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Green Synthesis of Manganese Dioxide Nanoparticles and its Biological

Sabeeha K. Shanshool ***, *Zainab J. Shanan***

Department of Physics, College of science for Women, University of Baghdad, Baghdad, Iraq.
*Corresponding Author.

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Abstract

The goal of this research is to prepare Manganese dioxide (MnO_2) nanoparticles from plant extracts (*Puncia granatum L peel, Artemsiai herba-alba Asso, Matricaria chamomilla L, and Camellia sinensis*) and test their antibacterial activity. To evaluate the presence of phytochemicals in plant extracts, were used UV-Visible spectrophotometer analysis (UV– Vis), Zeta potential (ZP), and X-ray Diffraction (XRD). The production of MnO_2 nanoparticles by plant extracts was shown to have distinctive features. The average crystallite size of manufactured MnO_2 NPs was calculated to be between 14.22-21.0 nm, with zeta potential values -23.6, -20.04, -25.6, -16.7 mV, respectively. Manganese dioxide nanoparticles (MnO_2 NPs) have significant antibacterial activity against fungi and diverse gram-positive and gram-negative bacteria. The findings results showed that the synthesized MnO_2 NPs by *P granatum L peel*, *A herba-alba Asso*. plants had the highest percentage of bacterial and fungi inhibition, while *M. chamomilla L*.and *C. sinensis* extracts showed lower antifungal activity

Keywords: Antimicrobial, Fungi, Manganese dioxide (MnO_2) , *P granatum L peel*, Zeta potential (ZP).

Introduction

Nanoparticles are microscopic things that act in terms of transport and properties as if they were a single entity. Examples include liposomes, Nanocrystals, Nano suspension, Solid lipid nanoparticles (SLN), and other nanoparticles. As a new generation of ecologically benign catalysts, nano-MnO_2 has a lot of potential in the field of environmental protection. A lot of investigations have discovered that the structure of semiconductor materials influences their functional activity.[1] Manganese dioxide is one of the most intriguing materials since it has a diverse structure and large surface area. Manganese dioxides' diverse structural and chemical properties are used to benefit possible applications such as cation-exchange, ion and molecular separation; absorbents, sensors, batteries, and catalysis.[2] Microorganisms capable of generating nanoparticles include bacteria, fungi, and algae. The principle is the reduction of metal ions to nanoparticles.[3] Manganese dioxide NPs have got a lot of attention because of their wide range of applications in sectors like catalysis, chemical sensing devices, ion-sieves, microelectronics,[4] rechargeable batteries, and optoelectronics.[5] They are also based on earth's extra elements due to the presence of many attachments for a variety of activities. Mn is the twelfth most prevalent element on the planet and the third most common transition element after Fe and Ti.[6] For dye degradation, MnO_2 NPs show a strong photocatalytic activity.[7,8] Although physical and chemical methods can be used to synthesize nanoscale materials, green

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manufacturing is the best alternative because it is more environmentally friendly.[9,10] Plant extracts can be used to create nanoscales instead of conventional biological techniques such as the microbial approach, which includes the time-consuming process of maintaining cell cultures.[11,12] MnO_2 NPs were used to test antibacterial activity against four strains of bacteria (*B. Subtilus*, *S. aureus*, *E. coli*. and *K. pneumonia*) and one yeast (*C. albicans*). The aim of the research is to prepare the oxide in an environmentally friendly and inexpensive way, use of several plants in the preparation of manganese oxide nanoparticles Like (*P granatum L peel* ،*A herba-alba.Asso and* ،*M chamomilla L, C sinensis*), study the physical properties of the prepared material and measure the biological activity of the prepared material on several types of bacteria and fungi.

Materials and Methods

Green methods of synthesis Manganese dioxide nanoparticles

Collecting of plant material:

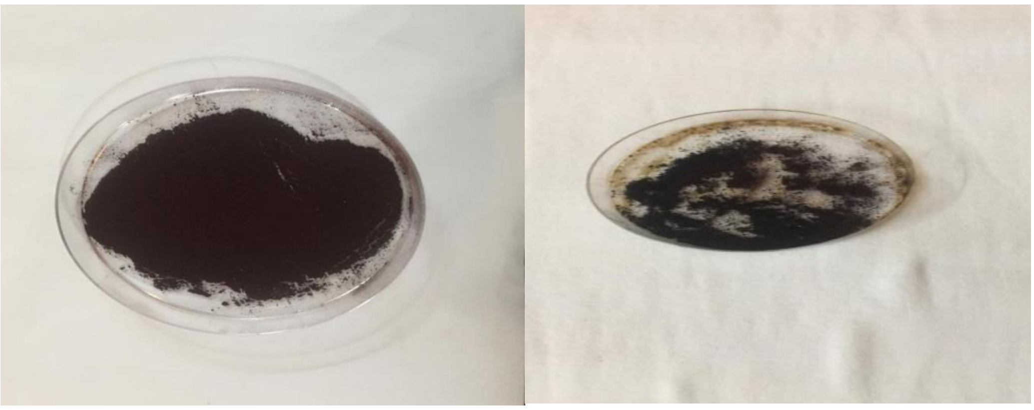
A herba-alba Asso. M chamomilla L, P granatum L peel and *C sinensis* were collected from the local market in Baghdad, the plants were kept at room temperature until use.

The preparation of hot aqueous extract of *P granatum L peel, A herba-alba Asso, M chamomilla L,* and *C sinensis*.

The leaf powder (10gm) was mixed well with 150 ml distilled water boiled at 80°C, and then homogenized on a magnetic stirrer for 2 hours, although the color of the aqueous solution was different. The aqueous solution was then1filtered and centrifuged for 20 minutes at 6000 rpm before being stored at 4°C until being used.

The synthesis of MnO_2 NPs by using aqueous extract of *P granatum L peel, A herba-alba Asso, M chamomilla L.* and *C sinensis*.

The initial solution was made by, 0.2 g of manganese acetate$(CH_3COOH)_2Mn.6H_2O$ was mixed with 100 ml of distilled water and heated for ten minutes with magnetic stirring. To prepare MnO_2 nanoparticles, 50 ml of the manganese acetate solution was mixed with 5 ml aqueous extracts from *P granatum L peel, A herba-alba Asso, M chamomilla L* and *C sinensis* extracts on a magnetic stirrer at 70°C for 60 minutes, despite the fact that the color of the aqueous solution varies. The hue of the solution changed to brown when manganese acetate was added to both *P granatum L peel, M chamomilla L* and *C sinensis* extracts, The hue of the solution changed to dark brown when it was mixed with extracts of *A. herba- alba.Asso.* The sample was then centrifuged at 8000 rpm for 15 minutes. The solution was baked at 250°C for 2 hours, and the powder was carefully collected and kept for future use see Fig 1.



(a) (b)

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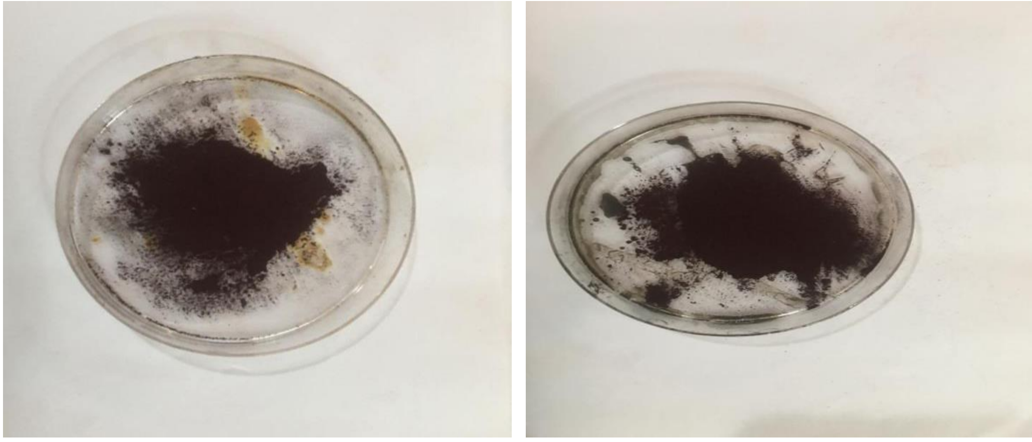

(c) (d)

Figure1. Manganese dioxide nanoparticles green synthesized via (a) *P granatum L peel,* (b) *A herba-alba Asso* extract, (c) *M chamomilla L* extract, (d) *C sinensis* extract.

Biological efficacy method

The antibacterial activity of manganese dioxide nanoparticles on Mueller-Hinton agar (MHA) was examined using the good diffusion method (Merck). Inhibition1zones against gram negative bacteria (E. coli, K. pneumonia), gram positive bacteria (*S aureus*, *B Subtilus*), and fungal isolates (*C albicans*) were measured in millimeters (well size was 6 mm).[13,14] MHA agar plates were1infected with bacterial strains under aseptic circumstances, and wells were filled with 50 MnO_2 NPs at several concentrations (100, 75, 50, 25%) and incubated at 37°C for 24 hours. The inhibition zones were measured after the incubation period. [15]

Characterization

UV–visible spectrometer(UV amersham bioscience libra s32 england), Powder XRD (XRD Siemens model D500) and Zeta potential (Dls zeta Malvern Zetasizer nanozs) were used to characterize the produced MnO_2 nanoparticles in order to explore their structural and optical properties.

Results and Discussion

UV – visible spectrum Studies

The UV-visible of green MnO_2 NPs was used to assess band gap and the absorption maxima of biological molecules involved in the reduction and capping from *P granatum L peel, A herba-alba Asso, M chamomilla L,* and *C sinensis* extracts. as shown in Table 1. The UV–visible study revealed that plant extracts have absorption maxima in the UV region 310–390 nm as shown in Fig 2, The energy gap is determined by Eq 1[16]:

$$Eg= hv/ \lambda \;\ldots\ldots 1$$

Where Eg: Energy gap, λ : wavelength, hv: Photon energy.+

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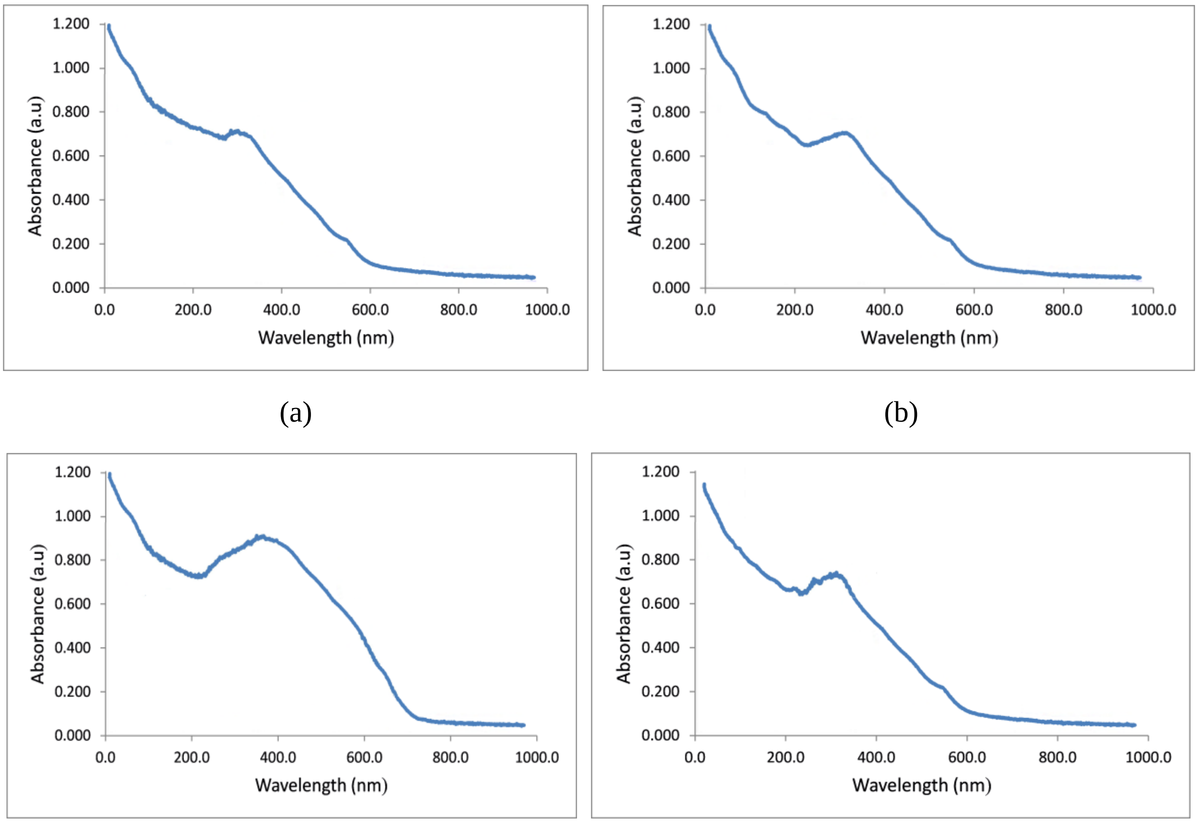

(a) (b) (c) (d)

Figure 2. UV-visible of green-synthesized of MnO_2 NPs using, (a) *P granatum L peel* extract, (b) *A herba-alba Asso* extract, (c) *M chamomilla L* extract, (d) *C sinensis* extract.

Table 1. Band gaps from UV-Vib absorption spectra of the MnO_2.

MnO_2 NPs isolates	Absorption Wavelength (nm)	Band gap (mV)
P granatum L peel	300	4.13
A herba-alba Asso	320	3.87
M chamomilla L	380	3.26
C sinensis	305	4.06

Powder XRD Studies

The manganese dioxide nanoparticles' crystalline size and nature were confirmed by X-ray diffraction analysis. Fig 3a, b shows an XRD pattern of green-synthesized MnO_2 NPs via *P granatum L peel, A herba-alba Asso.* XRD pattern demonstrates distinguish peaks at 2θ=28.51°, 30.51°, 35.20°, 43.17°, 50.72°, 56.41°, 68.32° and 75.71 indexed to (310), (101),(222),(301), (411),(110),(002) and (217) crystal plane of MnO_2 NPs (JSPDF 44-0141).[17,18] The obtained results are in good agreement with the JCPDS pdf number of 10799; [19] In addition, the XRD pattern shows that MnO_2 NPs are extremely crystalline, which is demonstrated by the intensity of the peaks and supported by a comparison of the acquired XRD spectrum with the reference material. Eq 2 the Debye Scherrer condition is used to calculate the crystallite measure[20].

$$D = 0.9\lambda/\beta \cos\theta \ldots 2$$

Where λ is the X-beam wave length, β is the line expanding at a large portion of the greatest power in radians, θ is the Bragg point. The average particle size was discovered to be 14.22 and 18.61 nm, respectively. The XRD pattern of green-synthesized MnO_2 NPs by *M. chamomilla L.* and *C. sinensis*

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was shown in Fig 3c,d. The XRD pattern results showed that various diffraction1peaks , which agree with JCPDS no. 78-0424 as indexing to (101),(301),(210),(411), (116),(220) and (300) crystal planes at crystal planes 30.28, 43.70, 44.67, 50.67, 56.27,64.73 and 72.35 degrees, respectively. The Miller index (101) and (301) had the highest peak intensity at a 30.28 and 43.70-degree angles. The enhanced peak intensity and crispness of the synthesized NPs further confirmed that they are extremely crystalline.[21] The average particle size was discovered to be 21.0 and 16.07 nm, respectively, as shown Table 1. The inexplicable reflections could be caused by the crystallization of bio-organic molecules on the surface of nanoparticles.[22]

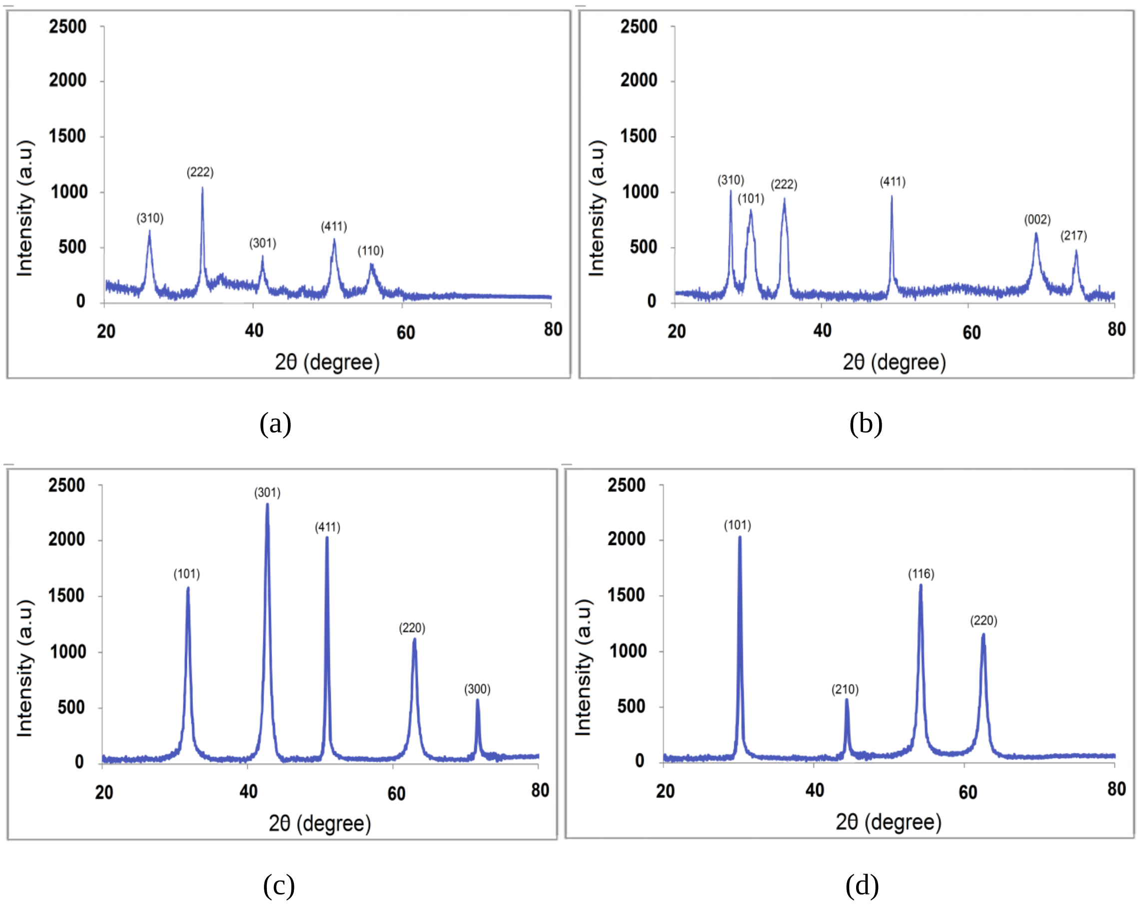

Figure 3. X-ray diffraction (XRD) pattern of MnO_2 NPs synthesized via (a) *P granatum L peel* extract, (b) *A herba-albaa Asso* extract, (c) *M chamomilla L* extract, (d) *C sinensis* extract.

Table 2. Crystalline size of MnO_2 NPs.

MnO_2 NPs isolates	Average grain size (nm)
P granatum L peel	14.22
A herba-alba Asso.	18.61
M chamomilla L	21.0
C sinensis	16.07

Zeta potential analysis

The stability of colloidal nanoparticles is demonstrated by ZP. at 500 mJ laser power and 500 pulses, the zeta potential values of green-synthesized MnO_2 NPs utilizing *P granatum L peel*, *A herba-alba Asso*, *M chamomilla L*, and *C sinensis* extracts were -23.6, -20.04, -25.6, and -16.7mV, respectively[23]. More negative values in the colloidal solution suggest that the NPs are more stable,[24] as shown in Fig. 4 .

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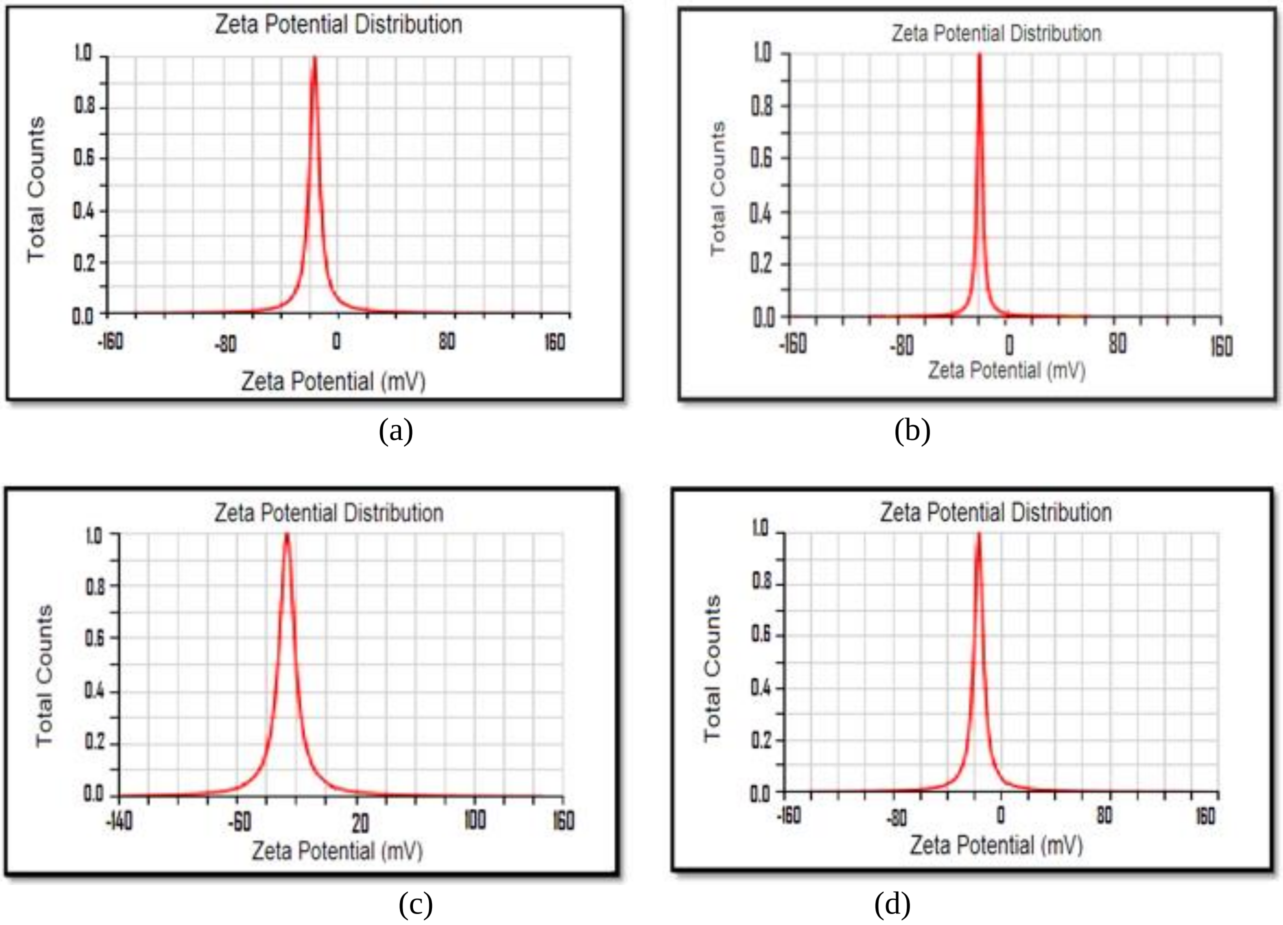

Figure 4. Zeta potential analysis of green-synthesized of MnO_2 NPs using (a) *P granatum L peel* extract (b)*A herba-alba Asso* extract (c) *M chamomilla L* extract (d) *C sinensis* extract

Antimicrobial activity

The results also showed that the producer of green synthesized MnO_2 NPs via *P granatum L peel*, *A herba-alba Asso*, *M chamomilla L* and *C sinensis* extracts. were more active toward Gram-negative species(*E. coli*, *K. pneumonia*) than Gram-positive species (*S aureus*, *B Subtilus*) Manjula *et al*, were also reported the same inhibitory effect of MnO_2 NPs toward Gram-positive than Gram-negative bacteria[25] . This could be due to structural and compositional differences in the cell1walls of Gram-negative and Gram-positive bacteria[26,27], and as shown in Fig 5. According to several studies, the majority of nanoparticles antibacterial activity is caused by physical and chemical degradation, such as the dissolution of lipid molecules and oxidative stress.[28] A synergistic interaction between the physical characteristics of the nanoparticle and the adsorption of biologically active phytomolecules from the *P granatum L peel*, *A herba-alba Asso*, *M chamomilla L*, and *C sinensis* extracts on their surface and other plant materials may also contribute to the enhanced antibacterial action of MnO_2 NPs[29-31] To demonstrate the findings the results showed that the synthesized MnO_2 NPs significantly reduced the killing efficiency of MnO_2 NPs using *P. granatum.L peel* and *A herba-alba Asso.* extracts of *C albicans*, as shown in Fig 6. MnO_2 NPs by *M. chamomilla L.* and *C sinensis* extracts showed lower antifungal activity than MnO_2 NPs by *P granatum L peel* and *A herba-alba Asso.* extracts, as shown in Table 3. This shows that physiologically active phytomolecules, particularly efficient at killing fungal strains, are present in the *P granatum L peel* and *A herba-alba Asso.* extract.[32]

The numbers 9, 10, 11 and 12 refer to Manganese dioxide nanoparticles prepared by the extracts (*P granatum L peel*, *A herba- alba Asso*, *M. chamomilla L* and *C sinensis*) respectively.

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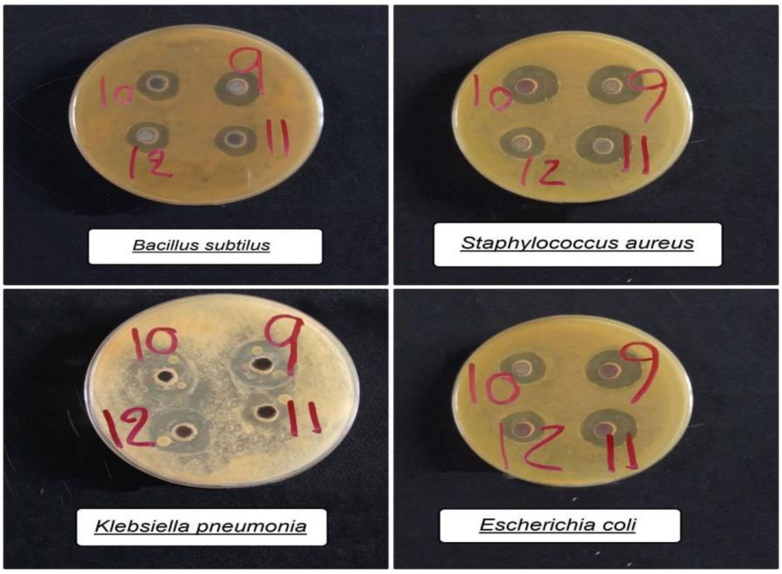

Figure 5. Representative images of control and treated for gram-positive and gram-negative bacteria

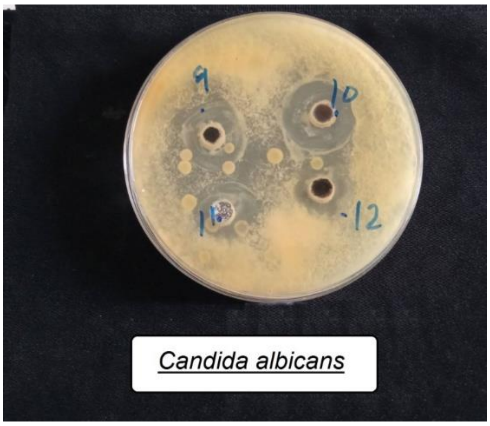

Figure 6. Representative images of control and treated for fungi (*C albicans*)

Table 3. Measurement of inhibition diameters of MnO_2 nanoparticles on bacteria and fungi

Samples	Zone of inhibition (mm)				
	S aureus	***B Subtilus***	***E coli***	***K pneumonia***	***C albicans***
9	23	15	16	25	26
10	23	17	24	24	25
11	15	16	23	23	23
12	20	15	17	16	17

Conclusion

In this research, we focused on the various plant species' capacity to green synthesis NPs. Over conventional physical and chemical methods, plant-mediated NP synthesis offers undeniably considerable advantages. Using a green synthesis method, MnO_2 nanoparticles were produced from the peels of *P granatum L*, *A herba-alba Asso*, *M chamomilla L*, and *C sinensis*. The creation of pure and crystalline MnO_2 nanoparticles was validated by XRD and UV-visible. Based on the results of the XRD, we deduced that the generated manganese dioxide nanoparticles were extremely crystalline in nature. The manganese dioxide nanoparticles ranged in size from 14.22 to 21.0 nm on average and they showed significant antibacterial action. The research results demonstrate Manganese dioxide nanoparticles from the green technique can inhibit the growth of *E coli, K pneumonia, S aureus, B subtilus*, and *C albicans,* where we conclude from the above results that manganese dioxide prepared by *A herba-alba Asso the* extract has the highest percentage of inhibition of bacteria and fungi.

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Authors' Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in University of Baghdad.

Authors' Contribution Statement

Z. J. Sh. proposed the topic of research and guidance and the review and proofreading the research. S. K. Sh. prepared the samples and analyzed parameters and writing, as well as the costs and publishing process.

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التوليف الأخضر لجسيمات ثاني أوكسيد المنغنيز النانوية وتقييمها البيولوجي

صبيحة خليل شنشول ، زينب جاسم شنان

قسم الفيزياء، كلية العلوم للبنات، جامعة بغداد، بغداد، العراق

الخلاصة

الهدف من هذا البحث هو تصنيع جسيمات نانوية لثاني أوكسيد المنغنيز من المستخلصات النباتية (*P granatum L peel*، *A herba-alba Asso* ، *M chamomilla L.*، *C sinensis*) واختبار نشاطها المضاد للبكتيريا. لتقييم وجود المواد الكيميائية النباتية في المستخلصات النباتية ،تم استخدام تحليل مقياس الطيف الضوئي المرئي بالأشعة فوق البنفسجية (UV-Vis) ، إمكانات زيتا (ZP) و حيود الأشعة السينية (XRD). تبين أن تحضير جزيئات MnO_2 النانوية عن طريق المستخلصات النباتية له سمات مميزة. تم حساب متوسط الحجم البلوري لمركبات MnO_2 NPs المصنعة بين 21.0_14.22 نانومتر ، مع قيم جهد زيتا -23.6 ، -20.04 ، -25.6 ، -16.7 ملي فولت على التوالي. الجسيمات النانوية لثاني أوكسيد المنغنيز (MnO_2 NPs) لها نشاط مضاد للجراثيم ضد الفطريات والبكتيريا المتنوعة الموجبة والسالبة لملون غرام. أظهرت النتائج أن MnO_2 NPs المحضر بواسطة *P granatum L peel* ، *A herba-alba Asso.* كان أعلى نسبة من تثبيط البكتيريا و الفطريات، بينما أظهرت مستخلصات *M chamomilla L* و *C sinensis* نشاطًا مضادًا اقل.

الكلمات المفتاحية: مضادات الميكروبات, الفطريات, ثاني أكسيد المنغنيز MnO_2 , قشر الرمان, جهد زيتا.