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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 concern for humans. The research on specific methods and technologies to remove pollutants from wastewaters is in high demand currently. There are number of water purification techniques, but the adsorption is one of the simplest, effective, and economical methods for wastewater purification. Among various used adsorbents, soil, and its composite materials such as fiber soil composites and volcanic soil composites with magnetite are widely used for removing the toxic pollutants from aqueous solutions and wastewater due to their wide availability and cost efficiency. The present review was carried out to investigate the behavior of soil composites as adsorbents 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 article. The effect of various parameters including contact time, adsorbent dosage, pH, initial concentration, agitation speed and particle size is critically evaluated and described in this review. The characterization of adsorbents using SEM (Scanning Electron Microscopy), Fourier Transform Infrared Spectroscopy (FTIR), UV-Visible Spectroscopy and X-Ray Diffraction Analysis (XRD) is also discussed.

Keywords: Wastewater, soil composite, kaolinite, green composite, gastropod shell, alluvial soil composite, calcareous soil composite, smectite-A composite

 Full length article
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1. Introduction

Environmental issues like global warming and water pollution have drawn the global attention since last few decades [1-5]. Large volumes of highly polluted and toxic wastewater are produced by or mainly come from many industries [6]. For the survival of all living organisms, water is the prime necessity of life and extremely essential component of biosphere [7-8]. Water has great economic, social and environmental importance and is a basic need for survival of all life forms on earth [9]. 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 [8-10]. Through different anthropogenic and natural activities, various organic, inorganic and biological impurities are added in water [11-14]. 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 [15-16].

Toxic materials have very hazardous effects on human beings as these substances may enter the human body either directly through drinking water or indirectly via eating food, plants and fish [17]. Synthetic dyes are well known for their ecotoxicological effects and are among most dangerous pollutants [18-19]. These dye compounds are released from different industrial products like paint, paper, textile, leather, cosmetics, plastics, and pharmaceuticals [20]. 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 in effluents [21-22]. Azo-group, nitro-group or sulfo-groups present in these dye molecules makes them recalcitrant to biodegradation and their residues thereby accumulate in surrounding biota [22]. Allergic reactions to skin and eyes, cancer, inflammations in gastrointestinal tract and inhibition of mitotic cell divisions are the major toxic effects of these dyes [23-24]. The removal of various impurities from wastewater is essential for survival of humans and maintenance of healthy environment [25-26].

Adsorption through membrane filtration [27], oxidative processes and biological treatment [25] are the most common methods used for removing these impurities till now. The currently applied methods include ion exchange and electrodialysis. These methods are known to have relatively high operational 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 [28].

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 biopolymers [29]. Soil has low porosity, minute diffusivity and high cohesive and adhesive properties [30], so it may be considered as more acceptable adsorbent for the filtration of dye compounds present in wastewater [1]. Kaolin is one of the common clay minerals which is being used as adsorbent for wastewater treatment [25]. 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, ecofriendly nature, high mechanical and chemical stability [17]. 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 [31].

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.

2.1. Chemical methods

In chemical treatment methods, chemicals are extensively used for discoloration of wastewater. They include reduction, oxidation, complexometric methods, ion exchange and oxidation processes. Chemical precipitation is cheap but it is still unable to remove trace concentrations of heavy metal ions [32]. *Maqsood et al.*, 2019

2.2. Oxidation

Oxidation is the most used chemical discoloration process as it is easy to handle. Hydrogen peroxide is the commonly used oxidizing agent. Fenton's reagent [hydrogen peroxide, activated with Fe(II) salts] is very appropriate for wastewater oxidation. Fenton process is preferred for wastewater treatment in the cases when a municipality permits the release of Fenton sludge to the sewer [33].

2.3. Biological methods

Biological methods are very popular for the removal of pollutants from wastewater [34]. Biological techniques involve biodegradation or breakdown by living organisms that is the most important removal process of organics which are transferred from industrial processes into soil and aquatic ecosystems. 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 ¹⁴C labeled test substances, but this is a time taking and costly procedure [32].

2.4. Physical methods

Physical methods involve coagulation-flocculation, adsorption, electrocoagulation, and adsorption. Coagulationflocculation based physical methods are beneficial for the discoloration of wastewater containing disperse dyes. These methods have low discoloration efficiency for the wastewater containing reactive and vat dyes [35].

3. Adsorption

Adsorption is a phenomenon that is most 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 gathered 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 absorbed, called as an adsorbent [36]. Adsorption is one of the most successful methods used to remove dyes and other pollutants from wastewater [31]. Adsorption is a low cost process. 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 [29].

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 composites (admixtures) were prepared with gypsum, kaolinite, cement, betonies, fly ash and calcium oxide separately, to study the flow of dyes through the membrane [1].

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 due 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 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 [37]. 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 [17].

3.1.2. Reinforcement of soil

Reinforcement of soil with natural or synthetic fibers are used as 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 of micro-cracks may also develop in the soil because 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 Maqsood et al., 2019

systematically reinforced soils, fiber reinforced soils with random distribution of fibers exhibited more advantages in water treatment [38].

3.1.3. Fiber soil composite

Cover and bottom liners are utilized in the waste dump landfills to limit the evacuation of gases as well as liquids into the atmosphere [39-40]. The fine quality fiber content for efficacious reduction of cracking should be determined for relatively better design of a fiber-soil composite to be used as barriers. The use of natural fibers as augmentation in fiber-soil composite present good workability [41-42]. The utilization of synthetic fibers such as polypropylene fibers, polyester fibers and rubber fibers has been evaluated by several researchers. The use of polyethylene terephthalate (PET) fibers is of specific interest as PET is broadly utilized for the manufacturing of bottles and packaging of number of products [43].

3.1.4. Properties of fiber-soil composites

Fibers have reflected improvement in mechanical behavior of the fiber-soil composites, mainly regarding to the apex strength, irrespective of the form of fiber used for reinforcement. Some fluctuations and 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 [44]. Significantly, fiber-reinforced soil holds stronger toughness and ductility and smaller loss of post peak strength, as compared to soil alone [45]. 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 [38].

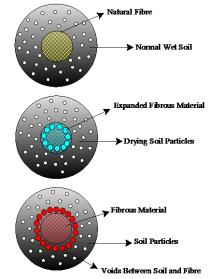


Figure 1: Interaction of natural reinforcing fiber and drying soil

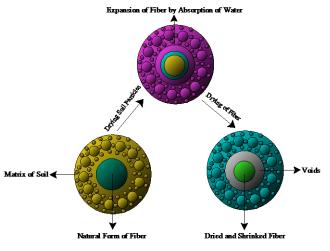


Figure 2: Soil-fiber reinforcement

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 [46].

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 defluoridation 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 prepared by combining gastropod shell (GS) with laterite soil (LS) in aqueous fluoridation. Tropical soil 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 contents. It often contains phyllosilicates minerals and iron (goethite and hematite). Gastropods have worldwide distribution and shell has the same chemical composition as other mollusks shells (contains majorly CaCO₃ and nominal organic compounds) [47-48].

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. The competency of volcanic soil as adsorbent was improved through compositing the volcanic soil with magnetite (Fe₃O₄). Magnetite shows strongest magnetism in iron oxides and generally used in numerous fields. Several methods have been used in magnetite synthesizing (Fe₃O₄) including chemical vapor deposition (CVD), hydrothermal deposition, solvo-thermal deposition, electro-deposition, decomposition of high temperature organometallic and coprecipitation methods [49]. Co-precipitation method was preferred in some researches as it is simple, requires low temperature and easy to handle [50]. Volcanic soil and magnetite samples showed similar adsorption capacity for dyes but lower than composite sample [31].

Soil Composites	Pollutants Removed	Removal Efficiency	pH	References
Alluvial Soil Composite	Crystal Violet (CV) Dye	99.98%	6.4	[1]
Kaolinite/Smectite A Composite	Pb (II) Ion	5.84 mg/g	3	[17]
Kaolinite/Smectite B Composite	Pb (II) Ion	6.51 mg/g	2.8	[17]
Green Composite Lateritic Soil and	Groundwater Contaminated	43.7 mg/g	8.4-8.5	[51]
Gastropod Shell	with Fluoride (F ⁻)			
Cinnamon Soil Colloid Composite with	Cd (II)	160.4 mg/g	6.3	[52]
Bacterial Cell				
Cinnamon Soil Colloid Composite with	Cr (IV)	215.7 mg/g	5.5	[52]
Bacterial Cell				
Calcareous Soil–Alginate Composites	Fe (III) Ions	4.0 mg/l	-	[29]
Calcareous Soil–Alginate Composites	Mn (II) Ions	0.5 mg/l	-	[29]

Table 1: Adsorption efficiency of soil composites

4. Factors affecting adsorption by composite

Different factors affect the adsorption process of soil such as pH, temperature, and initial dye concentration. Polydopamine (PDA) coated materials are becoming exceedingly popular in dye removal. A novel composite of PDA-kaolin with reduced graphene oxide hybridization known as graphene hybridized poly-dopamine-kaolin composite (PDA-rGO-kaolin) was used for the removal of methylene blue (MB) dye. Adsorbent dosage was set as 0.8g/L and MB removal was observed at various dye concentrations. At higher initial MB concentration, PDAkaolin and its composite have lower removal efficiency. Dye removal for 5-40 mg/L of MB was 84.32%-50.77% by PDA-kaolin. The resultant MB removal efficiency reached 97.28%-77.13% at same MB concentration using PDArGO-kaolin as adsorbent [53-54]. Removal of MB by PDAkaolin 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 [25].

5. Analysis techniques

Composites are analyzed 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) [25].

6. Conclusions

Water is one of the renewable resources and is 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 is one of the best methods for the 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, Fe(III) ions and Mn(II) ions were effectively removed by calcareous soil-alginate composites. Kaolinite/smectite-A and kaolinite/smectite-B composites were used to remove Pb(II) ions from water at pH 2.8. Green composite lateritic soil and gastropod shell was used to remove fluoride (F⁻) from groundwater. Cinnamon soil colloid composite with bacterial cell was used to remove Cd(II), Cr(IV) from water. In conclusion, soil composite possesses enough potential to act as potential natural adsorbent, when used properly. They are cost efficient thereby providing sustainable solution to polluted wastewater.

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