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Preparation of New PMMA/PEG/Si₃N₄ Nanocomposites for Biological Applications

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The PMMA–PEG–Si₃N₄ nanocomposites' films are fabricated by casting method with different ratios of Si₃N₄ nanoparticles (NPs) of 0, 1.6, 3.2, 4.8 and 6.4 wt.% to use for antibacterial defence and γ -radiation shielding. The PMMA–PEG–Si₃N₄ nanocomposites' films are tested for antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*. The results show that the PMMA–PEG–Si₃N₄ nanocomposites have high antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*. In addition, the PMMA–PEG–Si₃N₄ nanocomposites' films are tested for γ -radiation shielding. The results show that the PMMA–PEG–Si₃N₄ nanocomposites' films have high linear attenuation coefficients for γ -rays.

Плівки нанокompозитів поліметилметакрилат–поліетиленгліколь–Si₃N₄ (ПММА–ПЕГ–Si₃N₄) виготовляються методом лиття з різними співвідношеннями наночастинок Si₃N₄ у 0, 1,6, 3,2, 4,8 і 6,4 мас.% для використання для антибактеріального захисту та захисту від γ -випромінювання. Плівки нанокompозитів ПММА–ПЕГ–Si₃N₄ перевірено на антибактеріальну активність проти золотистого стафілокока та кишкової палички. Результати показують, що нанокompозити ПММА–ПЕГ–Si₃N₄ мають високу антибактеріальну активність проти золотистого стафілокока та кишкової палички. Крім того, плівки нанокompозитів ПММА–ПЕГ–Si₃N₄ проходять випробування на екранування γ -випромінювання. Одержані результати показують, що плівки нанокompозитів ПММА–ПЕГ–Si₃N₄ мають високі коефіцієнти лінійного згасання для γ -променів.

Key words: polymethyl methacrylate, polyethylene glycol, Si₃N₄, nanocomposites, antibacterial defence, radiation shielding.

Ключові слова: поліметилметакрилат, поліетиленгліколь, Si₃N₄, нанокompозити, антибактеріальний захист, радіаційний захист.

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1. INTRODUCTION

Nanocomposites are materials with at least one nanometer of dimension. It demonstrates high-level performance, and composite combinations have unusual properties and uniqueness in design possibilities. They are made up of two or more different composites with varying physical and chemical properties, and they usually have different phases separated by an interface [1].

PMMA (polymethyl methacrylate) is a transparent, rigid thermoplastic polymer that is widely used as a shatterproof substitute for glass. Because of the numerous technical advantages, which it has over other transparent polymers, it is frequently used as a lightweight or shatter-resistant alternative to glass in sheet form. Because of its moderate physical and mechanical properties, ease of processing, and low cost, PMMA is widely used. Non-modified PMMA is brittle, when subjected to a load, particularly an impact force, and is more prone to scratching than conventional inorganic glass, whereas, modified PMMA can achieve high scratch and influence resistance under certain conditions [2]. PMMA has been a popular choice for the preparation of polymer–inorganic composites, and it was chosen as a supporting matrix in this study because of its lightweight, smoothness, flexibility, environmental durability, high clarity, light transmission, milder processing conditions, and biocompatibility [3]. PMMA can withstand temperatures ranging from 70°C to 100°C. Furthermore, it has very good optical properties with a refractive index ranging between 1.3 and 1.7. PMMA is one of the best organic optical materials, and it is widely used as a substitute for inorganic glass due to its high impact strength, lightweight, and shatter resistance [4]. PMMA has numerous applications in manufacturing, and several technologies benefit from its unique combination of excellent optical properties with chemical inertness, good spectroscopic properties, thermal stability, electrical properties, and ease of forming and shaping [5].

Because polyethylene glycol (PEG) has important properties such as excellent water solubility, good protein adsorption resistance, and low toxicity, it is used in a variety of applications, including as a coating material for biotechnical applications, the optical band gaps, refractive indices, and optical dielectric constant imaginary part [6]. PEG is a type of polymer that is highly biocompatible and is widely used in contamination-resistant surfaces, medicine, and antibacterial activities [7].

Silicon nitride Si_3N_4 is a material with excellent mechanical and wear properties, as well as high thermal-shock resistance and ther-

mal conductivity. Silicon nitride-based composites have the potential to be used as an advanced structural material due to their excellent property combinations, such as high mechanical strength and resistance to wear and corrosion, as well as high chemical and thermal stability at normal and elevated temperatures [8]. Si₃N₄ is used in a variety of applications, including passive and active devices, as well as nonlinear optics. Broadband light sources from near-IR to mid-IR wavelengths have also been realized using silicon nitride [9]. Si₃N₄ chemical inertness also makes it useful as a shielding layer for magnetic thin films in disk drives [10].

There are numerous studies on the composites and nanocomposites applications like optical, electronics and optoelectronics applications [11–31], piezoelectric and sensors [32–42]. This work aims to preparation of PMMA–PEG–Si₃N₄ nanocomposites films to use for antibacterial and gamma-radiation shielding.

2. EXPERIMENTAL PART

The nanocomposites of PMMA–PEG blend as matrix and silicon-nitride (Si₃N₄) nanoparticles as additives were prepared by the casting method with concentration of polymer blend (PMMA 85%, and PEG 15%) were dissolved in 30 ml of chloroform and mixed with a magnetic stirrer. Different concentrations of silicon-nitride nanoparticles 0, 1.6, 3.2, 4.8 and 6.4 wt.% are added to the blend. The samples were examined by optical microscope. The antibacterial activity of the PMMA–PEG/Si₃N₄ nanocomposites is measured using the disc diffusion method. Gram-positive (*Staphylococcus aureus*) and gram-negative (*Escherichia coli*) organisms were used to perform antibacterial activities. The diameters of the inhibition zones in all samples of PMMA–PEG–Si₃N₄ nanocomposites were measured. The gamma-ray shielding application was investigated. A gamma-radiation source (Cs-137) was used, samples were placed at 3 cm in front of the radiation source, and attenuation factors were measured using a Geiger meter.

3. RESULTS AND DISCUSSION

The optical microscopic images of PMMA–PEG–Si₃N₄ nanocomposites are shown in Fig. 1. At lower concentrations, the Si₃N₄ nanoparticles aggregate as clusters. When the concentrations of nanoparticles are increased, the nanoparticles form a network of paths inside the polymer matrix [43–46].

Figure 2 depicts the antibacterial properties of the PMMA–PEG–Si₃N₄ nanocomposites against gram-positive bacteria (*Staphylococ-*

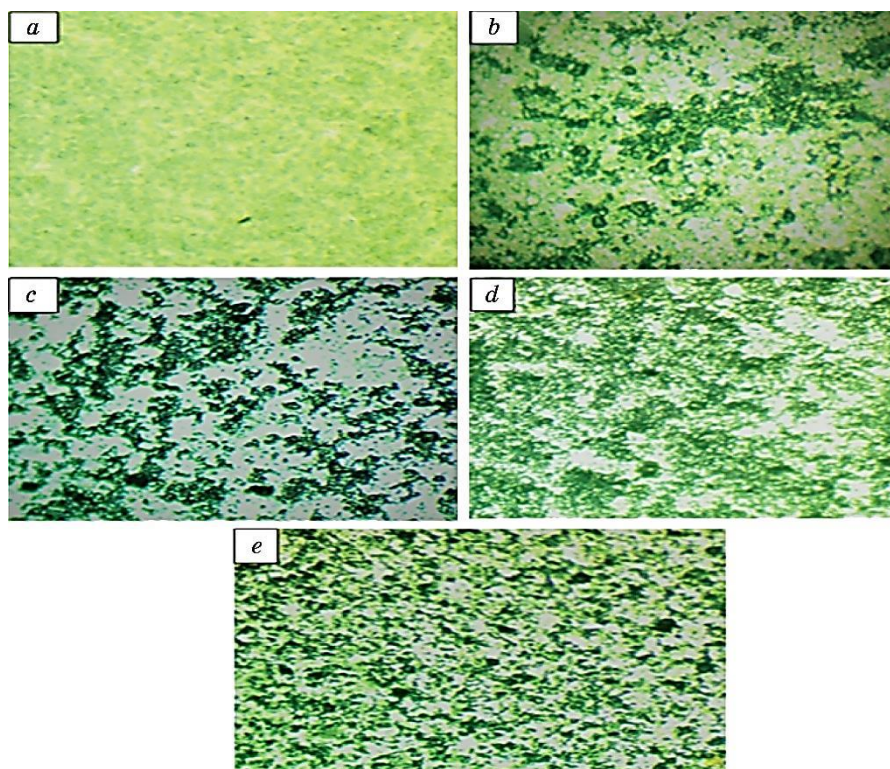


Fig. 1. Microscopic images of PMMA-PEG/Si₃N₄ nanocomposites: (a) for pure PMMA-PEG, (b) 1.6 wt.% Si₃N₄ NPs, (c) 3.2 wt.% Si₃N₄ NPs, (d) 4.8 wt.% Si₃N₄ NPs, (e) 6.4 wt.% Si₃N₄ NPs.

cus aureus). Figure 3 depicts the antibacterial properties of the PMMA-PEG-Si₃N₄ nanocomposites against gram-negative bacteria (*Escherichia coli*). As shown in these figures, the diameter of the inhibition zone increases as the concentration of Si₃N₄ nanoparticles increases.

The presence of reactive oxygen species (ROS) generated by various nanoparticles may be the cause of nanocomposites' antibacterial activity. The chemical interaction between hydrogen peroxide and membrane proteins, or the chemicals produced in the presence of nanocomposites, and the outer bilayer of bacteria, could be the cause of nanocomposites' antibacterial activity.

The hydrogen peroxide produced enters bacterial cell membranes and kills them. Once the hydrogen peroxide is produced, the nanocomposites continue to interact with dead bacteria, preventing further bacterial action and continuing to produce and release hydrogen peroxide into the medium. The nanoparticles in nanocomposites

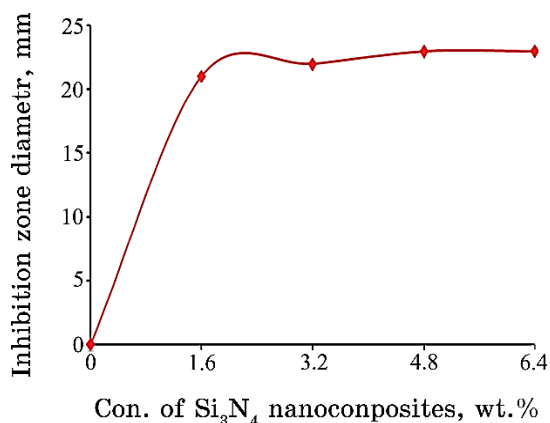


Fig. 2. Antibacterial properties of the PMMA–PEG/Si₃N₄ nanocomposites against gram-positive *Staphylococcus aureus*.

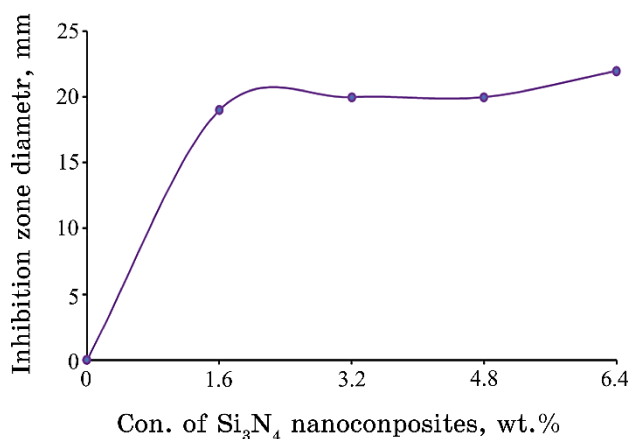


Fig. 3. Antibacterial properties of the PMMA–PEG/Si₃N₄ nanocomposites against gram-negative *Escherichia coli*.

may be negatively charged, creating an electromagnetic attraction between the nanoparticles and the microbes. When the attraction is formed, the microbes oxidize and die. The main mechanism causing the antibacterial properties of nanocomposites caused by nanoparticles could be oxidative stress caused by ROS. ROS includes radicals such as superoxide radicals (O₂⁻), hydroxyl radicals (•OH), and hydrogen peroxide (H₂O₂); and singlet oxygen (1O₂) may be the cause of protein and DNA damage in bacteria. The current metal oxide could have produced ROS, resulting in the inhibition of most pathogenic bacteria [47–51].

Figure 4 shows the N/N_0 variation with Si₃N₄ nanoparticles in

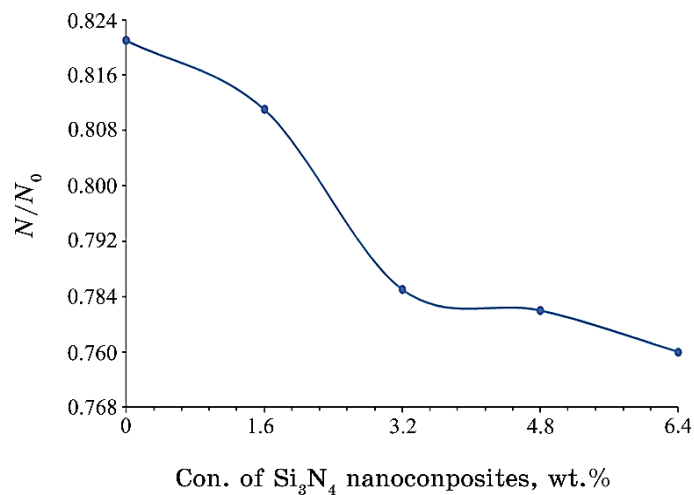


Fig. 4. N/N_0 variation for the PMMA-PEG- Si_3N_4 nanocomposites with different concentrations of Si_3N_4 nanoparticles.

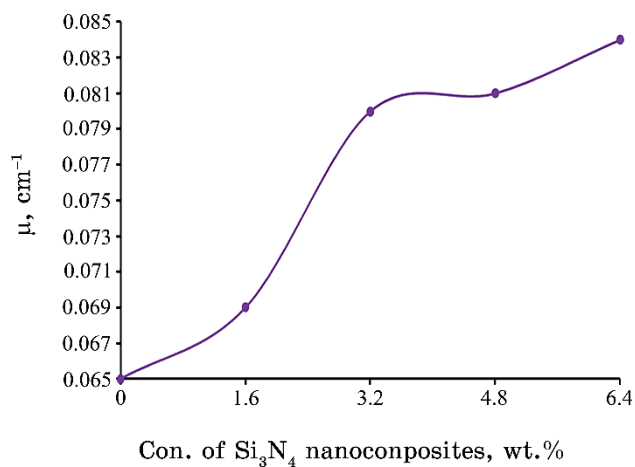


Fig. 5. Variation of gamma attenuation coefficient of PMMA-PEG- Si_3N_4 nanocomposites with concentration of Si_3N_4 nanoparticles.

various concentrations. The transmission radiation decreases as the attenuation of radioactive radiation increases as the Si_3N_4 nanoparticles increase.

Figure 5 depicts the gamma-radiation attenuation coefficients for the PMMA-PEG- Si_3N_4 nanocomposites. As the number of nanoparticles increases, the attenuation coefficient increases. This behaviour is because the gamma-radiation is absorbed or reflected by the nanocomposite shielding material [52-57].

4. CONCLUSIONS

In this study, the PMMA-PEG-Si₃N₄ nanocomposites' films were fabricated to employ for antibacterial defence and gamma-radiation shielding. The PMMA-PEG-Si₃N₄ nanocomposites' films were tested for antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*. The results showed that the PMMA-PEG-Si₃N₄ nanocomposites have high antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*. In addition, the PMMA-PEG-Si₃N₄ nanocomposites' films were tested for gamma-radiation shielding. The results showed that the PMMA-PEG-Si₃N₄ nanocomposites' films have high linear attenuation coefficients for gamma-ray.

REFERENCES

1. M. M. Shameem, S. M. Sasikanth, R. Annamalai, and R. G. Raman, *Materials Today: Proceedings*, **45**: 2536 (2021); doi:10.1016/j.matpr.2020.11.254
2. H. M. Alhusaiki-Alghamdi, *Results Phys.*, **24**: 104125 (2021); doi:10.1016/j.rinp.2021.104125
3. J. Bagawade, *Int. J. Curr. Sci. Res. Rev.*, **4**, No. 4: 246 (2021); doi:10.47191/ijcsrr/V4-i4-01
4. A. M. Alsaad, A. A. Ahmad, I. A. Qattan, A. R. El-Ali, S. A. Al Fawares, and Q. M. Al-Bataineh, *Polymers (Basel)*, **13**: 1 (2021); doi:10.3390/polym13111715
5. N. S. Alghunaim, *Results Phys.*, **5**: 331 (2015); doi:10.1016/j.rinp.2015.11.003
6. Z. K. Heiba, M. Bakr Mohamed, and S. I. Ahmed, *Alexandria Eng. J.*, **61**, No. 5: 3375 (2022); doi:10.1016/j.aej.2021.08.051
7. D. Tekin, D. Birhan, and H. Kiziltas, *Mater. Chem. Phys.*, **251**: 1 (2020); doi:10.1016/j.matchemphys.2020.123067
8. R. K. Arya and A. Telang, *Int. J. Eng. Adv. Technol.*, **9**, No. 3: 3366 (2020); doi:10.35940/ijeat.C6032.029320
9. T. Sharma, J. Wang, B. K. Kaushik, Z. Cheng, R. Kumar, Z. Wei, and X. Li, *IEEE Access*, **8**: 195436 (2020); doi:10.1109/ACCESS.2020.3032186
10. V. M. Bermudez and F. K. Perkins, *Appl. Surf. Sci.*, **235**, No. 4: 406 (2004); doi:10.1016/j.apsusc.2004.02.065
11. A. Hazim, A. Hashim, and H. M. Abduljalil, *Trans. Electr. Electron. Mater.*, **21**: 48 (2019); https://doi.org/10.1007/s42341-019-00148-0.
12. H. Ahmed, A. Hashim, and H. M. Abduljalil, *Ukr. J. Phys.*, **65**, No. 6: 533 (2020); https://doi.org/10.15407/ujpe65.6.533
13. H. Ahmed, A. Hashim, and H. M. Abduljalil, *Egypt. J. Chem.*, **62**, No. 9: 1659 (2019); doi:10.21608/EJCHEM.2019.7154.1590
14. H. Ahmed and A. Hashim, *Silicon*, **14**: 6637 (2022); https://doi.org/10.1007/s12633-021-01449-x
15. A. Hashim and Z. S. Hamad, *Egypt. J. Chem.*, **63**, No. 2: 461 (2020); doi:10.21608/EJCHEM.2019.7264.1593
16. H. Ahmed and A. Hashim, *Silicon*, **13**: 4331 (2020);

- <https://doi.org/10.1007/s12633-020-00723-8>
17. H. Ahmed and A. Hashim, *Transactions on Electrical and Electronic Materials*, **22**: 335 (2021); <https://doi.org/10.1007/s42341-020-00244-6>
 18. A. Hazim, A. Hashim, and H. M. Abduljalil, *Nanosistemi, Nanomateriali, Nanotehnologii*, **18**, Iss. 4: 983 (2020); <https://doi.org/10.15407/nnn.18.04.983>
 19. A. Hashim and Z. S. Hamad, *Nanosistemi, Nanomateriali, Nanotehnologii*, **18**, No. 4: 969 (2020); <https://doi.org/10.15407/nnn.18.04.969>
 20. A. Hashim, A. J. Kadham, A. Hadi, and M. A. Habeeb, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 2: 327 (2021); <https://doi.org/10.15407/nnn.19.02.327>
 21. A. J. K. Algidsawi, A. Hashim, A. Hadi, and M. A. Habeeb, *Semiconductor Physics, Quantum Electronics & Optoelectronics*, **24**, No. 4: 472 (2021); <https://doi.org/10.15407/spqeo24.04.472>
 22. A. Hazim, A. Hashim, and H. M. Abduljalil, *Egypt. J. Chem.*, **64**, No. 1: 359 (2021); [doi:10.21608/EJCHEM.2019.18513.2144](https://doi.org/10.21608/EJCHEM.2019.18513.2144)
 23. H. Ahmed and A. Hashim, *Silicon*, **13**: 2639(2020); <https://doi.org/10.1007/s12633-020-00620-0>
 24. A. Hazim, H. M. Abduljalil, and A. Hashim, *Transactions on Electrical and Electronic Materials*, **22**: 185 (2021); <https://doi.org/10.1007/s42341-020-00224-w>
 25. A. Hazim, H. M. Abduljalil, and A. Hashim, *Transactions on Electrical and Electronic Materials*, **21**: 550 (2020); <https://doi.org/10.1007/s42341-020-00210-2>
 26. H. Ahmed and A. Hashim, *Journal of Molecular Modeling*, **26**: 1 (2020); [doi:10.1007/s00894-020-04479-1](https://doi.org/10.1007/s00894-020-04479-1)
 27. H. Ahmed and A. Hashim, *Silicon*, **14**: 4907 (2022); <https://doi.org/10.1007/s12633-021-01258-2>
 28. H. Ahmed and A. Hashim, *Silicon*, **14**: 4079 (2021); <https://doi.org/10.1007/s12633-021-01186-1>
 29. H. Ahmed and A. Hashim, *Trans. Electr. Electron. Mater.*, **23**: 237 (2022); <https://doi.org/10.1007/s42341-021-00340-1>
 30. H. Ahmed and A. Hashim, *Silicon*, **14**: 7025 (2021); <https://doi.org/10.1007/s12633-021-01465-x>
 31. H. Ahmed and A. Hashim, *Silicon*, **13**: 1509 (2020); <https://doi.org/10.1007/s12633-020-00543-w>
 32. A. Hashim and A. Jassim, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 4: 883 (2021); <https://doi.org/10.15407/nnn.19.04.883>
 33. A. Hashim and Q. Hadi, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 4: 873 (2021); <https://doi.org/10.15407/nnn.19.04.873>
 34. A. G. Hadi, Z. Al-Ramadhan, and A. Hashim, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 4: 865 (2021); <https://doi.org/10.15407/nnn.19.04.865>
 35. D. Hassan and A. Hashim, *Bulletin of Electrical Engineering and Informatics*, **7**, No. 4: 547 (2018); [doi:10.11591/eei.v7i4.969](https://doi.org/10.11591/eei.v7i4.969)
 36. D. Hassan and A. H. Ah-Yasari, *Bulletin of Electrical Engineering and Informatics*, **8**, No. 1: 52 (2019); [doi:10.11591/eei.v8i1.1019](https://doi.org/10.11591/eei.v8i1.1019)
 37. H. Ahmed and A. Hashim, *International Journal of Scientific & Technology Research*, **8**, Iss. 11: 1014 (2019).

38. A. Hashim, A. J. K. Algidsawi, H. Ahmed, A. Hadi, and M. A. Habeeb, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 2: 353 (2021); <https://doi.org/10.15407/nnn.19.02.353>
39. A. Hashim, A. J. K. Algidsawi, H. Ahmed, A. Hadi, and M. A. Habeeb, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 1: 91 (2021); <https://doi.org/10.15407/nnn.19.01.091>
40. B. Mohammed, H. Ahmed, and A. Hashim, *Nanosistemi, Nanomateriali, Nanotehnologii*, **20**, No. 1: 187 (2022); <https://doi.org/10.15407/nnn.20.01.187>
41. A. Hashim and A. Jassim, *Nanosistemi, Nanomateriali, Nanotehnologii*, **20**, No. 1: 177 (2022); <https://doi.org/10.15407/nnn.20.01.177>
42. A. Hashim and Z. S. Hamad, *Nanosistemi, Nanomateriali, Nanotehnologii*, **19**, No. 4: 893 (2021); <https://doi.org/10.15407/nnn.19.04.893>
43. A. Hashim, *J. Mater. Sci.: Mater. Electron.*, **32**: 2796 (2021); <https://doi.org/10.1007/s10854-020-05032-9>
44. A. Hashim, *Journal of Inorganic and Organometallic Polymers and Materials*, **31**: 2483 (2021); <https://doi.org/10.1007/s10904-020-01846-6>
45. N. Al-Huda Al-Aaraji, A. Hashim, A. Hadi, and H. M. Abduljalil, *Silicon*, **14**: 4699 (2022); <https://doi.org/10.1007/s12633-021-01265-3>
46. A. Hashim, *Opt. Quant. Electron.*, **53**: 1 (2021); <https://doi.org/10.1007/s11082-021-03100-w>
47. A. Hashim, I. R. Agool, and K. J. Kadhim, *Journal of Bionanoscience*, **12**, No. 5: 608 (2018); doi:10.1166/jbns.2018.1580
48. H. B. Hassan, A. Hashim, and H. M. Abduljalil, *Nanosistemi, Nanomateriali, Nanotehnologii*, **20**, No. 3: 821 (2022); <https://doi.org/10.15407/nnn.20.03.821>
49. H. B. Hassan, A. Hashim, and H. M. Abduljalil, *Nanosistemi, Nanomateriali, Nanotehnologii*, **20**, No. 3: 815 (2022); <https://doi.org/10.15407/nnn.20.03.815>
50. A. Hazim, A. Hashim, and H. M. Abduljalil, *International Journal of Emerging Trends in Engineering Research*, **7**, No. 8: 68 (2019); <https://doi.org/10.30534/ijeter/2019/01782019>
51. A. Hazim, H. M. Abduljalil, and A. Hashim, *International Journal of Emerging Trends in Engineering Research*, **7**, No. 8: 104 (2019); <https://doi.org/10.30534/ijeter/2019/04782019>
52. A. Hashim and Z. S. Hamad, *Journal of Bionanoscience*, **12**, No. 4: 504 (2018); doi:10.1166/jbns.2018.1561
53. B. Abbas and A. Hashim, *International Journal of Emerging Trends in Engineering Research*, **7**, No. 8: 131 (2019); <https://doi.org/10.30534/ijeter/2019/06782019>
54. K. H. H. Al-Attiyah, A. Hashim, and S. F. Obaid, *Journal of Bionanoscience*, **12**: 200 (2018); doi:10.1166/jbns.2018.1526
55. D. Hassan and A. Hashim, *Journal of Bionanoscience*, **12**, No. 3: 341 (2018); doi:10.1166/jbns.2018.1533
56. A. Hashim and Z. S. Hamad, *Journal of Bionanoscience*, **12**, No. 4: 488 (2018); doi:10.1166/jbns.2018.1551
57. D. Hassan and A. Hashim, *Journal of Bionanoscience*, **12**, No. 3: 364 (2018); doi:10.1166/jbns.2018.1537