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Role of Soil pH for Enhancing DDT Remediation by Using Synthesised Iron Oxide Nanoparticles

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Abstract: This study investigates the influence of soil pH on the catalytic degradation of dichlorodiphenyltrichloroethane (DDT) using iron oxide (Fe_2O_3) nanoparticles, monitored by the characteristic absorption peak of DDT at 270 nm within the 220–300 nm spectral range. To determine the ideal conditions for nanoparticle-mediated reductive dechlorination, the degradation efficiency was assessed in acidic, neutral, and alkaline pH ranges. The results showed that DDT degradation achieved its maximum (58.7%) at pH 5, which was explained by increased corrosion of Fe_2O_3 nanoparticles and increased production of reactive Fe(II) species in acidic environments. Degradation efficiency reduced to 22.5% at neutral pH 7, most likely as a result of oxide/hydroxide layers stabilising the surface and decreasing electron transport. Due to the creation of passivating layers and reduced Fe(II) release, alkaline (pH- 9) condition inhibited reductive reactions and caused minimal degradation (0.44%). In order to optimise iron nanoparticle catalytic activity for successful DDT remediation in contaminated soils, our results emphasise the crucial role that pH plays in regulating nanoparticle surface chemistry and reactivity, highlighting the necessity of pH optimisation or buffering in field applications. Overall, the results show that the effectiveness of DDT remediation is highly dependent on the pH of the surrounding environment and indicate that maintaining the soil slightly acidic could enhance the catalytic efficacy of iron oxide nanoparticles in field applications.

Keyword: DDT, nanoparticles, degradation efficiency, pH value.

I. INTRODUCTION

The Organochlorines pesticides (OCPs) comprise the largest class of organic pollutants, including aldrin and dichlorodiphenyltrichloroethane (DDT), followed by organophosphorus (OPs) pesticides, then by flame retardants and polyaromatic hydrocarbons (PAHs) and other chemicals (polychlorinated biphenyls PCBs); the by-products of the above-mentioned compounds also constitute for the same¹⁻². India currently ranks twelfth in the world for pesticide consumption and is the leading producer of pesticides in Asia. In India, the organochlorine chemical class accounts for 40% of all pesticide applications. India has an average DDT consumption of 48 $\mu\text{g/g}$, which is rather high. However, environmental contamination and cumulative poisoning have resulted from their stable chemical structure and resistance to degradation in the natural environment.

The wide range of OCPs that are extremely hazardous are unable to be effectively eliminated by the current treatment techniques, such as oxidation, activated carbon, activated sludge, and reverse osmosis. Moreover, the by-product generated during the process of degradation may turn out to be more dangerous than the parent chemical³. Numerous studies that demonstrate the inadequacy of commonly employed conventional treatment procedures have led to the demand for sophisticated and effective methods of removing organic pollutants (OPs) from waters and wastewaters⁴⁻⁵.

The application of nanomaterials to treat environmental contaminants is known as nano-remediation. This novel method of safely removing organic pollutants like pesticides, PCBs, heavy metals, brominated chemicals, and other dangerous substances is called nano-remediation. Nanomaterials have special optical, thermal, mechanical, structural, and electromagnetic qualities that make them useful for a wide range of potential uses⁶.

In several environmental and biochemical applications, iron oxide nanoparticles are effectively used because of their low toxicity, biocompatibility, and biodegradability. Different techniques including chemistry, biology, and physical processes have been used to create iron oxide nanoparticles with altered surface chemistry⁷. Iron oxides exist in many forms in nature, with magnetite (Fe_3O_4), hematite ($\alpha\text{-Fe}_2\text{O}_3$) and maghemite ($\gamma\text{-Fe}_2\text{O}_3$) being most probably common and important technologically. It has been reported that surface effects have a strong influence on the magnetic properties of iron oxide nanoparticles.

Several studies prove the efficiency of nanomaterials for the adsorption of various pollutants, as more than 90% removal efficiency was achieved in most of the studies for up to ten cycles⁴⁻⁶.

By applying UV/visible light and appropriate nanoparticles to the contaminated site, pollutants can be gradually oxidised into low molecular weight intermediate products, which can then be converted into CO_2 , H_2O , and anions (NO_3^- , PO_4^{3-} , and Cl^-). This process of photocatalysis removes pollutants and contaminants from wastewater. Nevertheless, it works best in pH levels that are almost neutral as opposed to very basic or acidic⁸.

Therefore in present study, we assessed the reactivity of the synthesized nanoparticles towards organochlorine pesticides as DDT through batch degradation experiments under various pH values.

II. MATERIALS AND METHODOLOGY

The batch degradation experiment carried out on various pH value and degradation of the dichlorodiphenyltrichloroethane (DDT) by using synthesised iron oxide nanoparticle concentration at 5 mg/kg soil. The soil amended with 40 mg DDT/kg soil, incubated with iron nanoparticles suspension at $30 \pm 2^\circ\text{C}$. The incubation carried out on a rotatory shaker at constant rpm for 15 days. Experiments were carried out in triplicate. The suspension centrifuged, and the supernatant and the solid phase separated using Whatman filter paper for DDT analysis. DDT degradation was analysed by UV-Vis spectrophotometry³. The absorbance of the supernatant was measured using a UV-Vis spectrophotometer at various wavelengths. The degradation of DDT was estimated according to the following equation:

$$\%D = (1 - A_t / A_0) * 100$$

Where

A_0 is the initial absorbance and

A_t is the absorbance after time t at the wavelength corresponding to the peak maximum.

III. RESULTS AND DISCUSSIONS

The characteristic absorption spectrum of DDT was typically recorded in the range between 220 nm to 460 nm. The catalytic activity of the Fe_2O_3 nanoparticles on the degradation of DDT was mapped at different pH by the intensity of the DDT absorption peak at 270 nm. The degradation reaction was estimated to be completed when the absorption peak approached the baseline.

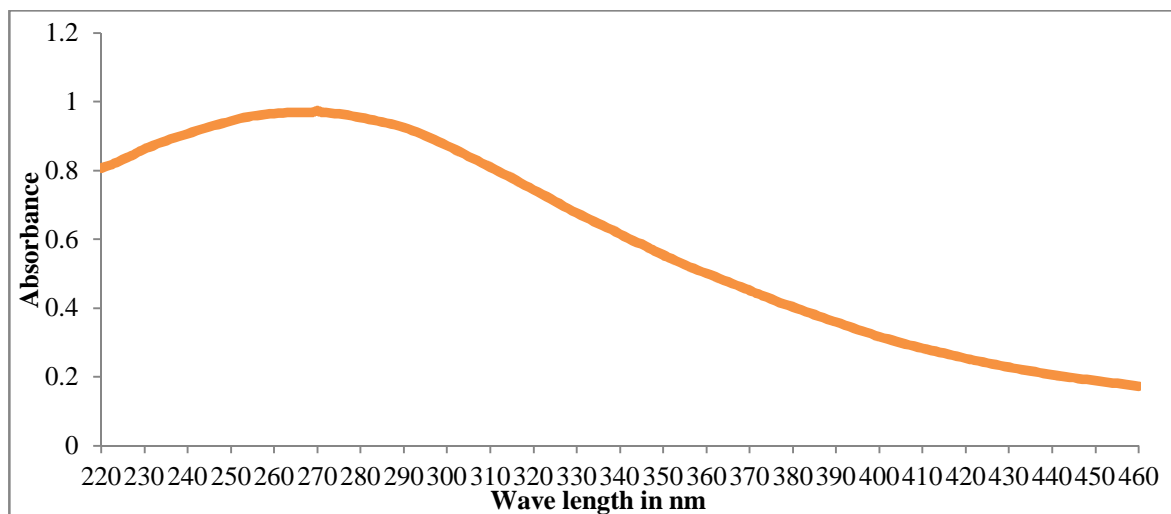


Fig-1. Recorded Absorbance for DDT concentration at different wavelength in nm

When using iron nanoparticle therapy for DDT breakdown, soil pH optimisation is essential. The pH has a crucial role in controlling the corrosion and surface chemistry of nanoparticles, which in turn determines how easily contaminants may break them down. To maintain the ideal conditions for nanoparticle activity in field applications, pH buffering or amendments can be required. pH is important for the reactivity and life-time of iron nanoparticle in water, with low pH increasing oxidation and higher pH resulting in passivation of iron nanoparticle⁹. According to studies, iron oxide nanoparticles are most reactive in pH ranges that are slightly acidic to neutral, and their efficacy drastically decreases in alkaline environments.

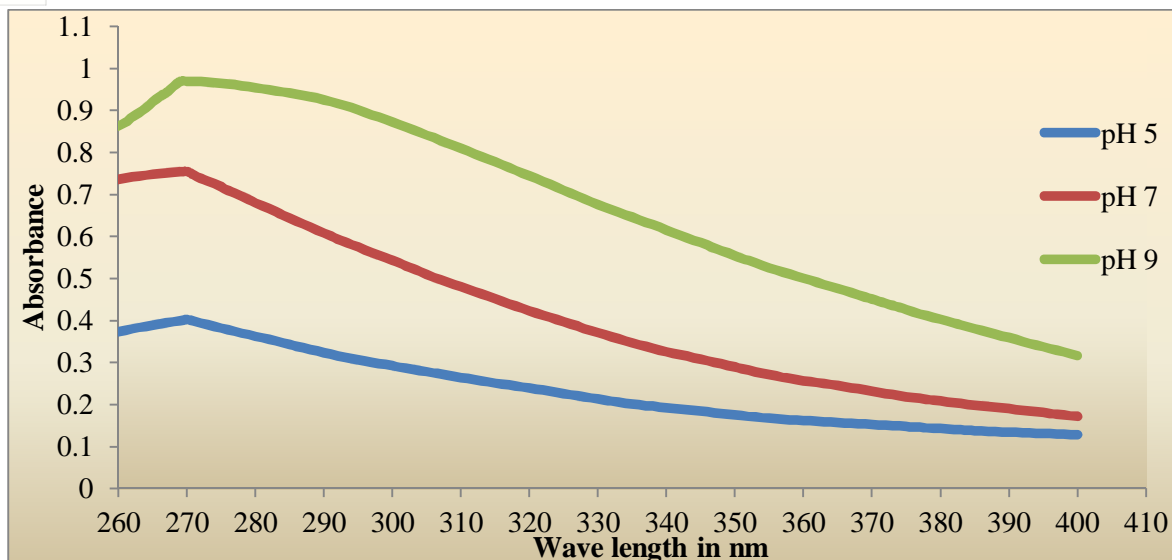


Fig.-2. Effect of variation in pH on DDT degradation by treatment of synthesised iron oxide nanoparticles

DDT degradation peaked at pH 5, at around 58.7%. In our experiments, significant degradation percentage at acidic pH is consistent with the accelerated reductive dechlorination of DDT caused by Fe(II) in acidic settings. Iron oxide nanoparticles corrode more readily and produce more reactive Fe(II) species at acidic pH values (around 5), which improves DDT's reductive dechlorination. Increased DDT breakdown results from this. Iron oxidation and the production of reactive species, which propel the pollutant transformation, are made easier by the acidic environment⁵.

Table-1 Effect of pH on DDT degradation by treatment of synthesised iron oxide nanoparticles

pH value	A_0	A_t	A_t/A_0	$1-A_t/A_0$	DDT degradation in %
pH 5	0.9741	0.4024	0.413099	0.586901	58.69007
pH 7	0.9741	0.7549	0.774972	0.225028	22.50282
pH 9	0.9741	0.9698	0.995586	0.004414	0.441433

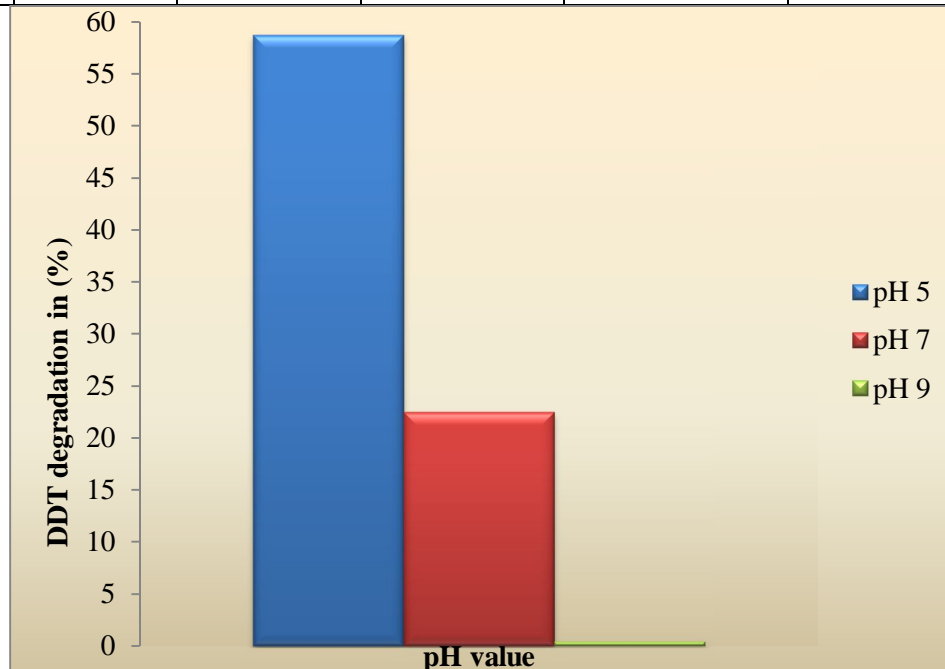


Fig.-3. Effect of variation in pH on DDT degradation by treatment of synthesised iron oxide nanoparticles

Degradation decreased to around 22.5% at neutral pH 7. At neutral pH (7), the activity is moderate. An oxide/hydroxide layer on iron oxide nanoparticles may stabilise their surface, decreasing reactivity and, as a result, degradation rates. For many iron oxides, this pH is close to the point of zero charge, which reduces reactive electron transfer or surface contact with DDT.

Degradation was over 0.44% at alkaline pH 9. Degradation is significantly reduced in alkaline circumstances (pH 9). Fe(II) release is reduced and reactivity is low when the passivation layer deepens and iron nanoparticle corrosion slows down significantly. Poor DDT degradation is the outcome of this. Higher pH additionally promotes the formation of iron oxides, which may hinder reductive degradation-related electron transfer activities¹⁰⁻¹¹. Because of the underlying chemistry of redox processes and the surface characteristics of the nanoparticles, soil pH has a major impact on the breakdown efficiency of DDT with iron oxide nanoparticles¹². Similarly Liu and Lowry (2006) studied the reactivity and life-time of two nZVI products, their results showed that nZVI remained reactive for several months at pH 8.9 but for only a few weeks at pH 6.5, while the nZVI made using BH_4 was fully oxidized in less than two weeks at pH 8.9. The reaction with oxygen and water increases pH, presumably most so for nZVI due to their smaller particle size¹³.

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