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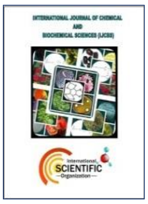

Wastewater Treatment using Soil Composites as Effective Adsorbents – A Comprehensive Review

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Abstract

Wastewater contamination by toxic heavy metal ions, dyes and other pollutants remains a serious public health problem for humans, so attention on specific methods and technologies to remove these pollutants from wastewaters are desired. There are number of water purification techniques but the adsorption is one of the most simplest, effective and economical method for wastewater purification. Among various used adsorbents, soil and its composite materials such as fibre soil composite and volcanic soil composite with magnetite are widely used for removing the toxic pollutants from aqueous solutions and wastewater due to its wide availability and cost efficiency. The present review was carried out to investigate the behavior of soil composites as adsorbent for pollutants removal. Soil based composites can remove the dyes and other wastewater pollutants more efficiently than simple soil. Alluvial soil composite, calcareous soil–alginate composites, kaolinite/smectite A composite, kaolinite/smectite B composite, green composite lateritic soil and gastropod shell are discussed in detail in this review. Various parameters including contact time, adsorbent dosage, pH, initial concentration, agitation speed and particle size were studied. Characterization of adsorbents was also studied by using SEM (Scanning Electron Microscopy), Fourier Transform Infrared Spectroscopy (FTIR), UV-Visible Spectroscopy and X-Ray Diffraction Analysis (XRD).

Keywords: Wastewater, Soil Composite, Kaolinite, Green Composite, Gastropod Shell, Alluvial Soil Composite, Calcareous Soil Composite, Smectite A Composite

Full length article **Corresponding Author*, e-mail: farwa668@gmail.com

1. Introduction

Environmental issues like global warming and water pollution have drawn the global attention for last few decades which is more closely related to industrial sector. Large volumes of highly polluted and toxic wastewater are produced by or mainly come from textile industry that is of great concern now-a-days [1]. For the survival of all living organisms, water is the prime necessity of life and extremely essential component of biosphere [2]. Water has great economic, social and environmental importance and is a basic need for survival of all life forms on earth [3]. Various activities of living organisms require the supply of good quality water. However, day by day water quality is deteriorating mainly because of anthropogenic activities, population growth, unplanned urbanization, rapid industrialization and unskilled utilization of natural water reservoirs [4]. Through different anthropogenic and natural activities, various organic, inorganic and biological impurities are added in water [5]. Different toxic heavy metals are the main constitutes of inorganic pollutants. For biological life, the presence of these heavy metals in water makes it harmful. Toxic dyes, herbicides, antibiotics and aromatic compounds are different organic moieties that are also present in wastewater [6].

Toxic materials have very hazardous effects on human beings as these substances may enters into human body either directly through drinking water or indirectly via eating food, plants and fish [7]. Synthetic dye, which is rendered due to its eco toxicological effect, is the example of one of dangerous pollutants [8]. These dye compounds are released from different industrial products like paint, paper, textile, leather, cosmetics, plastics and pharmaceuticals, which extensively use such kind of toxicants to color their products. Among these, the textile industry itself has been reported to be responsible for releasing approximately 146,000 tons of dye/year along with its wastewater. An inefficient dying procedure often causes a loss of 2% (basic dyes) to 50% (reactive azo dyes) synthetic dyes, which are released as effluent [9]. Azo-group, nitro-group or sulfo-groups present in these dye molecules makes them recalcitrant to biodegradation materials and their residues thereby accumulating in surrounding biota. Allergic reactions to skin and eyes, cancer, *methaeno globinaemia*, relentless inflammations in gastrointestinal tract and inhibition of mitotic cell divisions are the major toxic effects of these dyes [10]. The removal of various impurities from wastewater is essential for survival of humans and maintenance of healthy environment [11].

Adsorption through membrane filtration [12], oxidative processes and biological treatment [11] are the most common methods used for removing these impurities till now. A currently applied method includes ion exchange and electro dialyses that are known to have relatively high operation and maintenance costs. Chemical precipitation is however inexpensive but still unable to remove trace concentrations of heavy metal ions. Adsorption strategy is being expected to overcome such shortcomings and is being used for pollutants removal due to its properties of reasonable cost, high-efficiency and easy-pattern.

Soil and various soil composites such as fiber soil composites have been accepted as feasible materials for reducing the cost of wastewater treatment that is among various other widely used adsorbents of natural origin such as polymers [13]. Soil has low porosity, minute diffusivity and high cohesive and adhesive properties [14], so it may be considered as more acceptable adsorbent for the filtration of dye compounds present in wastewater [15]. Kaolin is the one of the common clay minerals which is being used as adsorbent for wastewater treatment [11]**.** Kaolinite and smectite are also common component of soils and sediments. These soil particles can combine with the pollutants present in wastewater and can be potential adsorbent having low cost, rich natural abundance, eco-friendly nature, high mechanical and chemical stability [7]. Volcanic soils which are naturally produced can be used for wastewater treatment due to high adsorbing ability based on their surface characteristic which contain Si/Al atom. Its effectiveness as adsorbent can be improved by mixing iron ore magnetite with volcanic soil [16]**.**

2. Methods for Pollutants Removal

Physical, biological and chemical methods are the well-known techniques for treatment of wastewater. Physical method comprises different precipitation methods such as coagulation, flocculation and sedimentation, adsorption (on activated carbon, biological sludge and silica gel), filtration and reverse osmosis etc. Biological treatments can further be differentiated in accordance with the presence or absence of oxygen. Since biological treatment resembles degradation processes that naturally occur in environment that is also known as biodegradation**.**

2.1 Chemical Methods

In chemical treatment methods, chemicals are extensively used for discoloration of wastewater. They include reduction, oxidation, complex metric methods, ion exchange and oxidation process. Chemical precipitation is cheap but it is still unable to remove trace of concentrations in heavy metal ions [17].

2.2 Oxidation

Oxidation is the most commonly used chemical discoloration process because it is easy to handle. Hydrogen peroxide is the commonly used oxidizing agent and this property is mainly attributed to its stability in pure form which further needs to be activated. A discoloration method differs in the way in which hydrogen peroxide is activated. Fenton’s reagent [hydrogen peroxide, activated with Fe(II) salts] is very appropriate for wastewater oxidation which prevents biological treatment or are poisonous. Fenton process is preferred for wastewater treatment in the cases when a municipality permits the release of Fenton sludge to the sewer [18].

2.3 Biological Methods

Biological elimination of heavy metals in wastewater comprises the use of biological techniques for the excretion of pollutants from wastewater. Microorganisms play a role of settling solids in the solution in this method [19]. Biological techniques involves biodegradation or breakdown by living organisms that is the most important removal process of organics which are transferred from industrial processes into solid and aquatic ecosystems. Heterotrophic micro-organisms are the most important living beings. Most presently used laboratory methods for screening biodegradation includes aerobic micro-organisms which employ molecular oxygen as hydrogen acceptor during respiration. The simplest method for the anaerobic biodegradability screening is the use of ^{14}C labeled test substances, but this is a time taking and costly procedure [17].

2.4 Physical Methods

Physical methods involve coagulation-flocculation, adsorption, electrocoagulation and adsorption. Coagulation-flocculation based physical methods are beneficial for the discoloration of wastewater containing disperse dyes. Also they have low discoloration efficiency for the wastewater including reactive and vat dyes. These techniques also restrict their use because of the low discoloration property and generation of large amount of resultant sludge [20].

3. Adsorption

Adsorption is a phenomenon that is most commonly employed for removal of organic and inorganic pollutants. When a solution comprising absorbable solute contacts with a solid, liquid–solid intermolecular force of attraction causes some of the solute molecules from the solution to be alleviated at the solid surface. The solute present (on the solid surface) in adsorption method is known as adsorbate, whereas the solid on which it is floating is called as an adsorbent [21]. Adsorption is one of the most successful methods used to remove dyes and other pollutants from wastewater [16]. Some of the benefits of adsorption are possible regeneration at low cost, chance of known process equipment, sludge-free operation and improvement of the sorbet. Different adsorbents have been used for adsorption in wastewater treatment systems and filtration units. For example naturally existing materials, such as natural polymers, soil, fiber soil composites, kaolin, kaolinite and smectite, volcanic soil and its composite have been used for reducing the cost of wastewater treatment [13].

3.1 Soil Composite as Adsorbent

Now-a-days, dye-rich effluents are predominantly treated using different low-cost adsorbents. Owing to its low porosity, minute diffusivity and high cohesive and adhesive properties, soil and its composites are good adsorbents. Soil particles are able to resist the path of charged molecules and hence can perform the functions of membrane as well. This study is established to check the adsorption behavior of soil and its composite materials as adsorbent with enhanced properties to reduce the dyes and groundwater contamination. In order to achieve this objective, many different techniques had been followed such as column study (to determine the effect of different composites on effectiveness of the adsorbent). Soil composites (admixtures) were prepared with gypsum, kaolinite, cement, betonies, fly ash and calcium oxide separately, to study the flow of dyes through the membrane [15].

3.1.1 Kaolinite and Smectite

Kaolinite and smectite are the most common constituents used to remove wastewater pollutants of soils and sediments. When different types of pollutants in industrial effluents, domestic sewage sludge and other solid wastes are dumped on the surface of earth, then soil particles including kaolinite and smectite minerals can interact with the pollutants. The kaolinite and smectite minerals in soil may play a major role in scavenging pollutants from the environment. Kaolinite and smectite have a low CEC (cation exchange capacity) of the order of 3–20 meq/100 g, so it is not expected to be an ion exchanger of high order. Therefore, kaolinite and smectite mineral can be potential adsorbents owing to its low cost and rich natural abundance. Small numbers of exchange sites are located on the kaolinite surface and it has no interlayer exchange site. Therefore, kaolinite and smectite minerals should exhibit significant potential for the use in adsorption process and separation of heavy metals. However, the small CEC and adsorption process may also play effective role in scavenging inorganic and organic pollutants from wastewater [22]. Different tools can be used for the management of wastewater. Removal of these metals from industrial wastewater has recently become more intense with increasing industrial activities [7].

3.1.2 Reinforcement of Soil

Reinforcement of soil with natural or synthetic fibers are used as a mechanical methods for improving the mechanical activities (e.g., strength and load bearing capacity) of soil. In various cases, the mechanical improvement is achieved by placing the fibers in critical locations in soil mass. This is referred to as oriented or systematic reinforcement technique. Reinforcement can also be done through mixing the fiber with soil. This method is known as "random reinforcement". Mixing cement with the soil has great results in chemical reaction between soil, cement and water. The compressive strength of soil-cement is increased by increasing the cement content and this leads to delicate behavior or sudden failure. On the other hand, by increasing the cement to soil ratio for cohesive soils, shrinkage micro-cracks may also develop in the soil as a result of loss of water content during hydration or drying of cement. The work on reinforced cemented clay, soil is very limited and is mostly focused in the investigation of the effect of fiber content at constant length on the behavior of cemented clay soil. In comparison with oriented or systematically reinforced soils, fiber reinforced soils with random distribution of fibers exhibit some advantages [23].

3.1.3 Fiber Soil Composite

In the waste dump landfills, covers and bottom liners are utilized to limit the evacuation of gases as well as liquids into the atmosphere [24-25]. For relatively better design of a fiber-soil composite to be used as barriers, the fine quality fiber content for efficacious reduction of cracking should be determined. The use of natural fibers as augmentation in fiber-soil composite present good workability [26-27]. Furthermore, the employment of synthetic fibers has been evaluated by a number of researchers. Example includes polypropylene fibers, polyester fiber and rubber fibers. The use of polyethylene terephthalate (PET) fibers is of specific interest because PET is broadly utilized for the manufacturing of bottles and packaging of number of products [28].

3.1.4 Properties of Fiber-Soil Composites

Fibers have reflected improvement in mechanical behavior of the fiber-soil composites, mainly regarding the apex strength, irrespective of the form of fiber used for reinforcement. Some fluctuations and the rupture mechanism have also been witnessed, ranging from brittle to ductile behavior. This behavior is mainly related to the content, type, length and diameter of fiber reinforcement [29]. Significantly, fiber-reinforced soil holds stronger toughness and ductility and smaller loss of post peak strength, as compared to soil alone [30]. However, it should be noted that natural fibers are biodegradable and may not be long lasting. Nylon fibers are not depreciated by the presence of salts in soils, biodegradation and ultraviolet degradations. The tensile strength of nylon fiber is greater than many of the other materials i.e. paper and rubber tires [23].

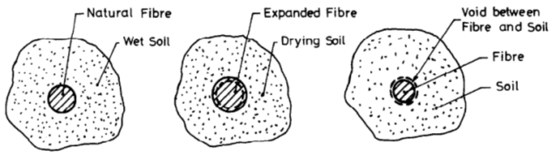

Fig 1 Interaction of natural reinforcing fiber and drying soil

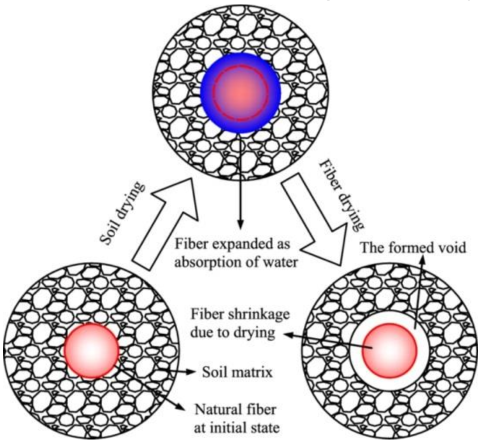

Fig 2 Soil-fiber reinforcement

3.1.5 Coconut-and-Sisal-Fiber-Reinforced Soil

Coconut-reinforced and sisal-reinforced soil has shown unexpected results. The presence of 4% of fibers, by weigh, reduced the amount of visible cracks to resist and gave high ductility soil blocks. Fibrous composite blocks are consisted of three-pronged local soils when mixed with sisal and coconut fibers. These variables control the strength and performance of developed composite, water absorption of fibers, tensile strength and bonding with soil. The influence of fiber/matrix ratio has also been observed in detail [31].

3.1.6 Green Composite Lateritic Soil and Gastropod Shell

Ground water (GW) contamination with fluoride F^- is highly pernicious to environment and there is need to minimize the F^- by using different absorbents in absorption based water treatment systems and filtration units. In such materials, the F^- binding sites are utilized by multivalent metallic species which is equal to high affinity of metallic species for F^-. Synthesis of functional reactive materials for potable water DE fluoridation using metal rich materials has been preferred. Some metal rich materials have been observed including calcium chloride altered zeolite, calcium aluminate-diatomaceous earth composite, gastropod shell, aluminum-diatomaceous earth composite, granular anionic clay composite and iron oxide/aluminum based nano-absorbents. Green composite reactive material was also prepared by combining gastropod shell (GS) with laterite soil (LS) in aqueous fluoridation. Tropical soil is abundant in iron oxide is known as "laterite". It is rusty red soil that is widely divided in tropics, subtropics and mediterranean climate zones. This soil is flexible and complemented with the rich oxide content, it often contain phyllosilicates minerals and iron (goethite and hematite). Gastropods have worldwide distribution and shell has the same chemical composition as other mollusks shells (contains a majorly $CaCO_3$ and nominal organic compounds) [32-33].

3.1.7 Magnetite/Volcanic Soil Composite

Adsorption process with natural materials like volcanic soils can be used for wastewater treatment. Volcanic soil has high adsorbing ability that is based on their surface characteristics having Si/Al atom. Volcanic soil can be used as adsorbent; it is difficult to separate supernatant and adsorbent within short time. The competency of volcanic soil as adsorbent was improved through compositing the volcanic soil with magnetite (Fe_3O_4). Magnetite is strongest magnetism between other iron oxides that generally used in numerous fields. Several methods have been used in magnetite synthesizing (Fe_3O_4) such as CVD (chemical vapor deposition), hydrothermal, solvo-thermal, electro-deposition, decomposition of high temperature organometallic and co-precipitation methods [34]. Co-precipitation method was used in this research because it is simple, requires low temperature and easy to handle [35]. Volcanic soil and magnetite samples showed similar adsorption capacity for dyes but lower than composite sample [16].

Table 1 Adsorption efficiency of soil composites

Soil Composites	Pollutants Removed	Removal Efficiency	pH	References
Alluvial Soil Composite	Crystal Violet (CV) Dye	99.98%	6.4	[15]
Kaolinite/Smectite A Composite	Pb (II) Ion	5.84 mg/g	3	[7]
Kaolinite/Smectite B Composite	Pb (II) Ion	6.51 mg/g	2.8	[7]
Green Composite Lateritic Soil and Gastropod Shell	Groundwater Contaminated with Fluoride (F^-)	43.7 mg/g	8.4-8.5	[36]
Cinnamon Soil Colloid Composite with Bacterial Cell	Cd (II)	160.4 mg/g	6.3	[37]
Cinnamon Soil Colloid Composite with Bacterial Cell	Cr (IV)	215.7 mg/g	5.5	[37]
Calcareous Soil–Alginate Composites	Fe (III) Ions	4.0 mg/l	-	[13]

Calcareous Soil–Alginate Composites	Mn (II) Ions	0.5 mg/l	-	[13]

4. Factors Affecting Adsorption by Composite

Different factors affect the adsorption process of soil such as pH, temperature and initial dye concentration. PDA-rGO-kaolin and graphene hybridized poly-dopamine-kaolin composite was used for the removal of MB dye. Adsorbent dosage was set as 0.8g/L and then decreasing trends of MB removal were observed. At higher initial MB concentration, PDA-kaolin and its composite have lower removal efficiency. Dye removal for 5–40mg/L of MB was 84.32%–50.77% by PDA-kaolin. Nevertheless, the resultant MB removal efficiency reached 97.28%–77.13% at same MB concentration using PDA-rGO-kaolin as adsorbent [38-39]. Removal of MB by PDA-kaolin and PDA-rGO-kaolin was examined at pH range of 3.5–11.0. Removal efficiency of MB through PDA-kaolin and PDA-rGO-kaolin was increased with increase in pH [11].

5. Analysis Techniques

Analysis of composites in adsorption was done by using scanning electron microscopy (SEM).), UV visible spectroscopy, fourier transforms infrared spectroscopy (FTIR and x-ray diffraction analysis (XRD). The surface morphology and chemical compositions of PDA-kaolin and soil composites were categorized by using x-ray photoelectron spectroscopy (XPS) and transmission electron microscope (TEM).

6. Conclusion

Water is one of the renewable resources, essential for sustaining all forms of life. A tremendous increase in the demand for freshwater is due to rapid growth of population and accelerated pace of industrialization. Human health is threatened by most of the agricultural activities and unsanitary conditions. Adsorption was best method for removal of pollutants. Soil composites as adsorbent were used for the dye removal. Soil composite such as alluvial soil removed approximately 99.98% of crystal violet (CV) dye at pH of about 6.4. Similarly, 4.0 mg/l of Fe(III) ions and 0.5 mg/l of Mn(II) ions was removed by calcareous soil–alginate composites. Kaolinite/smectite A composite was used to remove 5.84 mg/g of Pb(II) ion in water at pH 3 while kaolinite/smectite B composite was used to remove 6.51 mg/g of Pb(II) ion in water at pH 2.8. Green composite lateritic soil and gastropod shell was used to remove 43.7 mg/g groundwater contaminated with fluoride (F^-) at pH 8.4-8.5. Cinnamon soil colloid composite with bacterial cell was used to remove Cd(II) 160.4 mg/g at pH 6.3. Cinnamon soil colloid composite with bacterial cell was used to remove 215.7 mg/g Cr(IV) at pH 5.5. Kaolinite/smectite B composite have greater efficiency 6.51 mg/g to remove the Pb(II) ion at pH 2.8 as compared to kaolinite/smectite A composite that removed 5.84 mg/g Pb(II) ion at pH 3. Cinnamon soil colloid composite with bacterial cell have greater efficiency 215.7 mg/g to remove Cr(IV) at pH 5.5. So, in short we can say that soil composite possesses enough potential to act as potential natural adsorbent, when used properly. They are highly cost efficient there-by providing sustainable solution to polluted wastewater.

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