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Preparing Some Nano Composites Decorated with Some Pharmaceutical Compounds and Evaluating their Biological Effectiveness

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ABSTRACT: The aim of this study was to use electrochemical methods to synthesise and characterise several derivatives of graphene oxide or nanosheets of reduced graphene oxides (A3) and (A4). Melting-point analysis was performed on the produced compounds. SEM, XRD, TEM, and AFM. It's not environmentally friendly, but it's quick and doesn't cost a lot of money. All of the separated chemicals can be stored safely at room temperature. Bacteria have performed biological research on compounds created using the nanotechnology approach. And assess the produced compounds' capacity to limit bacterial growth and their biological efficacy against Gram-positive and Gram-negative bacteria.

KEYWORDS: Graphene, Reduced Graphene Oxide, Triazole, Gram bacteria

1. INTRODUCTION

Nanomaterials are considered advanced chemical materials that can be produced with dimensions ranging from 1 nm to 100 nm. The small sizes and scales of these materials have led them to behave differently from traditional large-sized materials whose dimensions exceed 100 nm, and to have strong qualities and properties. Excellence cannot be found together in traditional materials, and the smaller the scale of nanomaterials, the greater the effectiveness. [(1)]

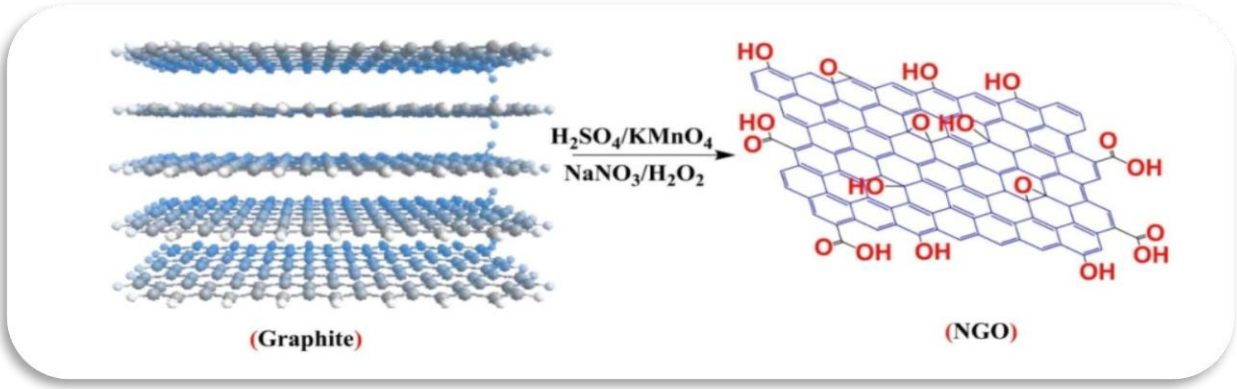
Nano size is described with many examples, such as being the same width as deoxyribonucleic acid or the size of ten hydrogen atoms,[(2)] and nanotechnology is the application of scientific concepts and methods in the process of development at the nanoscale level, although nanotechnology is relatively new, as it is considered a revolution. Industrial fields in the electronic, industrial, military, medical and other vital fields that greatly affect our lives.[(3)]

The emergence of new applications for nanotechnology has enabled researchers to control cell behavior and repair and develop human tissues, and it has also provided services in the field of energy and water purification.[(4)] Nanotechnology has also been used to study the structure of cancer cells and treat them.[(5)]

2. MATERIAL AND METHODS

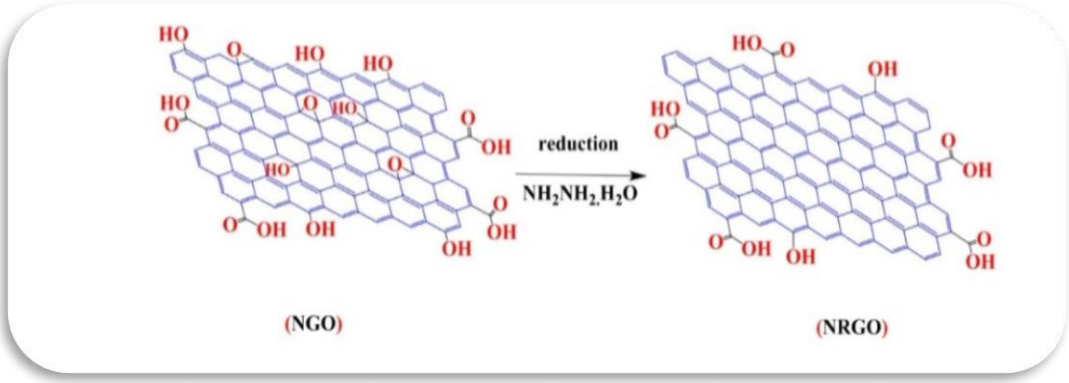
2.1 Synthesis Graphene Oxide (GO)

In a snow bath, 46 mL of concentrated sulfuric acid (H_2SO_4) was introduced to a 600 ml beaker while magnetic stirring was maintained. After 15 minutes of stirring at 0 C°, 1 g of graphite was added progressively over the course of 10 minutes. Then, over the course of 10 minutes, while keeping the temperature below 10 C° for two hours, add 6 g of potassium permanganate while stirring slowly and carefully. Finally, remove the mixture from the ice bath and add 46 ml of distilled water while stirring slowly and carefully for 15 minutes. The temperature is 98 C°, and then 140 ml of distilled water is added to bring it down to a comfortable but still warm 50 C°. Ten minutes later, add 15 ml of 30% hydrogen peroxide H_2O_2 and mix for another 30 minutes. The next step is to add 150 ml of distilled water to each half of the mixture. Finally, after 24 hours, drain the mixture and wash it once with 10% (HCl) to remove any remaining precipitate. The acidic function (pH=7) was attained after drying at 60–70 degrees Celsius and five washes with anionic water. [(6)]



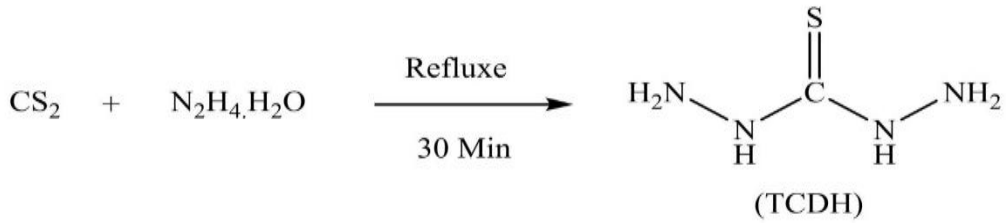
2.2 Synthesis reduced graphene oxide

In a heat-resistant circular flask of 50 ml, place 0.1 g of graphene oxide prepared in the Hummer method and add 1 ml of hydrochloric acid and keep it on constant stirring until it becomes a clear solution without plankton until homogenization, add 1 ml of hydrazine hydrate 80% was heated at 100 C° for 2 hours in the presence of an inverter condenser, collected NRGO by filtration, and washed with deionzed water three times to get rid of excess hydrazine, where it was dried in an oven at 100 C° for 12 hours. [7]



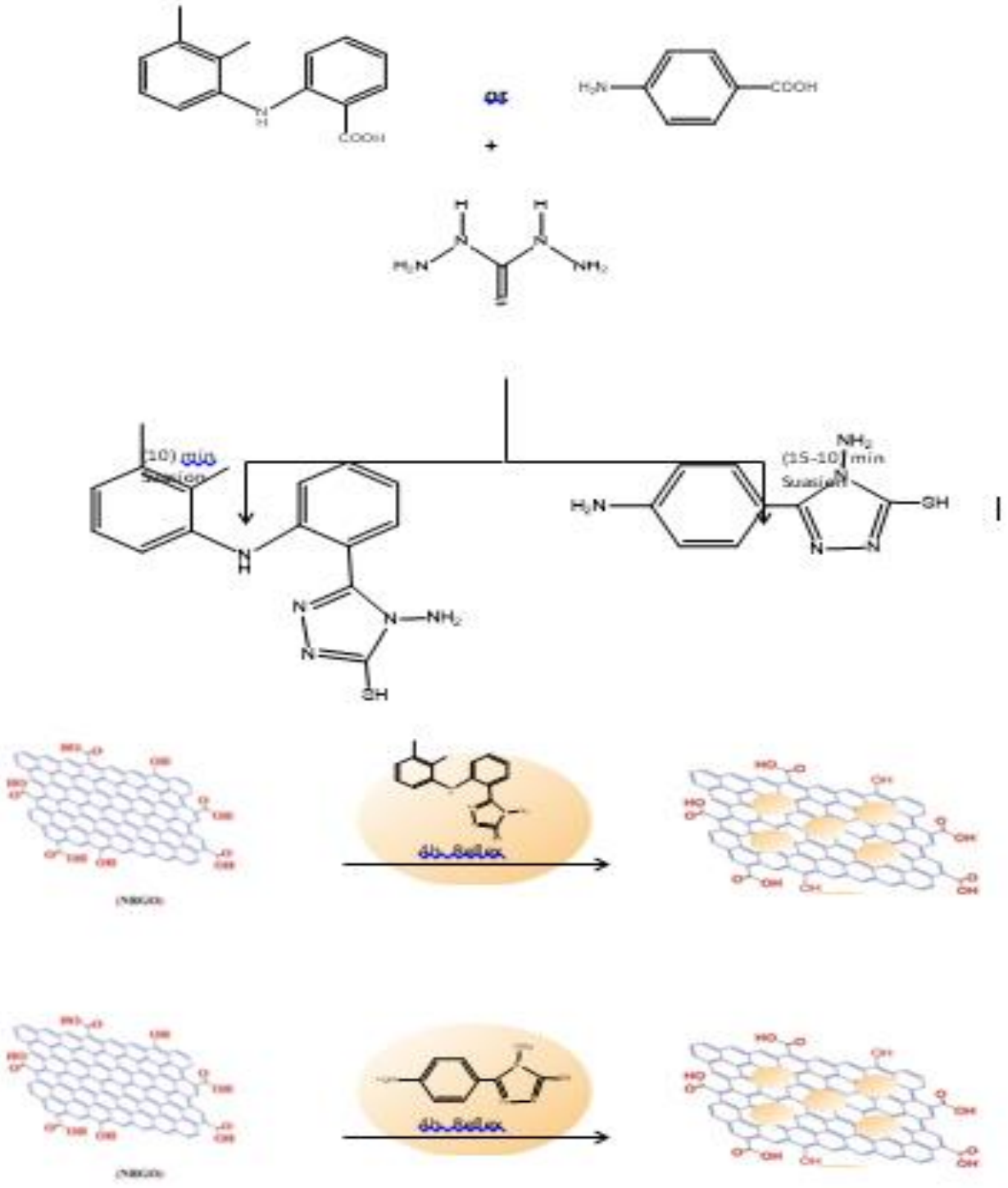
2.3 Synthesis Thiocode di Carbohydrazide TDCH

A 100 ml round flask was placed in an ice bath, 20 ml of 80% hydrazine was added, and 5 ml of carbon disulfide was added gradually over the course of 10 minutes with magnetic stirring. After 30 minutes, a yellow precipitate formed, and the mixture was cooled back down to room temperature. It was dried at 50 C° after being recrystallized with distilled water. The melting point was measured at (172 - 170) C° [8], which agrees with the published data. 66% of the total product. [9]



2.4 Synthesis 4-Amino-5-Phenyl-4H-1, 2, 4-Triazole-3-Thiol

0.01 mole of pyridine-6,2-dicarboxylic acid was mixed with 0.02 mole of TDCH (or 0.01 mole of benzene-5,3,1-tricarboxylic acid with 0.03 mole of TDCH) in a heat-resistant flask and heated in an oil bath with Stir for a period of (10-15) minutes at the melting point. The product was cooled to room temperature until the consistency and color of the melt changed. Then it was treated with a 10% sodium bicarbonate solution. The product was collected by filtration and washed with ethanol several times until the acidity of the filtrate disappeared and it was dried in a temperature oven 60 C°. [10]



2.5 Synthesis of reduced graphene oxide nanocomposites

Ultrasound at 50 Hz for 30 minutes is applied to 0.2 mole of the prepared nano-reduced graphene oxide (A2) in 10 ml of distilled water, then, 0.2 mole of one of the selected aromatic amines is added; the mixture is heated for 4 hours at 100 C°, finally, the product is collected, it is washed three times with 10 ml of deionized water; and it is dried in the oven at 80 C° until the weight is constant. [11] [12]

2.6 Biological application of graphene methyldopa compounds

2.6.1 Synthesis an antibacterial medical mask based on graphene pharmaceutical compounds [13]

Synthesis of colloidal solution of methyldopa [A3,A4] at different concentrations (50, 100, 150) ppm From the solution of methyldopa, a commercially purchased face mask was soaked in 50 ppm and 100 ppm solution and 150 ppm.

2.6.2 Synthesis of colloidal solution using Methylcellulose

A beaker containing 150 ml of deionized water at 90 C°, sprinkle 0.8042 g of methylcellulose (dynamic viscosity of a 2% solution 4000 cl) on it and stir vigorously to ensure that the substance is dispersed in the solvent, then cool the solution in an ice bath with constant stirring until it forms A clear colloidal solution. 0.02 grams of the nanomaterial was dispersed in a beaker containing 50 ml of deionized water for 60 minutes. Then, methylcellulose was added to the colloidal material after cooling. The reaction mixture was kept for (15-20) minutes at room temperature, then added to Mix 3-5 drops of diluted hydrazine solution (1 mL in 100 ml of DI water), stir the solution for 30 minutes at room temperature, and finally the formation of a silver colloidal solution of methyl-dopa nanopharmaceutical compounds can be observed.[14]

2.6.3 Synthesis the colloidal solution using Xanthan gum

A beaker containing 150 ml of DI water at laboratory temperature, sprinkle 0.8042 g of Xanthan gum (dynamic viscosity of a 2% solution 4000 cl) on it and stir vigorously to ensure that the material is dispersed in the solvent, and a clear colloidal solution is formed, and 0.02 g of the nanomaterial is dispersed In a beaker containing 50 ml of deionized water for 60 minutes, then add the colloidal substance of DI water and stir the solution for 30 minutes at room temperature, and finally the formation of a silver colloidal solution of methyl-dopa pharmaceutical nanocomposites can be observed.[15]

2.6.4 Synthesis a medical mask containing the nanocolloidal solution

From the concentrated solution of methyldopa nanocompounds, three different concentrations (100, 50, and 150) ppm were prepared. In the previously prepared solutions, small previously prepared pieces of masks measuring (55) cm in size began to be soaked for a period of (5-7) hours. At room temperature, then the pieces of masks were dried in room air for 30 minutes. Face masks treated with a colloidal methyl-dopa nanopharmaceutical solution will be further used in antimicrobial studies against Gram-positive and Gram-negative bacteria, as a mask treated with a 50 ppm solution is described as A50. While a mask treated with a 100 ppm solution is labeled A100, and a mask treated with a 150 ppm solution is labeled A150. [16]

2.7 Bacterial biological activity

2.7.1 Bacterial isolates

In this study, two types of Gram-positive bacteria and two types of Gram-negative bacteria were used. They were obtained and diagnosed from the Department of Biological Effectiveness at the General Company for Pharmaceutical Industry in Samarra. They were directly activated before use. The Gram-positive bacteria are Staphylococcus aureus. Gram-negative bacteria are Bacillus and Salmonella.

2.7.2 Synthesis of Bacterial inoculation

The bacterial suspension was prepared from a 24-hour-old colony, which was done by taking a smear of the isolated bacteria using a lube, and according to the type of bacteria to be worked on, it was placed in the prepared solution, Normal saline solution, in an amount of 10 ml, and incubated for 24 hours. [17] [18]

2.7.3 Synthesis bacterial plates to test the biological activity of the prepared compounds

The biological effectiveness of the prepared compounds was estimated according to the method (United States Pharmacopeia USP) under the antibiotics-microbial assays section, and the work was done using the cylinder plate method. Which is summed up by preparing a nutrient medium consisting of nutritional compounds for bacteria. [19] [20]

It was prepared and sterilized in an autoclave, and it consists of:

The acidity level (pH) after sterilization was (0.1 ± 6.6), after which the medium was cooled to a temperature of (37-40) C°, and the bacterial suspension prepared a day before was added, which was done by taking a swab of the bacteria isolated with a loop, and according to the type of bacteria to be worked on, and it was placed In a liquid nutrient medium prepared in advance in an amount of 10 ml and incubated for 24 hours, then an amount of (1-2) ml of the suspension is added to the basic nutrient medium, which usually has a volume of 100 ml. After placing the bacteria, the medium is poured into Petri dishes in an amount of 20 ml for each dish, then It cools and then makes discs of masks with a diameter of 6 mm, which were prepared in advance as mentioned in paragraph 4.6.2, and which were immersed in three concentrations of the prepared compounds (50, 100, 150) ppm, in addition to using a standard antibiotic for comparison purposes. The inhibitory effect is Gentamicin sulfate at the same concentrations above. It is an antibiotic effective against both Gram-positive and Gram-negative bacteria. It has many medical uses and is a standard substance approved in the quality control laboratories of the General Company for Pharmaceutical Manufacturing in Samarra, where it was grown on culture media in a manner (Antibiotic Disc) The dishes were incubated in an incubator at 37 C° for 24 hours, then the diameter of inhibition was read with a measuring device (zone reader), and then the diameter of the inhibition zone was taken in mm to determine the extent of antibacterial effectiveness.

3. RESULT AND DISCUSSION

3.1 Synthesis and characterization of graphene oxide nanosheets

X-ray diffraction of graphene oxide, GO (A1), showed an angle value of 2Θ=11.82° and a distance between the layers of d=0.74. Increasing the distance between the layers indicates oxidation, and then the grain size is calculated, which is equal to 5.43. [21]

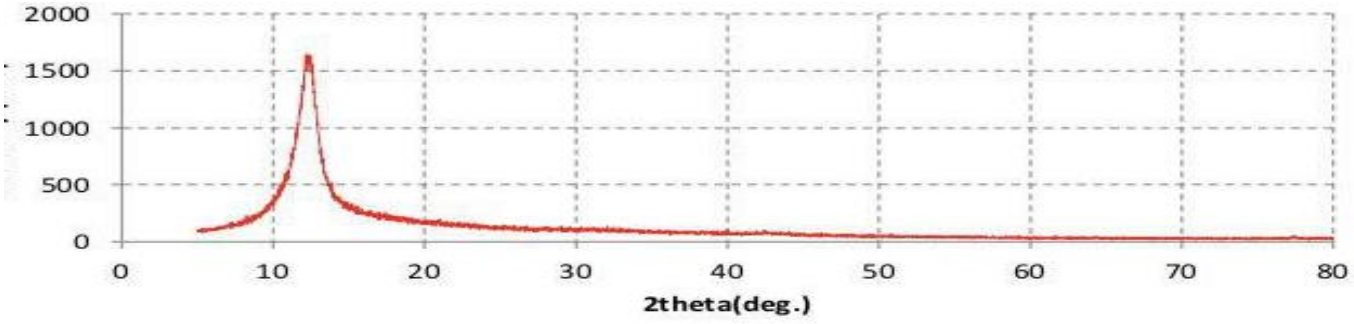


Figure (3-1) shows the XRD spectrum of compound (A1)

The SEM morphological images of compound (A1) show a peeling process resulting from oxidation and separation between the layers. Oxidation also appears on the edges and surfaces, with greater clarity of the rings on the plates.

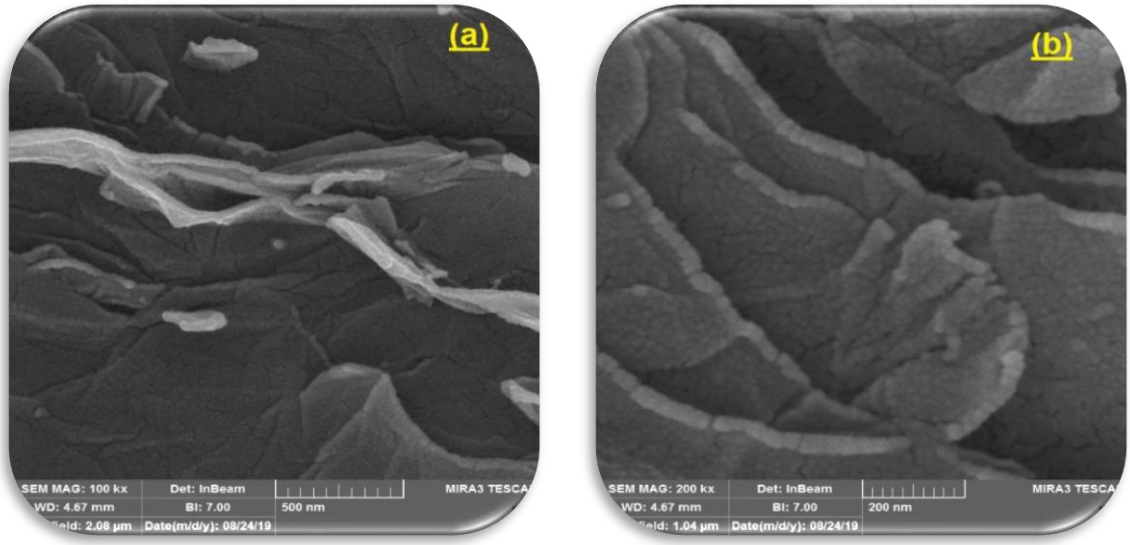


Figure (3-2) shows SEM images of the compound (A1)

AFM images of compound (A1) show a large sheet area (a) with the peeling process evident from observing the height of the sheet edges (b)

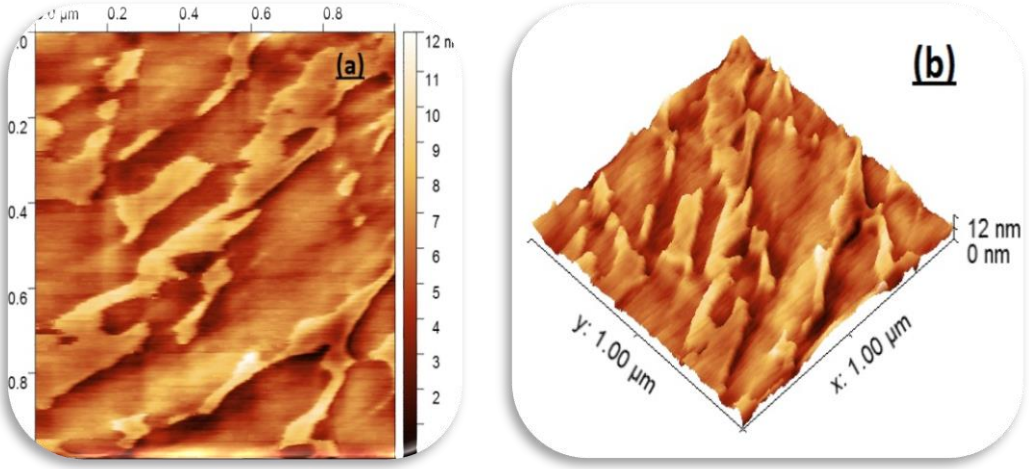


Figure (3-3) shows AFM images of the compound (A1)

TEM images clearly showed the presence of high peeling (a), where supporting materials were collected on the surface of the sheets and their edges (b), with excellent transparency commensurate with the clear peeling of the layers, which sometimes reached a single shot (c).

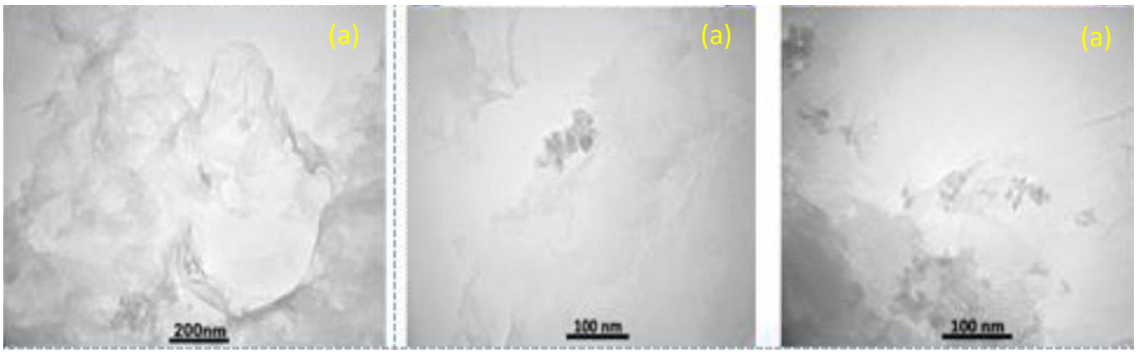


Figure (3-4) shows TEM images of the compound (A1)

3.2 Synthesis and characterization of reduced graphene oxide

The X-ray diffraction of RGO (A2) shows an angular value at 2Θ=32.26°, indicating the arrangement of the sheets along the stacking direction. This confirms that the sample is made of a few layers of RGO, with an interlayer distance of 0.74, which is smaller than GO due to the removal of most of the functional groups. [22]

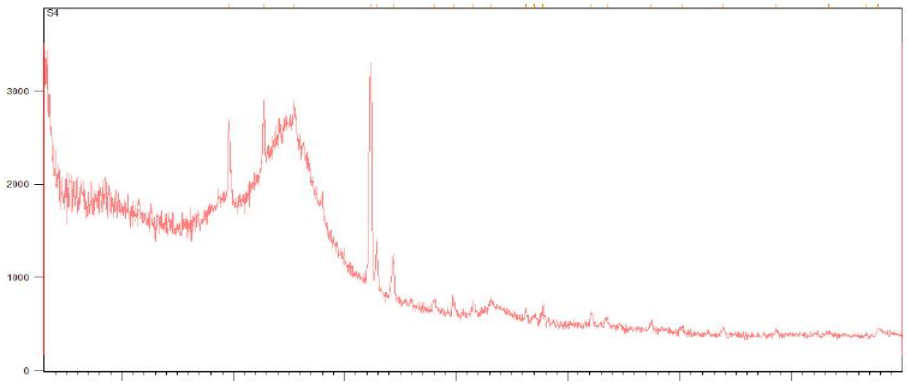


Figure (3-5) shows the XRD spectrum of the compound (A2)

SEM images of compound (A2) showed a decrease in the thickness of the sheets to 27.07 nm (a), with clear wrinkles on the sheets, while the oxidation state remained at the edges and some areas (b).

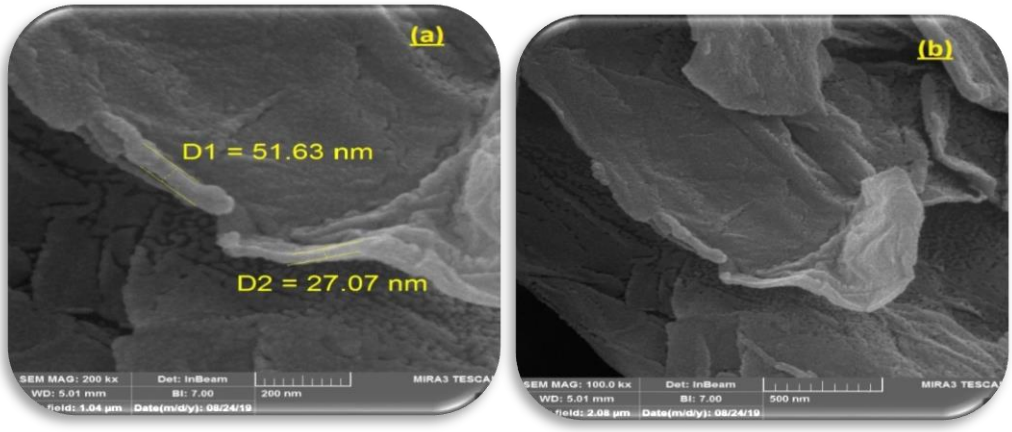


Figure (3-6) shows SEM images of the compound (A2)

As for the AFM images, they showed that the RGO (A2) in some areas on the surface still retained the oxygen groups (a) (b).

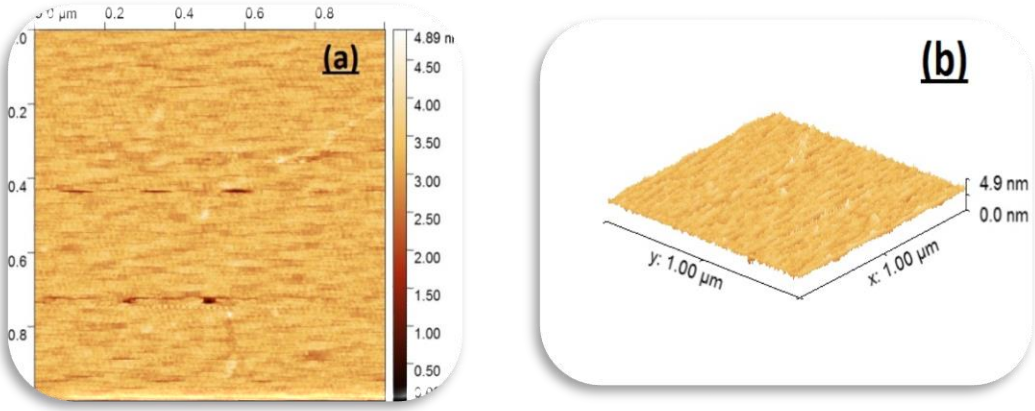


Figure (3-7) shows AFM images of the compound (A2)

TEM images revealed high peeling with high transparency (a), with obvious peeling layers (b) and reduced sheet cracks (c).

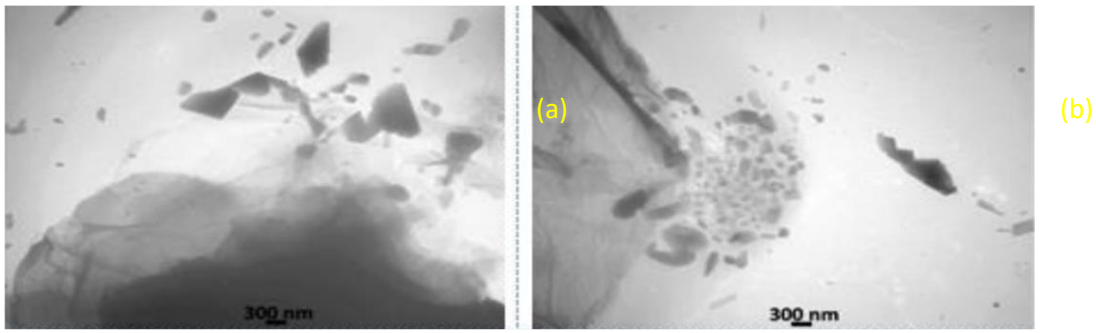


Figure (3-8) shows TEM images of the compound (A2)

3.3 Synthesis and characterization of reduced graphene oxide

With greater penetration and exfoliation, the X-ray spectra of compound (A3) showed an angle value of 2Θ=27.16°, indicating a decrease in both the interlayer distances (D = 3.1804 A°) and the grain size (D = 1.7569 nm).

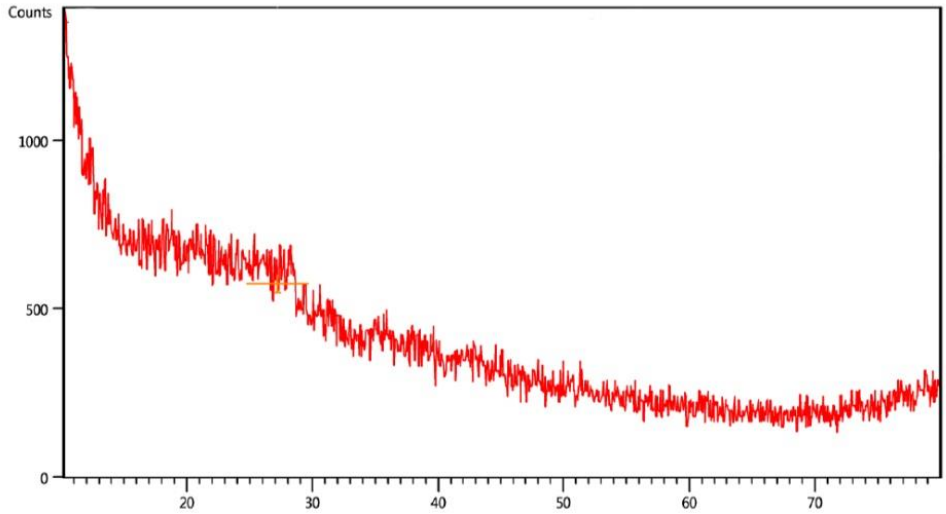


Figure (3-9) shows the XRD spectrum of the compound (A3)

The SEM images showed a roughness of up to 280 nm, and this corresponds to an increase in the surface area of 26.7418 nm^2 with an increase in the thickening of the plate edges of 35.1540 nm with an increase in cracks.

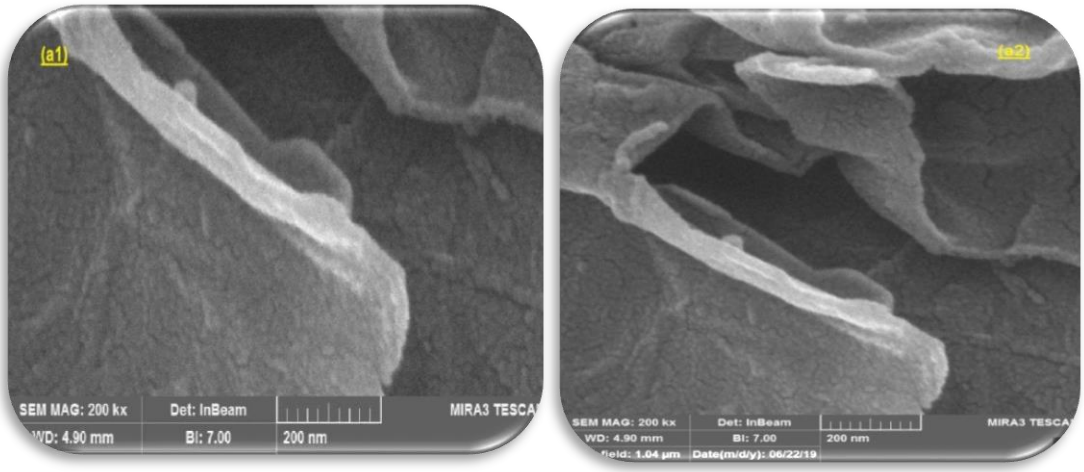


Figure (3-10) shows the SEM of the compound (A3)

The AFM images showed that the material to be overlaid with reduced graphene oxide spread well (a), in addition to maintaining the large area of the sheets and the distances between them (b).

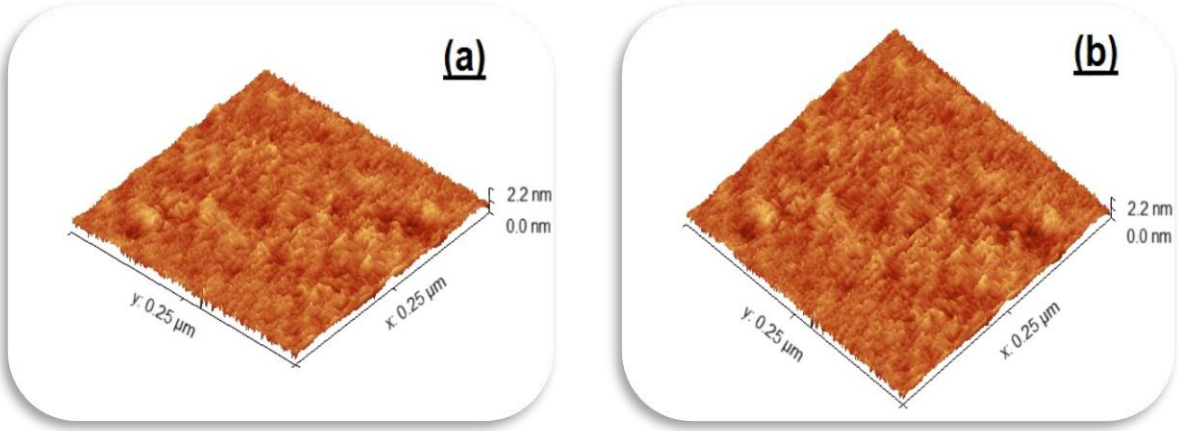


Figure (3-11) shows the AFM of the compound (A3)

TEM images clearly showed high peeling (a) as the support material collected on the edges (b) with a transparency commensurate with the clear peeling of the layers.

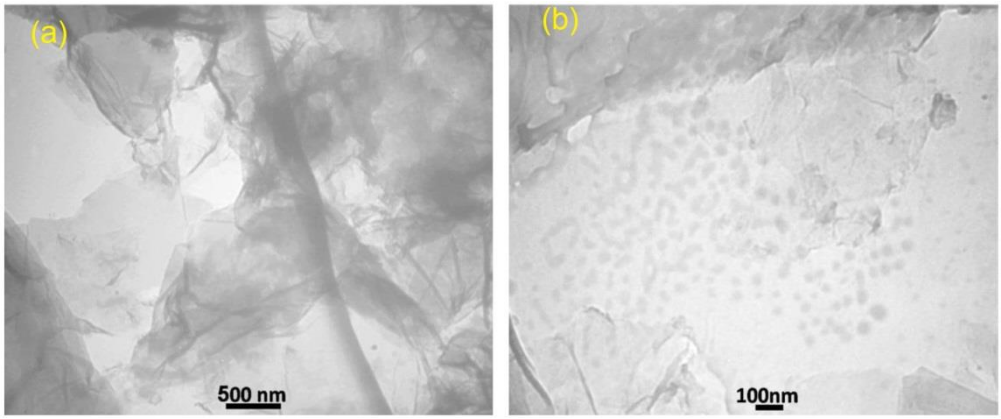


Figure (3-12) shows the TEM of the compound (A3)

The X-ray spectrum of compound A4 showed an angle value of 2Θ = 26.4522, with the presence of d layers equal to 3.3694 Ao, the grain size D equal to 19.5540, and the number of layers n equal to 5.8034. We notice a convergence between the distances for compounds A3 and A4, and we notice that the grain size of compound A4 is equal to compound A1.

3.4 Synthesis and characterization of reduced graphene oxide

The X-ray spectrum of compound (A4) showed an angle value of 2Θ = 26.4522°, with the presence of d layers equal to 3.3694 A°, the grain size D equal to 19.5540, and the number of layers n equal to 5.8034. We notice a convergence between the distances for compounds (A3) and (A4).

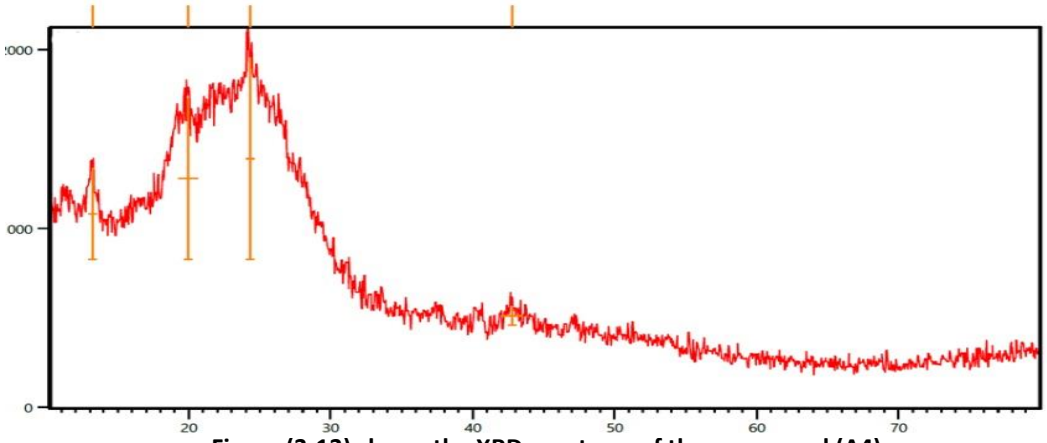


Figure (3-13) shows the XRD spectrum of the compound (A4)

From the SEM images of compound (A4), we notice the presence of high roughness, reaching 230 nm, and this is proportional to an increase in the surface area and an increase in thickening at the edges of the plate, and this explains the presence of cracks. [23]

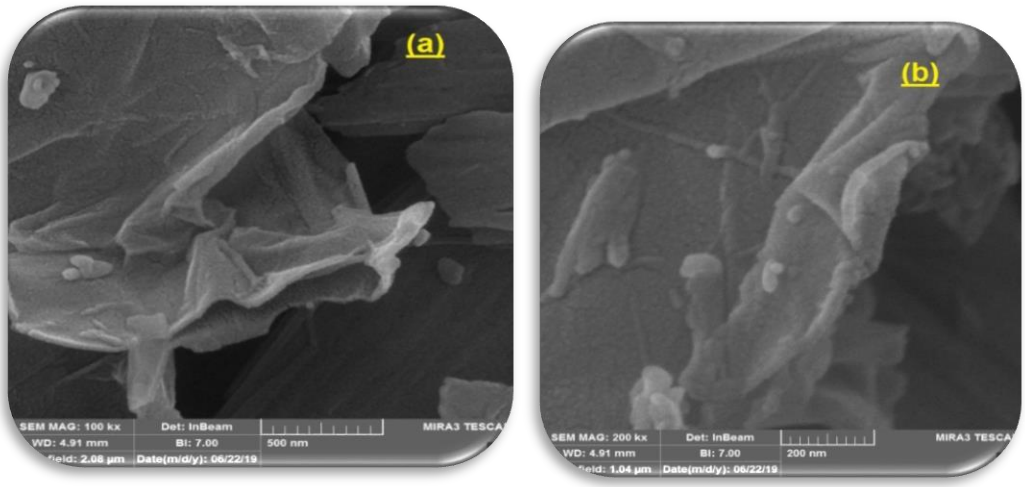


Figure (3-14) shows the SEM of the compound (A4)

The AFM images showed the spread of the superimposed materials on the surface of graphene oxide in an almost homogeneous manner (a), and the thickness of the sample was 1.9 nm. This indicates the nanosized, which supports the SEM and XRD images. It also indicates a decrease in the peak thickness of the sample (b).

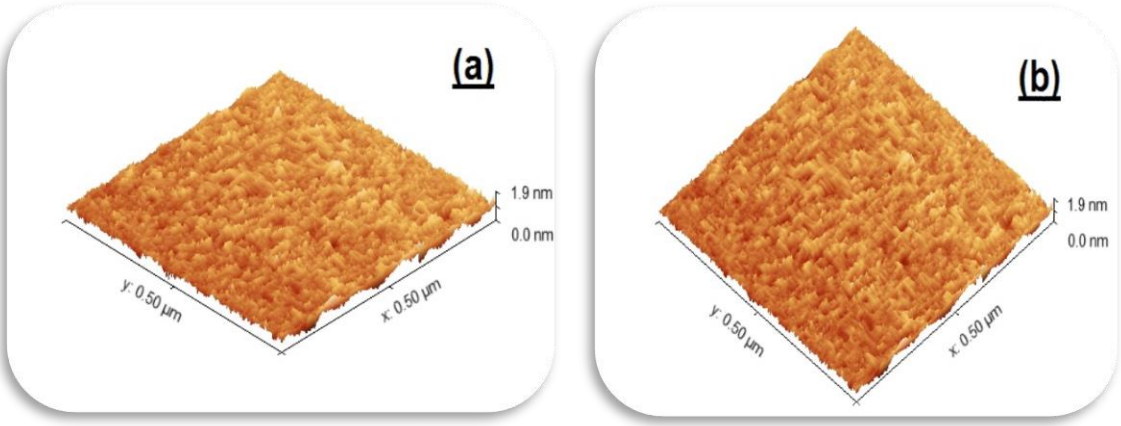


Figure (3-15) shows the AFM of the compound (A4)

The TEM images of compound (A4) clearly showed the presence of layers of material with cracks (a) and the supporting material is connected in a circular manner and far from the edges of the cracks (b).

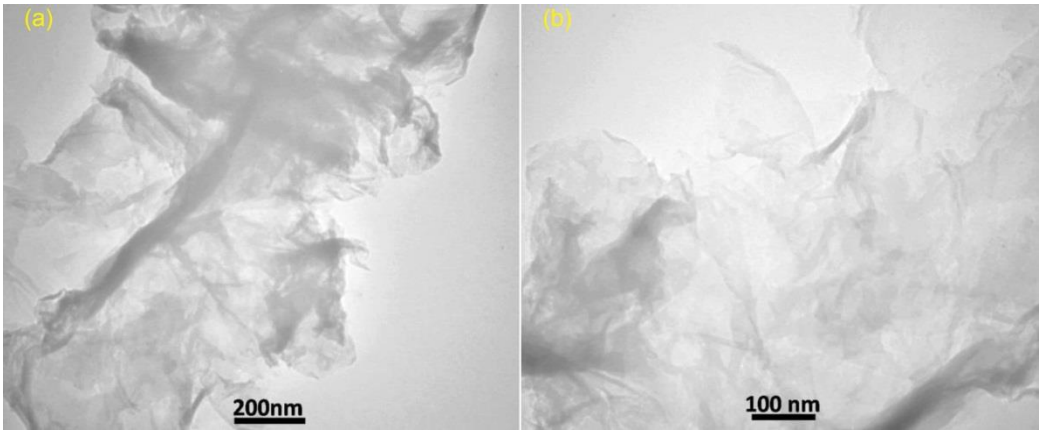


Figure (3-16) shows the TEM of the compound (A4)

3.5 Evaluation of the bacterial biological activity of some prepared compounds

Nanocomposites and heterocyclic compounds have different biological effectiveness against Gram-positive and Gram-negative bacteria. Two types of bacteria were used, and the effectiveness was evaluated by etching method, where three types of bacteria were used: Staphylococcus aureus, Bacillus bacteria, and Salmonella. These germs were chosen due to their medical importance. It causes many diseases, as well as its resistance to antibiotics. [24]

The results also indicate that the prepared compounds (A3) and (A4) have the ability to inhibit the growth of bacteria, both positive and negative, as the higher the concentration, the greater the inhibition. Concentrations of (50, 100, 150) p.p.m. were used, where the highest inhibition is when (150) p.p.m and the lowest at (50) p.p.m. The standard antibiotic used for comparison purposes in the effectiveness of inhibition is gentamycin.

When studying the inhibition using methylcellulose of gram-positive Staphylococcus aureus bacteria, it was found that the compound (A3) at a concentration of 150 p.p.m gave the highest inhibition of (A4) by 19.2 percent.

When studying the gram-positive bacillus bacteria, it was found that the compound (A4) gave the highest inhibition at a concentration of (150) p.p.m by 19.9, while when studying the gram-negative salmonella bacteria, it was found that the compound (A4) gave the highest inhibition at (150) p.p.m by 19.5. From this we conclude that the compound (A4) gave the highest rate of inhibition, and some concentrations showed a higher rate of inhibition than the antibiotic that was used in the study of gentamycin, and this is attributed to the large surface area that characterizes the compound (A4).

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