{-1}$  for RB4 at pH 3 and  $30^{\circ}\text{C}$ . The equilibrium data fitted the Langmuir model very well. Also, the kinetic data agreed very well with the pseudo-second order kinetic model. The removal of reactive dye (Blue D-5RN) from binary mixtures (Yellow D-5GN, Red D-8B, Black DN) using chitin by Filipkowska *et al* [31]. It was observed that the maximum adsorption capacity of the dye in the samples without pH adjustment accounted for 97 mg/g and in the sample with pH adjustment (pH = 3) it was 205 mg/g.

Sanghi *et al.* used a polymer derived from *Ipomoea dasysperma* seed gum and guar gum to decolourized dye solutions (Direct Orange 8, Acid Sandolan Red and Reactive Procion Brilliant Blue). It was observed that at optimum pH 9.5, the percentage removal was 95.61 and 79% for DO 8, ASR and RPBB respectively. Juang *et al.* [45] studied the adsorption of dye (Amido Naphthol Red G, AR1) onto hexadecyltrimethyl ammonium chloride modified montmorillonite and basic violet 10 onto MCM-41. It was observed that the adsorption capacity of AR1 onto organoclay increases slightly at low (2-3) and high (10-11) pH values. The isotherm data fitted Langmuir model very well and the low value of  $\Delta H$  of AR1 / organoclay indicated characteristics of physical adsorption while the slightly high  $\Delta H$  of BV10/MCM 41 confirmed the strong interaction between molecules of BV10 and surface hydroxyl groups of MCM-41. The aggregation of congo red on CPC (Cetylpyridiniumchloride) was studied by Liu *et al* [53]. The optimum pH was 2.03. The isotherm data followed Langmuir isotherm. At higher temperature, the adsorption capacity decreases. Congo red adsorption from aqueous solutions by using chitosan hydrogel beads impregnated with non anionic or anionic surfactant was studied by Chatterjee *et al* [21]. It was found that the adsorption capacity of chitosan hydrogel beads impregnated with Triton X-100 (TX-100) was enhanced at all concentrations of TX-100 while the increase in the concentration of SDS above 0.01% in the CS/SDS beads gradually reduced the adsorption capacity of the beads. Equilibrium adsorption isotherm data indicated a good fit to the Sips isotherm model and a heterogeneous adsorption process. The Sips maximum adsorption capacity of CS/TX-100 was 378.79 mg/g and 318.47 mg/g for CS/SDS beads, higher than the 223.25 mg/g of CS beads. Also, Chatterjee *et al.* [22] investigated the adsorption of congo red by chitosan hydrogel beads impregnated with carbon

nanotubes. It was observed that the equilibrium adsorption isotherm data of the adsorbent exhibited better fit to the Langmuir isotherm model than to the Freundlich isotherm model with maximum adsorption capacity of 450.4 mg/g.

### 3.7 Removal of Dyes By Activated Carbon Derived From Industrial/ Agricultural Waste:

Gimba *et al.* [33] investigated the adsorption of dye (Indigo dye) by powdered and granulated activated carbon from coconut shell. The agrowaste coconut shell was carbonized and was activated with different activating agents such as polymethyl methacrylate (PMMA), potassium carbonate, sodium hydrogen carbonate and their mixtures. The activated carbon produced by  $K_2CO_3$  and  $NaHCO_3$  and their mixtures gave 100% removal of the dye. PMMA alone and mixture with  $K_2CO_3$  gave lower than 50% and 55% respectively. The mixture of the three reagents with powdered and granular forms gave 52.4% and 99.8% respectively. Carbon from cassava peel, an agricultural waste as an adsorbent in the removal of dyes from aqueous solution by Rajeshwarivaraj [75]. The carbon was used in raw form and phosphoric acid modified form. It was found that there was a quantitative removal of 100% in the case of activated carbon prepared by chemical activation which showed that it was more efficient than the raw form. Rahman and Saad [77] utilized guava seeds as a source of activated carbon for the removal of methylene blue from aqueous solution. The carbon was activated by pyrolysis at temperature up to 700°C and by using  $ZnCl_2$  as activating agent, pyrolysis alone yields poor adsorption capability of the adsorbent but the optimum adsorption capacity was obtained when the sorbent was subjected to chemical activation, followed by pyrolysis at 700°C. The adsorption data showed best fit to the Freundlich model.

Decolourization of an acidic dye (acid orange) by charcoal from coffee ground was investigated by Nakamura *et al.* [64]. The result obtained revealed that the ratio of dye removal increased as the weight of sorbents increased and the removal ratio by sorbents carbonized at a higher temperature was also increased. Also, the intraparticle diffusion of the dye onto the adsorption was identified as the rate-limiting step in the adsorption process. Abechi *et al.* [2] investigated the comparison of adsorption of methylene blue by sawdust and walnuts shells carbon coated with ZnO. It was found that the percentage of dye removed decreased as the initial concentration of dye adsorbed increased but the amount of dye adsorbed increased with increase in the concentration of the dye. Also, there was an increase in the percentage of dye adsorbed as the dose of the carbon increased. The equilibrium data fitted better into the Langmuir model than the Freundlich model for both adsorbent. The adsorbent capacities of both adsorbents showed that sawdust carbon (10,000) was more efficient than walnut shell carbon (2,500).

The comparative study on the adsorption kinetics and thermodynamics of dyes (congo red, malachite green, rhodamine B) onto activated carbon from leaves of Pandanus was studied by Hema and Arivoli [39]. The data obtained correlated reasonably well with both Langmuir and Freundlich isotherm model. It also revealed that the amount of dyes adsorbed increased with increase in pH of the medium and the amount of the adsorbed dye slightly decrease with increasing ionic strength and increased with increase in temperature. The thermodynamic parameters gave values that showed spontaneity, feasibility and physical nature of the adsorption process. Sumanjit *et al.* [90] investigated the removal of health hazard causing acidic dyes (acid violet 17, acid violet 49, acid violet 54, acid blue 15 and acid red 119) from aqueous solution by adsorption. Carbon from bagasse, cow dung, groundnut shells, pea shells, used tea leaves, wheat straw and ash from brick kiln and cement kiln were used as adsorbent. The result obtained revealed that the optimum pH for the five dyes was 8.0. Also, the rate of dye removal increased with increasing contact time before equilibrium was established and the isotherm followed both Langmuir and Freundlich model for the adsorbents and dyes. Furthermore, the percentage of dyes sorbed increased as the sorbent dose was increased and on increasing the ionic strength of the solution there was decrease in adsorption percentage of dyes. Barley straw was utilized as a source of activated carbon for removal of methylene blue from aqueous solution by Hussein *et al.* [41]. The result showed that as the amount of adsorbent increased the percentage of dye removal increased and higher adsorption percentage are observed at lower concentrations of dye. Also, the optimum pH value for the adsorption was 7 and the equilibrium time was 90 minutes.

Kavitha and Namasivayam [48] studied the experiment and kinetic studies of methylene blue adsorption by coir pith carbon. It was observed that greater percentage of dye was removed with decrease in initial concentration of dye and increase in amount of adsorbent used. The kinetic data followed the pseudo-second order model indicating that the process was a chemisorption. The isotherm data fitted Langmuir and Tempkin isotherm best. The maximum removal of 97% was obtained at natural pH of 6.9 for an adsorbent dose of 100mg of 50ml of dye and 100% removal was obtained for an adsorbent dose of 600mg in 50ml of 10mg/L dye concentration.  $\Delta G$  value obtained indicated the feasibility and spontaneity of adsorption process. Arivoli and Thenkuzali [11] studied the kinetics, mechanistic and thermodynamics of the adsorption of rhodamine B by acid activated low cost carbon (phoenix sylvestric leaves). The result showed that the adsorption kinetics followed the pseudo-first order reaction and the rate is mainly controlled by intraparticle diffusion. The isotherm data followed Langmuir and Freundlich. The temperature variation study showed that the adsorption process was endothermic and spontaneous with increased

randomness at the solid solution interface. Almost 90% removal of rhodamine B was observed at 60°C. Orthophosphoric acid activated Babul seed carbon was used as an adsorbent for the removal of methylene blue by Sujatha *et al* [89]. The result revealed that the percentage of dye adsorbed increased with increase in pH. The kinetic data followed the pseudo second order model. The isotherm data fitted the Langmuir model and increase in temperature increased the adsorption capacity. The enthalpy change value revealed that the process was endothermic.

Malarvizhi and Sulochana [57] studied the sorption isotherm and kinetics of methylene blue uptake onto activated carbon prepared from wood apple shell. It was found that increase in pH of the initial concentration of dye solution and carbon dose increased the percentage adsorption. The equilibrium data were best fitted to Langmuir model and Redlich-Peterson model. The kinetic data follows pseudo-second order rate equation. Activated carbon prepared from Jack fruit (*Artocarpus heterophyllus*) peel waste was used for methylene blue removal by Prahas *et al* [72]. It was observed that the removal efficiency increased with increased pH. The equilibrium data fitted the Langmuir model and the kinetic data correlated well with pseudo-second order model. Ho *et al.* [40] studied the adsorption of methylene blue onto activated carbon prepared from *Delonix regia* pods. It was observed that the percentage adsorption increased as adsorbent dosage increased. The isotherm data fitted the Langmuir model with monolayer sorption capacity of 24.0 mg/g. Gimba *et al.* [34] used activated carbon prepared from *Khaya senegalensis* fruit to adsorb methylene blue from aqueous solution. It was discovered that out of all the activating agents (NaCl, KCl, CaCl<sub>2</sub>, MgCl<sub>2</sub>·6H<sub>2</sub>O, Na<sub>2</sub>CO<sub>3</sub>, K<sub>2</sub>CO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub> and ZnCl<sub>2</sub>), the NaCl activated carbon has the best adsorption characteristic. Somboon *et al.* studied the removal of direct dyestuff (Direct Blue 201, Direct Red 23 and Direct violet 9) by wood charcoal and activated wood charcoal. It was observed that the adsorption of the dyes on both adsorbents was pH independent over a pH range of 5-11. Adsorption isotherms were found to conform to both Langmuir and Freundlich isotherms. The order of dyes adsorption capacity of WC and AWC are Direct Red 23 > Direct violet 9 > Direct Blue 20 (AWC has a higher adsorption capacity than WC). The adsorption capacity increased with increasing temperature and the adsorption process was an endothermic reaction.

The equilibrium, kinetics and thermodynamic studies on adsorption of Remazol black 5 on the palm kernel shell activated carbon was done by Zawani *et al* [110]. It was observed that the optimum pH was 2. The isotherm data fitted Freundlich model very well. The kinetic data fitted the pseudo-second order model better. The negative values of  $\Delta G^0$  and  $\Delta H^0$  obtained indicated that the adsorption process was spontaneous and endothermic. Nourouzi *et al.* [68] studied the adsorption of reactive dyes (Reactive Black 5 and Reactive Red E) onto palm kernel shell-based activated carbon. The result showed that the external co-efficient of mass transfer decreased with increasing initial adsorbate concentration. Also, it was found that the adsorption process was better described by using film-surface diffusion. Rice husk carbon was used to adsorb dyes (crystal violet, direct orange and magenta) by Verma and Mishra [97]. It was found that the optimum time was 45 minutes and there was decrease in adsorption capacity in the low pH region. Also, the removal of dyes increased with increase in temperature and there was increase in removal of dyes with increasing adsorbent dose. The removal percentage decreased with increase in initial concentration of dyes. Santhi *et al.* [84] investigated the adsorption of methylene blue, methyl red and malachite green onto activated carbon from *Annona squamosa* seed. The result revealed that the adsorption of dyes on the adsorbent showed highest values at around pH 7.0 and the kinetic data followed the pseudo second order kinetic model with intraparticle diffusion as one of the rate determining steps. The adsorption–equilibrium was represented with Langmuir, freundlich, Dubinin-Radushkevich and Tempkin isotherms. The adsorption of acid red 18 by activated carbon from poplar wood was investigated by Shokoohi *et al* [87]. It was observed that the removal efficiency was increased with increasing adsorbent dosage, contact time and decreasing pH but with increase in dye concentration, the removal efficiency was decreased. The adsorption isotherm models showed that Langmuir isotherm model was best fitted onto collected data. In addition, kinetic models showed that sorption of the dye onto activated carbon prepared from poplar wood follows the pseudo-first order model.

The adsorption of basic dyes (malachite green, crystal violet and rhodamine B) from synthetic textile effluent by activated carbon prepared from *Thevetia peruviana* was investigated by Baseri *et al* [15]. It was found that the amount of dye adsorbed increased with increasing initial dye concentration. The dye adsorption increased with increase in temperature indicating that the adsorption was endothermic in nature. Kinetic studies showed that the adsorption of these dyes followed pseudo-second order model with multi-step intraparticle diffusion model. The data obtained from adsorption isotherms are well fitted with Langmuir model, the negative  $\Delta G^0$  value obtained suggested the process was spontaneous in nature.

#### 4.0 Conclusion:

This paper has attempted to give insight into a wide range of adsorption of dyes. This ranges from the materials used for adsorption, the dyes that were adsorbed and the method used in the adsorption. Certainly, the effectiveness

of adsorption onto commercial activated carbon was high but the cost is also a barrier compared to using any other materials. In particular, the alternative adsorbents such as natural and synthetic polymers, activated carbon from natural materials/natural materials shows a good replacement for commercial activated carbon on adsorption of dyes.

However, there is a strong need to investigate extensively on the following points;

- i. To improve on the report of competitive adsorption of dyes on various adsorbents.
- ii. To increase the studies on comparison of various adsorbents with commercial activated carbon in adsorption of dyes to ascertain the cost reduction
- iii. To study the effect of any other materials in the aqueous solution on the adsorption of dyes

With the above points been put into consideration, there will be little or no injury due to discharge of dye wastewater and this will contribute to the overall strategy of waste minimization.

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