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# A Novel Strategy for Remediation of Heavy Metal Removal and Oil/Water Separation from Carbon-Based Nanohybrid

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## ABSTRACT

**Increasing the world population results in the consequent consumption of primary resources, which produces high amounts of waste. The increasing amount of waste has negatively impacted the environment and ecosystem. The growing population demands a high amount of food resources, which increases anthropogenic activities. So, Environmental pollution has become a severe problem due to this anthropogenic activity affecting our planet. This problem cannot be neglected and has become the most challenging task of the 21st century. Researchers are putting effort into developing new nanohybrid, which should be environmentally friendly, with greener methods of synthesis to overcome and remediate the environment from pollutants. Scientists are trying to move towards nanotechnologies and nanomaterials to resolve the upcoming challenges related to environmental pollutants. This article describes heavy metal pollutants, recovery of oil from wastewater, etc. This article will also focus on the carbon-based nanohybrid, which can be used for the environmental recovery strategies of polluted areas. In particular, this article will give more attention towards the recent method developed and method to capturing heavy metal using silver fabricated reduced graphene oxide nanohybrid for the application of heavy metal capture and separation of oil water.**

***Keywords-*** heavy metal**,** oil recovery, environmentalremediation, Nanotechnology, nanomaterials, nanocomposites.

## I. INTRODUCTION

Environmental pollutants and contaminants due to human activities linked with agriculture and industries are increasing the dangerous and adverse effects on the ecosystem and human health [1-3]. In this aspect, our researchers are trying to handle and study the places contaminated with the pollutants emerging in the last century. As the population is increasing daily, which has significantly increased anthropogenic activities, heavy metal pollutants have become a severe problem for human beings and the ecosystem [4-6]. Heavy metals are one of the prime pollutants due to the mining of metals, smelting, and other metal-based activities, such as the leaching of metal from various sources like landfills, waste dumps, excretion, etc. [7-8]. Heavy metals are one of the persistent pollutants which result in the accumulation in the environment and contaminate the food chain constantly [9]. Accumulation of heavy metals in high amounts in the environment causes a potential health threat to the ecosystem and human beings [10]. Heavy metals have an atomic number larger than 20 and an atomic density of more than 5 $gm.cm^{-3}$ [11-12]. Example of heavy metals is Arsenic (Ar), Chromium (Cr), Copper (Cu), Cadmium (Cd), lead (Pb), Mercury (Hg), Nickel (Ni), Uranium (U), Manganese (Mn), etc. [13].

Another crucial environmental problem in the 21st century is oil spillage, which has become an

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essential environmental catastrophe [14,15]. Oil spillage is one of the biggest problems resulting from human activities via the process of release into coastal water and land [16,17]. Many reports say that this oil spillage problem is also coming into the picture at a larger level in developing countries. Still, the data is not being reported in a proper manner. Even if few reports are there on google, no effort is being made to restore the ecosystem to its previous state even when the oil spills are accounted for. It has been reported that crude oil spills damage the ecosystem all across the world [18-20]. The spillage of offshore oil has become one of the major concerns due to its hazardous impact on marine life [21-23]. There are many conventional countermeasures for oil spill remediation in marine life, including physical, thermal, chemical, and biological [24]. The biological method is one of the promising methods which uses native microorganisms in the water and soils [25]. Native microorganisms' biological methods are highly preferred in water as well as soil because these microorganisms utilize living organisms to carry out the remediation process in polluted sites [26]. However, biological methods also have some disadvantages, but this method has proven to be highly sustainable and economical. Figure 1 shows the most common environmental contaminants emerging in the last few decades.

**Figure 1: Environment pollutants and contaminations due to the anthropogenic activities**

Most of the reported remediation and waste management techniques require high cost of chemicals and high energy consumption; for example, in the case of wastewater treatment plants produce a source of secondary pollution [27-28].

To tackle this serious environmental problem, nano-materials and nanotechnologies are become emerging fields due to their unique properties such as high surface-to-volume ratio, good catalytic capacity, size and shape effect, and many more. It also has been observed that the properties of the nanoparticles can be easily tuned chemically or physically using the surface functionalized technique. The nanoparticles can be easily embedded into specific polymers and matrices to produce hybrid nanocomposites, which can provide specific chemical, mechanical, and physical properties. By altering their properties. This article describes the most recent innovations based on various nanomaterials, nanocomposites, and nanohybrid for environmental recovery strategies for the heavy metal pollution and oil-water spillage problems. This article will focus on eco-friendly and regenerative nano solutions and their safe-by-design properties to address the recent innovation activities toward sustainable strategies in environmental remediation and bioremediation [29-31].

## II. MATERIALS AND METHOD

All the chemicals were purchased from sigma and TCI company. Graphite powder was purchased from TCI. Sulfuric acid, potassium permanganate, hydrogen peroxide, Hydrochloric acid, hydrazine hydrate, and silver nitrate were provided by Sigma-Aldrich and TCI company. All other reagents are of analytical grade and were used without further purification. Deionized water (DI) was used throughout the complete experimental work.

## III. CHARACTERIZATIONS AND MEASUREMENTS

The surface morphology of the reduced graphene and silver nanoparticle-doped graphene hybrid was observed using a TEM. The contact angle measurements of the reduced graphene and silver nanoparticle doped graphene hybrid was determined using an OCA 40 Micro under normal conditions by using the sessile drop method. In brief, a drop of water was dropped using the pipette on the surface of the reduced graphene and silver nanoparticle-doped graphene hybrid. The contact angle was measured from the video camera, and images of the drop were taken at 0 seconds and 12 seconds.

## IV. SYNTHESIS OF AUNPS MODIFIED WITH GRAPHENE OXIDE

The first step was synthesizing reduced Graphene oxide using chemical exfoliation of Graphite, using the hummer method. In brief, Concentrated sulphuric acid was added into a filled with graphite at room temperature. The reaction mixture was cool at 0°C using an ice bath. $KMNO_4$ was added slowly to the reaction mixture slowly. The reaction mixture was taken to room temperature and stirred for two hours. After two hours, hydrogen peroxide was added to the reaction mixture until the gas evolution ceased. The resulting suspension was washed using diluted hydrogen chloride followed by

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distilled water and was also centrifuged to remove the excess unexfoliated graphite. The resulting solution was dried lyophilized and dried. It was referred to be graphene oxide. Afterward, the oxidized GO was treated thermally using the argon atmosphere. The Oxidized GO was placed in the furnace at 1100 °C to synthesize the reduced graphene for a period of 6 h. The Graphene oxide was reduced using hydrazine. GO was taken into an RB in the distilled water and ultrasonicated to become the homogenous yellow-brown dispersion. The dispersion was kept for sonication until a clear solution was observed. Hydrazine hydrate was added into the solution at 100 °C and kept for heating for 24 h. After 24 h, reduced graphene RGO was gradually precipitated out as a black solid. The product was isolated using centrifuged and washed with water a couple of times. The resulting solution was referred to as reduced graphene oxide.

## V. SYNTHESIS AND CHARACTERIZATION OF GRAPHENE/SILVER NANOCOMPOSITES

Graphene/silver nanocomposite was synthesized as follows. First, the gold(III) complex was reduced using sodium citrate. After that, graphene aqueous solution was added to the above solution, and the reaction mixture was kept stirring for 24 h. This promotes the interaction of gold ions with the graphene surface. Figure 2 shows the TEM images of reduced graphene and silver fabricated of reduced graphene oxide nanoparticles.

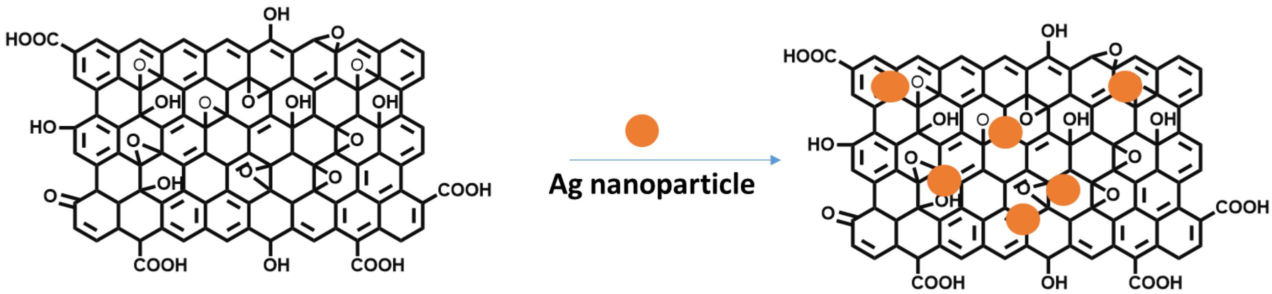


**Figure 2: Synthetic scheme of the formation of silver-graphene oxide nano-hybrid**

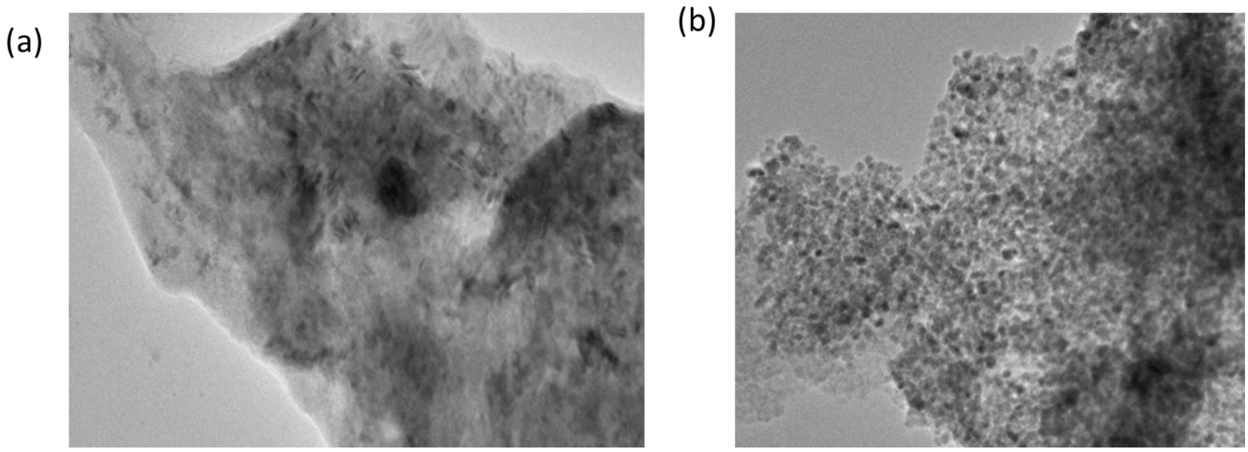


**Figure 3: TEM images of Reduced Graphene oxide (a) and Reduced Graphene oxide coated silver nanoparticles (b)**

## VI. CARBON-BASED MATERIAL TO DETECT THE HEAVY METAL

Silver nanoparticles and AuNPs modified with graphene oxide are used to detect the heavy metal in the water waste bodies. This particular hybrid material is able to detect the heavy metal such as $Cd^{2+}$, $Pb^{2+}$, $Cu^{2+}$, and $Hg^{2+}$ with a particular detection limit of 19.01, 47.55, 22.09, 29.30 $\mu A.\mu M^{-1}cm^{-2}$, respectively as shown in Table 1.

**Table 1: A detection limit of heavy metal using AuNPs modified with graphene oxide nanohybrid**

| | Heavy metal | Detection limit ($\mu A.\mu M^{-1}cm^{-2}$) |
|---|---|---|
| 1 | $Cd^{2+}$ | 19.01 |
| 2 | $Pb^{2+}$ | 47.55 |
| 3 | $Cu^{2+}$ | 22.09 |
| 4 | $Hg^{2+}$ | 29.30 |

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## VII. APPLICATION OF CARBON-BASED MATERIAL IN THE SEPARATION OF OIL/WATER MIXTURE

A field of remediation of the contaminated aquatic sites from the oil is one of the serious concerns. For this purpose, many researchers are trying to develop an economical and easier method to separate oil from water in aquatic sites. In this aspect, carbon-based monohybrid material can also be utilized for this problem. The hydrophilic and hydrophobic properties of carbon-based material can also be one of the aspects to tackle this problem. With carbon-based nanomaterials, π-π interactions and electrostatic interaction can also play a major role in this problem. Carbon-based material such as graphene oxide and reduced graphene oxide also has oxygen functionalities and hydrogen bonding interactions which can be used to trap water and can be used to separate from the oil at a bigger scale. So, doping the Ag nanoparticle on the graphene oxide can increase the adsorption efficiency and thereby be developed to remediate contaminants such as oil spilled on water.

## VIII. HYDROPHOBIC PROPERTIES

The wettability and water-repellent properties are one of the most important features for the application of separation of oil-water. The wettability of the reduced GO and GO-coated silver nanoparticle was measured using water contact angle measurement. As shown in figure 4, it can be seen that reduced GO exhibit a water contact angle of 0°for the smooth and capillary effect. However, by comparing the contact angle of silver-doped GO nanohybride shows the significant hydrophobicity of the hybrid material. The WCA value of the hybrid material was found to be 122.7°. The improvement of hydrophobicity of the nanohybrid is mainly attributed to the following reasons. First, strong interaction and good compatibility between the GO and Ag nanoparticles after doping the silver nanoparticle into the reduced graphene oxide. Second is the reduction of the hydroxyl group due to the doping of silver at high temperatures. So the physical interaction between silver nanoparticles and reduced graphene oxide results in the hydrophobic behaviour of the nano-hybrid, and the water contact angle was found to be significantly increased. Most of the oxygen functionalities were removed after the incorporation of silver nanoparticles.

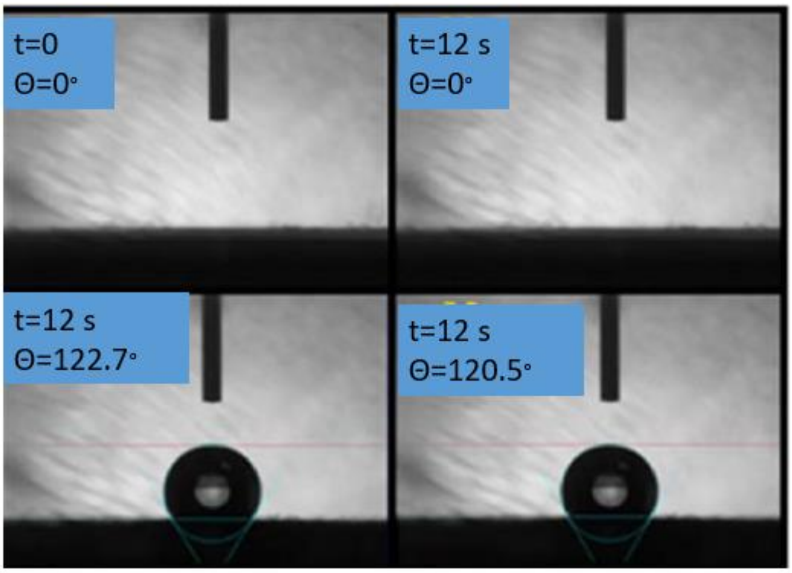


**Figure 4. Contact angle (CA) of water droplets of reduced graphene oxide and silver-doped reduced graphene oxide hybrid**

**Table 2: Water contact angle of reduced graphene oxide and silver doped reduced graphene oxide hybrid**

| Sample | Contact angle at 0 s (degree) | Contact angle at 12 s (degree) |
|---|---|---|
| Reduced GO | 0 | 0 |
| Ag-Reduced GO | 122.7 | 120.5 |

More interestingly, the fabrication of silver nanoparticles into reduced graphene oxide significantly affects the water contact angle, as shown in figure 2 and table 2. It also can be noted that the contact angle also did not change even after 12 s of time of reduced graphene oxide. Similarly, the contact angle of silver-doped reduced oxide nanohybrid did not change much from 122.7 to 120.5 degrees. From

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this experiment, it can also be observed that this hybrid can potentially be applied for oil/water separations.

## IX. CONCLUSION

In this article, the silver nanoparticle fabricated reduced graphene oxide nanohybrid was successfully synthesized and characterized using TEM analysis. The as-synthesized nanohybrid was used for heavy metal detection. It was concluded that the nano-hybrid was successfully able to detect with a very low-level limit. Further, the nano-hybrid was also assessed for the application of oil/water separation using the contact angle measurement. Significant increases in the contact angle were observed, indicating that hydrophobicity increases in the nanohybrid. So, the nano-hybrid can be useful for the environment in terms of removing heavy metals and applying oil-water separation.

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