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Application of Green Synthesized Iron Nanoparticles for Treatment of Slaughterhouse Wastewater from Abattoir in Zaria, Nigeria

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Abstract: Meat is an important diet globally; hence, slaughterhouse is a renowned industry. Slaughterhouse wastewater contains high level of nutrients and trace amount of heavy metals that can pollute groundwater and food chain when discharged into the environment untreated. In this study, already green synthesized iron oxide nanoparticles (Fe_3O_4-NPs) were used as a Fenton-like catalyst for the reduction of physiochemical parameters including pH, colour, turbidity, total dissolve solids (TDS), biological oxygen demand (BOD), total nitrate, total phosphate and metal elements (Fe, Pb, Cr, K and Ni) contained in wastewater collected from a slaughterhouse in Zaria, Nigeria. The physiochemical parameters were measured before and after treatment with the Fe_3O_4-NPs to study its effectiveness in the treatment of wastewater. There was significant percentage reduction in the levels of pH (38.47), colour (42.86), Turbidity (52.95), TDS (30.83), BOD (60), total nitrate (52.05) and total phosphate (100). Furthermore, there was percentage reduction in the concentrations (mg/L) of metal elements in the wastewater, for Fe (67.42), Ni (23.81), Pb (63.64), K (48.47) and Cr (66.67). Green synthesized Fe_3O_4-NPs is therefore efficient in the treatment of slaughterhouse wastewater before discharging into the environment.

1. Introduction

Water is an essential substance on earth and every living organism needs water to survive, directly or indirectly. Water makes up 71% of the earth's crust however; freshwater consists of only 2.5% (Rajasulochana *et al*., 2018). A large volume of wastewater from industries or urban areas is used for irrigation purposes in agriculture. Rapid industrialization and urbanization have increased water pollution, hence, the risk of consuming polluted water especially in developing countries. There is an increasing problem of water scarcity, which harms human livelihood, economic development and the environment (Abdouni *et al*., 2021; Ogbomida *et al*., 2016; Bouknana *et al*., 2014;). Meat processing industry produces a large volume of wastewater that is discharged into the environment without proper treatment and slaughterhouses can contaminated the environment with surfactants from the cleaning process. Surfactants are major components of detergents, that usually enter the aquatic environment as a result of inadequate slaughterhouse wastewater (SWW) treatment, causing changes in the ecosystem that affect human, fish, and vegetation (Bustillo-Lecompte and Mehrvar, 2017).

Poorly treated wastewater may contain heavy metals, organic or inorganic compounds, and pathogens that may result in soil contamination. Slaughterhouse wastewater contains nutrients such as calcium, sodium, magnesium, sulfur, and iron and a trace amount of heavy metals such as cadmium, cobalt, nickel, copper, and chromium (Nandomah and Tetteh, 2023). These pollutants find their way into the groundwater and food chain causing serious threats to the natural environment and human. Oftentimes, untreated wastewater is used for irrigation; this can lead to increase in soil salinity, organic matter, exchangeable Na, K, Ca, Mg, plant available phosphorus and microelements, and decreased soil pH (Kiziloglu *et al*., 2008).

The effects of heavy metals in human, when exposed to them depend on the dosage as well as the period of exposure. Humans are exposed to these metals through inhalation, ingestion, or direct contact, the presence of these metals in the environment is linked to anthropogenic activities like paper, paints, electronics, and metallurgical industries, which are major sources of metal pollution (Jaishankar *et al*., 2014). A wide range of wastewater from different industries such as meat processing plants, textile plants, sewage treatment plants, and milk-processing factories are sources of heavy metal pollution. Exposure to these metals can have a deleterious effect on human health such as; damage to the brain, heart, nervous system, and kidneys (Mohammadpourfard *et al*., 2015). Metals such as cadmium (Cd), nickel (Ni), lead (Pb), and chromium (Cr) are toxic at very low concentration and can pose health risk (Sandeep *et al*., 2019; Ubong *et al*., 2023). Heavy metal in wastewater can get it way into agricultural fields, and this can have effect on plant growth (Okon *et al*., 2023a). Metals such as iron and potassium are essential micronutrients for the metabolism of living organisms (Lieu *et al*., 2001), however, they pollute the aquatic environment, which causes eutrophication and reduces the level of dissolved oxygen, which is harmful to aquatic life (Arienzo *et al*., 2009).

Adsorption is efficient in the removal of heavy metals and other pollutants from wastewater, compared to other conventional methods. The ability for iron nanoparticle to adsorb molecules makes it suitable for wastewater treatment. The principle involved in adsorption is adhesion in which, the liquid molecules are bound to the large surface area of the adsorbent. Diverse studies have reported physical and chemical methods targeted at remediating contaminated wastewater, such as coagulation, sorption, chemical precipitation, ion exchange, membrane filtration, and biological remediation (Marcos-Hernández *et al*., 2021). However, there are limitations in employing these methods for wastewater remediation such as the high cost, low adsorption capacity, and the generation of toxic sludge (Kumar *et al*., 2013; Fazlzadeh *et al*., 2016).

Nanotechnology is a field of research that has been widely recognized since last century when Nobel laureate Richard P. Feynman presented “nanotechnology” during his famous lecture in 1959 “There’s Plenty of Room at the Bottom” (Feynman, 1960). Since then, various revolutionary developments have been made in the field of nanotechnology. Nanoparticles (NPs) are materials that include particulate substances that have at least a dimension less than 100 nm (Laurent *et al*., 2008). Among all the nanoparticles present, metal nanoparticles have gained much importance. They are unique because of their large surface-to-volume ratio. They create a link between bulk materials and atomic or molecular structure. In recent times, iron oxide nanoparticles have been employed in a wide range of applications such as imaging devices, nanomagnetic sensors, photocatalysis, batteries amongst others (Tong *et al*., 2017). Iron nanoparticles have also find application in the removal of petroleum oil from contaminated water and soils (Erika *et al*., 2018).

They have a large surface area to volume ratio, small size, thermally and chemically stable, and strong magnetic properties and are therefore suitable for application in various fields (Armijo *et al*., 2020; Zare *et al*., 2018; Zhang *et al*., 2017). Green synthesis of metal nanoparticles is cheap compared to other methods as plants contain phytochemicals that act as reducing and capping agents for successful synthesis of metal nanoparticles, this has been observed by Adamu *et al*. (2021) and Ekwumemgbo *et al*. (2023) when silver nitrate and iron oxide nanoparticles were synthesized and characterised. Many researchers have established application of green synthesized nanoparticles for wastewater treatment; this is due to simple structural properties of nanomaterials that provide a better surface area with high adsorption capacity (Nakum and Bhattacharya, 2022).

This study aimed at treatment and remediation of slaughterhouse wastewater using already synthesized green iron oxide nanoparticles (Fe_3O_4-NPs).

2. Methodology

2.1 Wastewater Sample collection

The wastewater sample used for this study was from a slaughterhouse (Abattoir) within Zaria in Nigeria. Wastewater sample was collected using airtight 4 litres polyethylene plastic (high-density) bottle. The sample container was washed thoroughly with solution of detergent and rinsed with deionized water, then soaked in 10% “Analar” hydrogen chloride acid (HCl) overnight followed by rinsing with deionized water to remove trace elements contamination before being used for sample collection (Abah, *et al*., 2016; Okon *et al*., 2022). Sample container was rinsed with the wastewater before collection of samples. Wastewater sample was collected from a central location where wastewater from several locations within the slaughterhouse flows into, and stored at 4°C in a refrigerator to maintain the sample integrity for further use.

2.2 Treatment of wastewater

A 0.4 g of the already green synthesized iron oxide nanoparticles was added to a 50 cm^3 slaughter house wastewater (SWW) sample, and the pH was maintained at 5 by the addition of dilute sodium hydroxide. To this, 0.36 M of aqueous H_2O_2 (30 %) was added and the resulting suspension was magnetically stirred at room temperature. The treated sample obtained was then analyzed for physiochemical parameters to observed the levels of parameters in the wastewater before and after treatment. Figure 1 shows the already synthesized iron oxide nanoparticles.

Figure1. Already synthesized iron oxide nanoparticles

2.3 Determination of the physiochemical parameters

The physiochemical parameters determined were pH, colour, total dissolve solids (TDS), biological oxygen demand (BOD), total nitrate, total phosphate and metal elements (Fe, Pb, Cr, K and Ni). Each of the physiochemical parameters in the wastewater was determined before and after treatment with already green synthesized iron oxide nanoparticles (Fe_3O_4-NPs). Analyses were carried out in triplicate.

2.4 pH and colour

The pH was measured using digital pH meter (Mettler MP 220, Switzerland) by immersion of the probe into the sample solution and then reading was recorded similar to Okon *et al.* (2023b).The colour of the wastewater before and after treatment was determined using a spectrophotometer. The colour was determined by comparison of the absorbance of the sample to the absorbance of standard coloured solution.

2.5 Turbidity

The turbidity of the sample was measured with a turbidity meter. A turbidity meter works by passing a beam of light through a water sample and detecting how much of the intensity of the beam is reduced as revealed by a light sensor. The turbidity vial was filled to mark, and placed in the instrument cell compartment and the turbidity value was recorded from the instrument (Brikowski, 2003).

2.6 Total Dissolved Solids

Total dissolved solid (TDS) is a measure of the total organic and inorganic matter dissolved in the wastewater. It was determined in proportion to the electrical conductivity of the wastewater using an electrical conductivity meter (Jakhrani, 2019). The TDS (mg/L) was calculated as shown in **Eqn. 1**

$$\mathrm{TDS} = ke\,\mathrm{EC} \qquad \textbf{Eqn.1}$$

Where TDS is Total dissolved solid (mg/L), *ke* is the constant of proportionality and EC is the electrical conductivity (µS/cm) at 25 °C

2.7 Determination of Biological Oxygen Demand (BOD)

To determine the BOD of the wastewater, dissolved oxygen (DO) was first determined by Winkler's method as described in Standard Methods for Examination of Water and Wastewater (America Public Health Association (APHA), 1998). The sample was prepared by dilution with dilution water; two BOD bottles (B1 and B2) were filled with the diluted sample, DO was determined in freshly prepared B1 while that of B2 was determine after a 5-day incubation at 20°C (Jouanneau *et al*., 2014). The DO observed in B1 is D1, while the DO observed in B2 after incubation is D2. The BOD was calculated as presented in **Eqn. 2**

$$\mathrm{BOD\ mg/L} = (\mathrm{D1} - \mathrm{D2}) \qquad \textbf{Eqn. 2}$$

where:

D1 = Dissolved oxygen of the diluted sample immediately after preparation
D2 = Dissolved oxygen of the diluted sample after 5-day incubation at 20°C

2.8 Determination of Total Nitrate

Fadiran and Mamba (2004) method for determination of nitrate was used for this study. To determine the total nitrate, 100 cm^3 of the wastewater was transferred to a petri dish and evaporated to dryness. To this, 2 cm^3 of phenol disulphonic acid was added and stirred with a bent rod. Consequently, 6 cm^3 of ammonium hydroxide was added to the mixture, and effervescence was observed. Afterward, the petri dish was fill with distilled water. The solution was filtered with filter paper and the filtrate was analyzed using a U.V spectrophotometer (Model 752) at a wavelength of 410 nm. The absorbance of the blank and sample was measured and compared with a standard curve to obtain the total nitrate.

2. 9 Determination of Total Phosphate

The method for the determination of dissolved oxygen nanoparticles by Farah (2016) was used for this study. The water sample (1 cm^3) was diluted with 99 cm^3 of distilled water. Phenolphthalein (1 drop), 4 cm^3 of ammonium molybdate, and 10 drops of stannous chloride were added and a light blue solution was obtained. The solution was filtered with filter paper and the filtrate was analyzed using a U.V spectrophotometer (Model 752) at a wavelength of 690 nm. The absorbance of the blank and sample was measured and compared with the phosphate calibration curve to obtain the total nitrate.

2.10 Determination of elemental composition

Some metallic elements were determined, to achieve this, the wastewater sample was digested in microwave digestion system prior to analysis. The wastewater sample was digested according to standard method as described by American Public Health Association (APHA, 1999). The elements determined include iron (Fe), Lead (Pb), Cromium (Cr), Pottasium (K) and Nickel (Ni). The elements were analyzed through microwave-assisted spectroscopy using Microwave Plasma Atomic Emission Spectrophotometer (MPAES, Agilent 4200, USA). Barros *et al*. (2016) also used microwave-assisted spectroscopy for metal analysis. For quality control, calibration curves were prepared separately for each of the metals by running different concentrations of standard solutions, the instrument was set to zero by running the respective reagent blanks.

3. Results and Discussion

3.1 Physiochemical parameters of untreated and treated wastewater

Physiochemical characteristics of the untreated and treated wastewater from the slaughterhouse are represented in **Table 1**. The mean levels of the various parameters in the untreated wastewater align with the report by Bustillo-Lecompte and Mehrvar (2017). The pH value of the wastewater before treatment was 8.11±0.16 (**Table 1**), which is within the permissible limit of 6 – 9 (WHO, 2011). However, the pH value of the treated wastewater was 4.99±0.01, this is at variance with the permissible limit of 6 – 9 by WHO (2011), an indication that the treated wastewater is acidic and this can be attributed to the effect of H_2O_2 from the Fenton process. The reduction efficiency of synthesized Fe_3O_4-NPs on pH was 38.47% as shown in **Figure 2.** The color of the wastewater before treatment was 70±7.07 CU (Hazen), which is above the WHO standard permissible limit of 10-50. However, the color of the treated wastewater at 40±0.41 CU (Hazen) was within the permissible limit. A reduction efficiency of 42.86 % in improving the color of the wastewater by the iron nanoparticle was obtained as presented in **Figure 2**. Turbidity level (NTU) observed for the wastewater before and after treatment was 112.0±2.83 and 52.7±1.70 respectively (**Table 1**)**,** this shows a percentage reduction of 52.95 % in the level of turbidity after treatment (**Figure 2**).

Meanwhile, turbidity level of the wastewater before and after treatment was within the permissible limit of 150 by WHO (2006).

The treatment of the slaughterhouse wastewater with the already green synthesized Fe_3O_4-NPs resulted in a 30.83 % reduction of the total dissolved solids (TDS) as the TDS of the untreated wastewater was 1667±5.66 mg/L and 11534.24 mg/L ± before and after treatment respectively. After treatment, the wastewater could be used for irrigation since the TDS of water used for irrigation purposes should not exceed 1500 mg/L after treatment (Aniyikaiye *et al*., 2019). The level of TDS observed (1667±5.66 mg/L) was higher than the 260 mg/L obtained by Mohammed *et al*. (2020), when TDS was measured in abattoir wastewater from Kano. This could have resulted from the level of slaughtering activity in Zaria abattoir. The 1153 ± mg/L obtained is below the permissible level of 1500 mg/L by WHO (2006). The high level of TDS (1667±5.66 mg/L) before treatment can cause dehydration of aquatic organisms if it finds its way to water system (WHO, 2011). Biological oxygen demand (BOD) for the untreated wastewater was observed to be 100±0 mg/L while that of treated sample was 40±2.83 mg/L, this shows a 60% reduction in level of BOD by the nanoparticle used. The BOD value of the treated wastewater aligns with the specification for effluent discharge of 60 mg/L (WHO, 2006). Total phosphate and nitrate levels are usually high in wastewater from slaughterhouses (Ogbomida *et al*., 2016), this study reports 73±0 mg/L and 0.35±0 mg/L as the value for the total nitrate and total phosphate levels respectively for the untreated wastewater as presented in **Table 1**. In the treated wastewater, the concentrations were 35±0.71 mg/L and 0±0 mg/L for nitrate and phosphate respectively. As presented in **Figure 2**, there was a drastic reduction in the level of phosphate and a 100 % reduction was obtained. The concentration of nitrate and phosphate obtained in the treated wastewater aligns with limits for effluent discharge of 45 mg/L and 15 mg/L respectively (WHO, 2006).

Table 1. Physiochemical parameters of the untreated and treated wastewater

Parameters	**Level in Untreated Wastewater**	**Level in Treated Wastewater**
pH	8.11 ± 0.16	4.99±0.01
Color (Hazen colour unit)	70 ±7.0	40±0.41
Turbidity (NTU)	112 ± 2.83	52.7±1.70
Total dissolved solids (TDS)(mg/L)	1667 ± 5.66	1153±4.24
Biological oxygen demand (BOD) (mg/L)	100 ± 0	40±2.83
Total nitrate (mg/L)	73 ± 0	35±0.71
Total phosphate (mg/L)	0.35 ± 0	0±0

3.2 *Elemental composition of untreated and treated wastewater*

Concentrations of metallic elements in treated and untreated wastewater are presented in **Figure 3**, the percentage reduction in the levels of elements after treatment are presented in **Figure 4**. Fe_3O_4-NPs exhibited high efficiency in the reduction of Fe, Ni, Pb, K, and Cr contained in the wastewater. The highest concentration (mg/L) before and after treatment was observed for Fe (7.52 ± 0.00; 2.45± 0.49) while the lowest concentration was observed for Pb (0.11± 0.00; 0.04 ± 0.007) as presented in **Figure 3**. The mean concentration of elements contained in the wastewater follows the order Fe > K > Cr > Ni > Pb. This is in variance with the report of Mohammed *et al*. (2020), where the concentration (mg/L) of Ni (1.54) was higher than that of Fe (0.11) and Cr (0.10). The result showed that the treatment of the wastewater using the already synthesized Fe_3O_4-NPs reduced the concentration of Fe, Ni, Pb, K, and Cr by 67.42 %, 23.81 %, 63.63 %, 48.47%, and 66.67.81 % respectively.

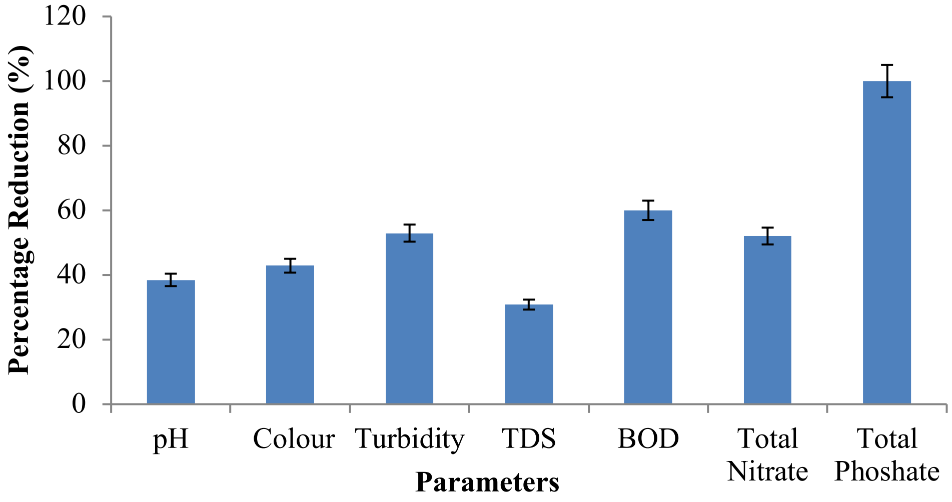

Figure 2. Percentage reduction of physiochemical parameters of wastewater after treatment

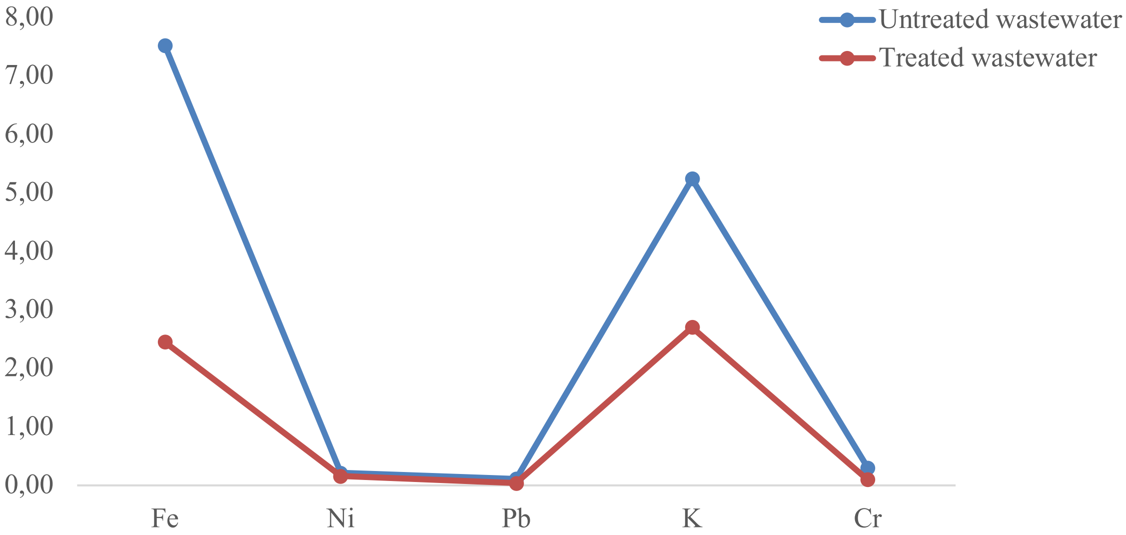

Figure 3. Concentration of elements in untreated and treated wastewater

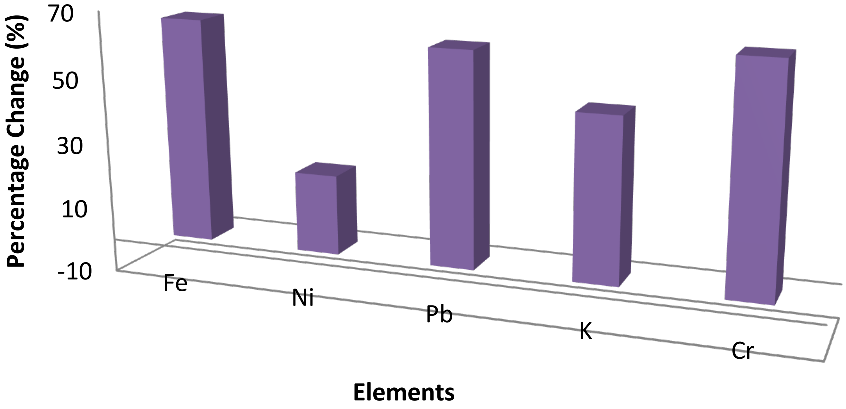

Figure 4. Percentage reduction in concentration of elements in wastewater

Although, there was a significant reduction in the mean concentration of Pb (0.04±0.007 mg/L) and Cr (0.1±0 mg/L) in the treated wastewater, the mean concentration of Pb and Cr after treatment exceeded the standard limits of 0.01 mg/L and 0.05 mg/L respectively (Aneyo *et al*., 2016; Ayeni, 2014). The mean concentration of Ni in the treated wastewater (0.16±0.028 mg/L) was also above the permissible level of 0.03 mg/L (WHO, 2006). Conversely, there was a significant reduction in the concentration of Fe in the treated wastewater (2.45±0.49 mg/L) which was below the WHO requirement of 5.0 mg/L (WHO, 2006). The mean concentration of K in the treated wastewater (2.7±0.28 mg/L) also complies with the WHO standard of 5.0 mg/L.

Conclusion

In this study, already green synthesized iron oxide nanoparticles (Fe_3O_4-NPs) were used to treat wastewater from abattoir to observe its efficiency in the reduction of physicochemical parameters of the wastewater. The parameters include pH, colour, turbidity, total dissolve solids (TDS), biological oxygen demand (BOD), total nitrate, total phosphate and metal elements (Fe, Pb, Cr, K and Ni). There was significant percentage reduction in the levels of the studied parameters when treated with the Fe_3O_4-NPs. This proves that Fe_3O_4-NPs is an effective Fenton-like catalysts for the degradation of the studied physiochemical parameters in wastewater.

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Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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