HYDROGEN
JURNAL KEPENDIDIKAN KIMIA
https://e-journal.undikma.ac.id/index.php/hydrogen/index

December 2024. 12(6)
e-ISSN: 2656-3061
p-ISSN: 2338-6487
pp. 1230-1243

Absorption of Metal Ions and Dyes in Liquid Waste : A Literature Review

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Article History
Received: 10-08-2024
Revised: 17-11-2024
Published: 31-12-2024

Keywords: absorption, metal ions, dyes, liquid waste

Industrial development and population growth cause a lot of waste to enter water bodies, causing pollution. A number of studies have been carried out to remove metal ions and dyes in waters. Adsorption is the most widely used method to absorb heavy metal ions and harmful dyes by utilizing natural resources that are still abundant, for example, agricultural, livestock, and fishery waste. Adsorption can also use activated or modified absorbent materials to increase porosity and expand the surface plane. In this review, the adsorption method and results obtained by the researcher with various absorbent materials are summarized which are mostly carried out in the last ten years. The purpose of this review is to provide input in finding alternatives to other absorbent materials or eliminating pollutants harmful to human health and the environment.

How to Cite: Indah, D., Afriani, A., & Nurwahidah, N. (2024). Absorption of Metal Ions and Dyes in Liquid Waste : A Literature Review. Hydrogen: Jurnal Kependidikan Kimia, 12(6), 1230-1243. doi:https://doi.org/10.33394/hjkk.v12i6.13823

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INTRODUCTION

Dyes are colored organic compounds that have chromophore functional groups such as (NR2, NHR, NH2, COOH and OH) and auxochromes such as (N2, NO and NO2) (Kausar et al., 2018). Most textile industries use dyes and pigments to dye their products. Chromophores are responsible for producing color, and auxophores complement chromophores that make molecules soluble in water and provide affinity with fabric fibers. Synthetic dyes come from a variety of industries such as leather, textiles, paper printing, food, pharmaceuticals, and cosmetics. Synthetic dyes can endanger industrial workers who do not prioritize safety and also wasted waste can enter water bodies and even be drunk by humans.

Hazardous dyes that are waste from the industry are complex molecules, stable and difficult to decompose. The most common danger of these dyes is respiratory problems due to inhalation of dye particles that interfere with breathing, watery eyes, sneezing and asthma symptoms such as coughing and wheezing (Hassaan and Nemr, 2017). In addition, azo compounds that are widely found in dyes have a complex and stable structure, causing greater difficulties in the degradation of textile waste. The toxicity of wastewater produced by the textile industry and the paint industry and others is a major challenge for ecological observers (Holkar et al., 2016).

The presence of dyes in the water, even in small amounts, will be very harmful to human health such as kidney dysfunction, disrupting the reproductive system, liver, brain and central nervous system as well as triggering cancer (Sharma, Dalai and Vyas, 2017). Acute and chronic effects on the organism, depending on the time of exposure and the concentration of the Azo dye. 1,4-diamino benzene is an aromatic amine which is the parent of azo dyes that can cause skin irritation, dermatitis, permanent blindness, vomiting gastritis, hypertension, vertigo, edema of the face, neck, pharynx, tongue and larynx as well as respiratory disorders (Hassan, Nemr and Hassaan, 2017). In addition, the waste generated during the process and operation, contains inorganic and organic contaminants that are harmful to the ecosystem and biodiversity that have an impact on the environment (Lavanya et al., 2014). Dyes can interfere with the growth of aquatic plants because they reduce sunlight transmission and increase toxicity in the water which is harmful to aquatic life.

Heavy metals can enter the environment through oil mining, coal, power plants, pesticides, ceramics, metal smelters, fertilizer factories and other industrial activities. Industrial waste has great potential as a cause of water pollution because it contains B3 waste, which is a hazardous and toxic material. Heavy metal pollution can enter through air, soil and water. From the air, metal pollution is produced from the discharge of motor vehicle smoke and industrial chimney smoke, and from the soil in the form of metal mining and industrial waste flushing (Widaningrum, Miskiyah and Suismono, 2017).

The presence of heavy metals in waters is toxic, even in low concentrations. Without realizing it, these heavy metal ions can accumulate in the human body, because they consume water and food such as rice, vegetables, fish and shellfish that are contaminated with heavy metals, so that they can cause metabolic disorders, neurological disorders and decreased intelligence, as well as the cause of cancer and even death (Yahaya and Don, 2014).

The degradation of dyes depends on several parameters such as pH, catalyst concentration, substrate concentration and the presence of acceptors. In photocatalytic reactions using ZnO/sunlight is better than ZnO/UV reactions. Another advantage besides taking advantage of sunlight, is the absence of sludge production and a considerable reduction in COD. However, the main drawbacks of this process are the presence of penetration, fouling of the catalyst, and separation problems from the treated liquid.

Photocatalysis with TiO_2-UV (Azkia Alma Ayesha, Akmal Mukhtar, 2015) can be used to reduce the content of dye waste in water. The degradation of yellow methanol solution was carried out photocatalytically by adding TiO_2 as catalyst and HNO_3 as oxidizer. The results of this study showed that 10 ppm yellow methanol was degraded by 30.755% after 120 minutes of radiation, while the addition of 0.008 g of TiO_2 at the same concentration could degrade 54.689% with an irradiation duration of 45 minutes. Sonolysis i.e., the use of ultrasonic waves for color removal and degradation of dyestuffs. The process depends on the power of the ultrasound and the total volume of the solution, and a decrease in the reaction rate is observed after changing the gas phase in the reactor from air to argon.

Initially, the selection of dyes in industry and their applications was not so considered, but in relation to their impact on the environment and human health, many researchers began to pay attention to the waste of these dyes. The most commonly used methods for removing dyes are biological oxidation and chemical precipitation, as well as coagulation (Fosso-Kankeu et al., 2017) and flocculation (T, Srimurali and K, 2014) for metal ions. In the last two decades, there have been many studies on the processing of dyes both physically and chemically (Karimifard and Alavi Moghaddam, 2018). Among them are ozone oxidation (Lambert et al., 2013), (Chollom, 2014), (Zhu et al., 2015), electrocoagulation (Karthik et al., 2014), (Ong et al., 2011), biocatalytic technology (Zhu, Chen and Wei, 2018), electrochemical oxidation (Zhu, Chen and Wei, 2018), photocatalytic degradation (Li et al., 2015), (Forgacs, Cserháti and Oros, 2004) reverse osmosis membrane separation technology (Ariyanti and Widiasa, 2011) and adsorption (Mo et al., 2018).

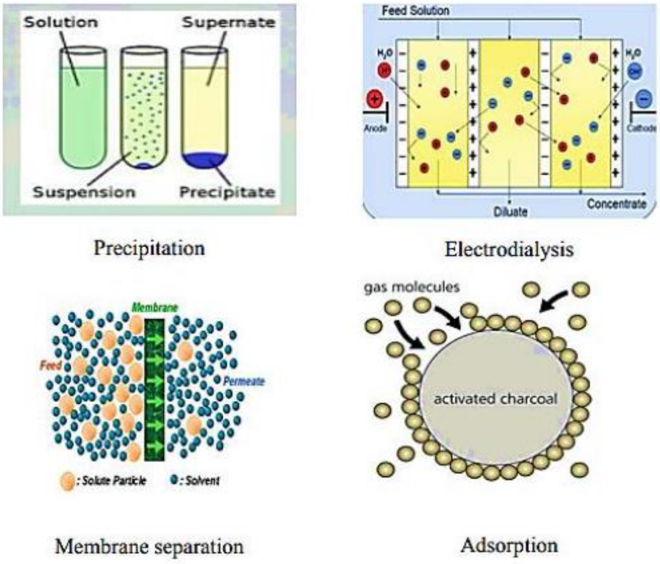

Figure 1. Several methods for removing metal ions (Raouf MS and Raheim ARM, 2017)

The absorption of metal ions and dyes using absorbent materials can be classified in two ways:

1. Based on their availability, namely: (a) Natural materials such as wood, peat, coal, lignite (b) Agriculture, Livestock Industry (Sa'adah, Hastuti and Prasetya, 2013), Fisheries (Annaduzzaman, 2015) or their by-products such as sludge, fly ash (Nguyen et al., 2017), domestic waste. (c) Synthesized products.
2. Based on its properties, namely: (a) Inorganic and (b) Organic.

METHOD

The research method used is literature study which is a method of problem identification, literature search, and data analysis by digging and collecting systematic data. Data collection was done by searching various references both national and international articles with online databases published in the last 10 years (2014-2024). This literature study uses 10 relevant articles that contain data on the methods used to absorption of metal ions and dyes in liquid waste. In addition, journals relevant to the topic were obtained by searching the database using the keywords “absorption”, “absorption metal ions”, “absorption dyes” and “absorption in liquid waste”.

RESULTS AND DISCUSSION

Adsorption using Natural Ingredients

Clay

Studies show that the adsorption ability of clay is due to the negative charge of silicate minerals neutralized by the adsorption of positively charged cations such as cationic dyes, heavy metals and others. Besides clay being used to make various types of ceramics, such as porcelain, bricks, tiles, and sanitary ware as well as essential components of plastics, paints, paper, rubber, and cosmetics, clay is non-polluting and can be used as a decontamination agent (Bergaya and Lagaly, 2013). Natural clay was used to remove Basic Yellow 2 (BY2) from the aqueous solution in a batch system and the maximum absorption capacity of the monolayer was found to be 833.33 mg/g at 25°C (at room temperature) (Öztürk and Malkoc, 2014).

Tunisian raw clay collected from the Tamra mine consisting of two species of clay minerals (kaolinite and halloysite) has shown efficiency to remove the azo dye Direct Orange 34 (DO34) from aqueous solutions. It was found that the rate of adsorption decreases with increasing temperature and the process is exothermal. Its adsorption kinetics follow a pseudo-second-order equation (Chaari, Moussi and Jamoussi, 2015) . Crystal violet removal from an aqueous solution was also carried out by Hamza et al using Tunisian Smectite raw clay. Batch studies were conducted to investigate the influence of experimental factors such as contact time (0–60 minutes), pH (2.5–11), adsorbent dosage (0.05–0.3 g/L), and initial dye concentration (12.5–100 mg/L) on Sono adsorption of Crystal violet dyes (Hamza et al., 2018)

The absorption of blue methylene cationic dye (MB) on the surface of kaolinite clay has also been carried out (Mukherjee et al., 2015). The results show that the MB adsorption isoterm without electrolyte follows the Langmuir model, while the presence of electrolyte follows the Freundlich model. At a constant concentration of dyestuffs, the absorption of dyes increases linearly as the ionic strength of the electrolyte solution increases. Among the four electrolytes NaCl, CaCl2, Na2SO4 and Na2HPO4 studied, it was found that Na2HPO4 had the highest increase in absorption. The study shows that the absorption capacity of kaolinite clay can be significantly improved with the use of electrolytes, which is very useful for the treatment of dye-contaminated wastewater.

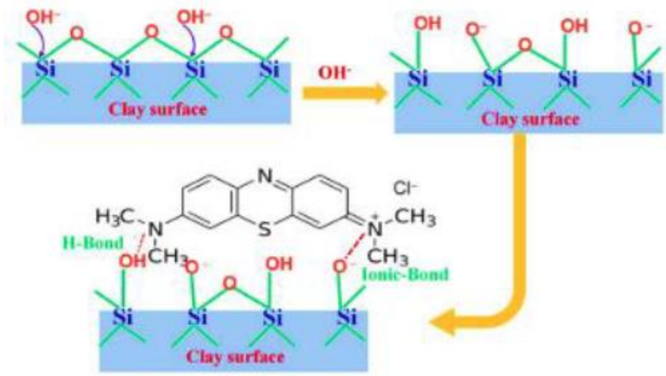

Figure 2. Interaction of clay molecules and blue methylene dye in alkaline pH.

Foorginezhad and Zerafat mix various proportions of clay, zeolite and polyethylene glycol for microfiltration of methylene blue, crystal violet and methyl orange from aqueous solutions. The results were 95.55% crystal violet removal, 90.23% blue methylene removal, and less than 10% orange methyl removal (Foorginezhad and Zerafat, 2017). Polyaniline-bentonite composites (Bent − APTES − PANI), as a new adsorbent successfully synthesized by oxidative polymerization method using ammonium persulfate as an oxidant. The positive charge on the adsorbent surface is generated using amino-activated bentonite (Bent − APTES) grafted with polyaniline treated with citric acid. Adsorption is applied to remove yellow methanol (MY) from the aqueous solution. The adsorption process is hardly affected by pH, as there are many amino groups (−NR3+) on the adsorbent surface. And the maximum adsorption capacity of Bent – APTES – PANI is 444.44 mg/g, which is better than other materials (Meng et al., 2017).

Zeolite

Zeolite is an aluminosilicate with a Si/Al ratio between 1 and infinity. There are 40 natural zeolites and more than 100 synthetic zeolites. They are also considered selective adsorbents. Zeolite is commonly used in the manufacture of detergents, ion exchange resins, catalytic applications in the petroleum industry (Pandiangan et al., 2017). For example, the manufacture of zeolite from rice husks and aluminum metal as a catalyst for the transesterification of palm oil, is also used in the separation process as an adsorbent for water (Shen, Zhao and Shao, 2014), carbon dioxide, and hydrogen sulfide.

Zeolite can be applied as a cation exchanger because zeolite contains various metal elements that can be interchangeable with other desired metals. Zeolite has a three-dimensional structure and has pores or chambers that can be filled by other cations or water molecules. The structural character of the hollow crystal lattice in zeolite functions to bind water molecules and metal ions. The metal ions and water molecules are free to move in the zeolite skeleton, making the zeolite can be used for ion exchange without experiencing changes in the crystal structure of zeolite (Atikah, 2017).

Wiyantoko et al. (2017) studied the effect of activation physics on natural zeolite minerals and natural clays through hydrothermal processes to determine their adsorption ability to blue methylene dyes. The results showed that natural zeolite and natural clay had adsorption capacities of 71.49 mg/g and 28.25 mg/g, respectively, while activated zeolite and activated clay were 75.77 mg/g and 69.78 mg/g, respectively.

Adsorption using Agricultural Waste

The skins and seeds of agricultural products after the fruit is used by the food industry, are often thrown away. In the last two decades, the use of agricultural waste has often been researched for its function as an absorbent of dyes and harmful metals in waters. In addition to its abundant availability, low cost and wide distribution, agricultural waste can also reduce solid waste that interferes with aesthetic value. The use of agricultural industrial waste as an absorbent is caused by the existence of functional groups contained in it that can bind to dyes or heavy metals.

Banana Peel

Banana peels have a high adsorption capacity for metals and organic compounds. Cellulose, hemicellulose, pectin, chlorophyll, and other low-molecular-weight species are its main constituents (Anastopoulos and Kyzas, 2014). Meanwhile, the hydroxyl and carboxyl groups of pectin function as metal ions or dye absorbers. Pb(II) and Cd(II) can be absorbed using kepok bananas with absorption capacities of 8.18 mg/g and 2.08 mg/g (Hafni, Zilfa and Suhaili, 2015).

Meanwhile, Ali and Said (2014) compared 4 ways of absorption of Cr (VI) and Mn (II) metal ions in banana peels, namely untreated banana peels (UTBP), banana peels hydrolyzed with alkalis (AlBP), banana peels hydrolyzed with acids (AcBP), and bleached banana peels (BBP). The maximum absorption capacity for Cr (VI) is UTBP (45%), AlBP (87%), AcBP, (67%) and BBP (40%). As for Mn (II), the maximum removal capacity of this adsorbent is UTBP (51%), AlBP (90%), AcBP (74%) and BBP (67%) at optimum conditions.

Corn Cob

Some agricultural waste has the potential to be adsorbents, namely corn cobs. Ningsih and Said (2016) used corn cobs to absorb Pb(II) by three methods, namely powder, charcoal and activated charcoal with HCl solution. The optimum conditions obtained for powder were 80 mg with 96.92% absorption, 80 mg charcoal with 97.29% absorption, and activated charcoal was 40 mg with 94.70% absorption.

Rice Husk

Rice husk ash contains more than 60% silica, 10–40% carbon and other small mineral compositions, which are byproducts during the gasification/pyrolysis process of rice husks. Many researchers are interested in how to use this industrial waste, because of its abundant amounts. Rice husk ash has been widely used as a construction material to make concrete, or as an adsorbent to absorb organic dyes, inorganic metal ions and waste in the form of gases (Shen, Zhao and Shao, 2014).

Ong and Hang (2013) performed Pb(II) absorption using rice husks, it was found that the absorption of 12.08 mg/g occurred in the pH range of 5 to 7 with a contact time of 30 minutes, Langmuir isotherms and pseudo-first-order kinetic models.

Silica can also be used to absorb metals and dyes. The silica made was extracted from rice husk ash. Agung et al.(2013) varied the KOH solution from 5%, 10% and 15% with variations in absorption temperature, the silica extracted from rice husk ash was the most obtained was 50.49% by using 10% KOH for 90 minutes. Separation of silica from straw (Manaa, 2015) with NaOH solution followed by acidification to obtain silica gel, then calcined and the resulting carbon is used to absorb Cr(VI) with absorption efficiency exceeding 98%.

Soursop Seeds

Adsorption of soursop seed tartrazine has been carried out by Fauzia et al to find the optimal conditions for the absorption of tartarrazine. The effects of pH, contact time, stirring rate, concentration and adsorbent dosage have been studied and the absorption capacity is 23.6310 mg/g at pH 2, contact time 120 minutes, stirring rate 100 rpm, initial concentration 600 mg/L and adsorbent dose 0.1 g. The IR spectrum shows hydroxyl groups and carbonyl groups as active sites (Fauzia et al., 2015).

Bark

Adsorption is a potential method to remove dyes from industrial wastewater. Gupta, Agarwal and Singh (2015) used maja fruit bark powder as an absorbent of Torque Blue. Experiments were conducted on different adsorbent doses, pH, temperature, dye concentration, particle size, as well as variations in contact time with the batch system. The results of the experiment revealed that the maximum absorption capacity of maja fruit bark in Torsi blue dye was 98% with an adsorbent dose of 0.7 g in a 100 ml dye solution which had a concentration of 15 mg/L at 310°K and pH 7.5.

Livestock Waste

Eggshells and eggshell membranes are waste materials that are widely produced from poultry, homes, restaurants, bakery businesses and food industry units. The surface of the membrane contains positively charged sites produced by the base side chains of amino acids with a very high surface area and contains functional groups such as hydroxyl (-OH), thiol (–SH), carboxyl (-COOH), amino (-NH2), amide (-CONH2) and others. Mittal et al., (2016) using eggshells and eggshell membranes has been carried out to absorb dyestuffs such as methylene blue, methyl orange, phenol and metals Pb, Cu, Cd, Cr and Hg. Comparison of absorption of Reactive Yellow to eggshells, eggshell membranes and eggshell mixtures, the absorption capacity of 32, 34, and 185 mg/g, respectively, proves that the adsorption affinity of the mixture of eggshells and membranes The eggshell is higher than using eggshell alone or eggshell membrane alone.

The addition of PVA with sulfuric acid to a mixture of corn cob - chicken feather biomass as an absorber of a mixture of Pb (II) and Cu (II) ions has also been carried out. The function of PVA and sulfuric acid is to enlarge the adsorbent pores of corn cobs - chicken feathers and increase the active group of OH. This is shown from the BET analysis with an increase in surface area of 43.42%, an average pore of 79.55% and a total pore volume of 2.5 times greater than adsorbents unmodified with PVA and sulfuric acid (Fatmawati, Hastuti and Haris, 2015). Meanwhile, chicken feather waste can absorb chromium metal from artificial waste containing potassium dichromate and an absorption percentage of 83.7% is obtained (Suseno, Mahayana and Darmawan, 2016).

Fisheries Waste

Zein et al., (2018) performed the absorption of Cd (II) and Cr (VI) metal ions using pencil shells (Corbicula moltkiana) activated with HNO_3 0.01 M. The absorption capacity obtained was 6.073 mg/g for Cd (II) and 1.286 mg/g for Cr (VI) metal ions, both of which followed the Langmuir isotherm model.

Activation and Modification

To increase the absorption capacity of biosorbents, functional groups are often activated before they are contacted with the sample. Mikati et al. (2013) activated Chaetophora Elegans algae before absorbing blue methylene, the result was that with the addition of 1 M HCl, the absorption capacity was higher compared to the addition of 1 M citric acid. The absorption capacity increased from 143 mg/g to 320 mg/g after the addition of HCl, while after the addition of citric acid decreased from 143 mg/g to 20 mg/g. Meanwhile, modified carbon can also be used to absorb Methylene Blue and Methaneyl Yellow dyes (P'yanova et al., 2017).

Jane et al (2014) compared the ability of activated carbon made from macadamia nut shells, baobab shells, pea shells, rice husks, moringa shells, and marula stones to absorb Pb(II), Zn(II), Cu(II), Ni(II), Fe(II), Mn(II), Hg(II), Cr(III), As(III) and Cd(II) from aqueous solutions. Experiments with batch systems were conducted at pH 4, 5 and 6. The adsorption of metal ions generally increases when the pH is raised from 4 to 6.

The percentage adsorption value is above 60% for the adsorption of Hg(II) by all activated carbon at pH 6. The adsorption of Pb(II) by carbon from Baobab shells, pea shells, moringa moringa shells oleifera and Marula stones is at least 22% higher than that of commercial carbon used as a comparator. Carbon derived from pea shells and baobab shells exhibits better adsorption of metal ions compared to other carbons and is used to determine the effects of initial metal concentration, contact time and adsorbent amount on metal adsorption.

Activated Carbon

Activated carbon is available in two main forms: activated carbon powder (PAC) and granular activated carbon (GAC). Most pollutant removal from water uses GAC because the granular form is more adaptable for continuous use and there is no need to separate the carbon from the liquid. The use of PAC presents several problems because it has to separate the adsorbent from the liquid after use. However, PAC is also used for wastewater treatment due to its low cost and faster contact. Activated carbon from castor fruit peels can be used to absorb metals Ni, Pb, Cu. The absorption capacity of Violet Crystals by castor skin activated carbon is 48.0 mg/g at an initial pH of 6.8 for particle sizes of 125-250 μM (Devi, Saraswathi and Makeswari, 2016).

The absorption of Rhodamin B using activated carbon from durian bark, after being activated with 3 M NaOH, obtained the KF value, which is the adsorbent adsorbent absorption capacity of 21.542 mg/g with Freundlich isoterm (Tanasale, Sutapa and Topurtawy, 2014). Absorption of erythrosin B using activated carbon from otapang activated with phosphate acid (Okoye and Dominic Onukwuli, 2016).

Graphene

Dyestuff waste has a strong color, high pH and is sometimes resistant to heat and difficult to degrade. The use of adsorbents against flame retardant dyes has increased significantly due to its simplicity, high efficiency, and wide availability. Nanomaterials have the potential to be efficient adsorbents because they have a large surface area. Graphene, a two-dimensional carbon-based material with a nanostructure, in addition to having thermal, mechanical, and electrical properties, can also absorb yellow methanol with an absorption capacity of 71.62 mg/g (Guo et al., 2013).

Magnetic Adsorbent

Metal ion removal using magnetic adsorbents has recently attracted attention in wastewater treatment (Kalia et al., 2014). Meanwhile, magnetic nanocomposites can be synthesized from chitosan and graphene oxide to absorb Pb (II) ions with an adsorption capacity of 76.94 mg/g (Fan L et al., 2013). In a similar study, chitosan-coated magnetite nanoparticles were successfully used to remove Pb(II) ions with a removal efficiency of 53.6% (Gregorio et al., 2012). A magnetization value of 70.1 m3/kg was also obtained for Fe3O4 nanoparticles without chitosan (Badruddoza et al., 2013).

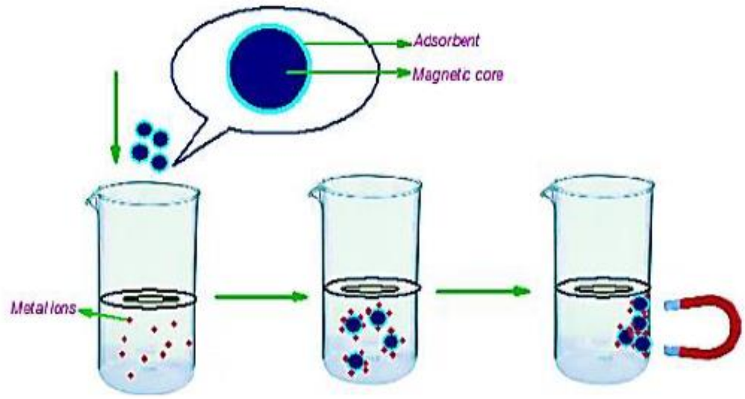

Figure 3. Representation of separation of adsorbed metal ions from aqueous media using magnetic nanocomposites (Kalia et al., 2014)

CONCLUSION

The absorption of heavy metal ions and harmful dyes using absorbent materials has been summarized both from natural materials and from agricultural, livestock and fishery waste as well as the synthesis of several other absorbent materials. From the data collected, in general, the research was carried out with a batch system with variations in pH, contact time, concentration of metals or dyes, temperature variations, adsorption isotherms and reaction kinetics. A number of studies that have been carried out can be a benchmark to find other absorbent materials to remove metal ions and harmful synthetic dyes in the waters by activating, modifying or mixing them with other materials.

ACKNOWLEDGEMENTS

Thank you to LPPM Undikma for funding research through internal research grants, laboratories at the Undikma Chemistry Laboratory and all parties who help researchers so that research can be completed properly and on time.

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