

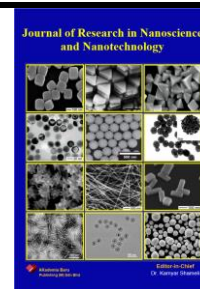


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Copper/Graphene Based Materials Nanocomposites and Their Antibacterial Study: A Mini Review

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ABSTRACT

Due to their biocidal activity properties, graphene based materials have been widely studied especially in biomedical, agriculture and water treatment process which focus on mitigating the microbial resistance problem. However, the antibacterial performance of these materials alone are relatively weak and need to be improved in order to enhance their biological activity. Copper nanoparticles is a low cost metal also has the antibacterial properties which is almost similar with the silver and gold nanoparticles. The combination of these two materials had produced to a new potential material as another alternative for the antibacterial agents. Therefore, in this work, a brief review of copper/graphene based material nanocomposites and their antibacterial study was discussed.

Keywords: Copper nanoparticles, graphene based materials, nanocomposites, antibacterial

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1. Introduction

The emergence of nanotechnology nowadays has open a new pathway for the researchers to find an alternative to solve the problems arise in most of the industries around the world such as agriculture, water treatment and medical industries. One of the common problems faced by these kind of industries is the growth of microbial resistance species that could harm the human health and also ecological system [1-4]. Due to that, nanomaterials were introduced in order to mitigate this problem as these materials have unique properties to be explored. Since then, an extensive amount of studies had been done during the last decade and it also has been widely expand to be utilized into multidiscipline applications [5-7]. Nanocomposite is one of the classifications in nanomaterials where

it could be defined as a materials that have at least one of them in a dimension of less than 100 nm scale [8]. A few nanocomposites that commonly explored are polymer nanocomposites [9], graphene based nanocomposites [10] and metal-metal oxide nanocomposites [11]. Usually nanoparticles that have size between 1 to 100 nm such as metal nanoparticles and metal oxide nanoparticles are used to produce the nanocomposite as it could enhance the application activities in the form of nanocomposite. In this paper, a short review on the production of copper/graphene based material nanocomposites is discussed including their antibacterial activity study.

2. Graphene Based Metal/Metal Oxide Nanocomposites

An extraordinary characteristic of graphene based materials related to their unique structure have attract researchers to investigate on their potential features for diverse applications. Generally, graphene based materials could be divided into three most common types which are known as graphene, graphene oxide (GO) and reduce graphene oxide (RGO). Graphene is known as an excellent support material because of its high surface area ($\sim 2600 \text{ m}^2\text{g}^{-1}$), high thermal/electrical conductivity, good mechanical strength and flexibility [12, 13]. Besides, due to the thick layer of carbon atoms bonded together by sp^2 hybridization which arrange in a hexagonal array structure of graphene, it lead to its other special properties such as high density and very high hydrophobicity. The difference structure of GO and RGO with graphene is the presence of oxygen functional groups attach to the layer of the sheet. Hence, that make them differ with the graphene as well as their properties which also change because of their structure. GO consists of an exogenous oxygen – containing group on its surface while RGO has less oxygen functional groups attach compare to GO.

GO could be produced by the oxidation of graphite through several methods such as Staudenmaier's method, Brodie's method and Hummer's method [14]. Hummer's method and its modification have been used extensively nowadays to produce GO as it is less hazardous route and could produce high level of oxidation [15]. Chemical exfoliation of graphite to graphite oxide was pioneered by William S. Hummers and Richard E. Offeman in the year 1958 [16]. Shahriary and Athawale studied on the production of GO by using Hummer's method and they found that this method was an efficient method to produce the GO [17]. By the present of the various oxygenated functional groups on the GO layer such as hydroxyl, carbonyl and epoxy, it leads for a better dispersion of metal nanoparticles as they could act as nucleation sites for the nanoparticles. RGO normally obtained through the reduction of GO and it could be done through the chemical reduction [18], green synthesis [19], thermal [14] and photochemical method [20]. Usually, the negative charge of functional groups such as $-\text{COOH}$, $-\text{OH}$ and $\text{C}=\text{O}$ of the GO were used as the precursor to produce the RGO. It leads to an easy dispersion of the material in aqueous solution and enable for this material to also acts as an effective anchor sites for nanoparticles immobilization [21].

GO and RGO are widely used in metal/metal oxide nanocomposites since these materials consist the oxygen-functional groups that could interact with the metal ions for the nucleation and growth to produce nanocomposites. Yin *et al.*, reported that several routes to synthesis GO loaded with the nanoparticles were through chemical reduction process, hydrothermal, electrochemical and ex-situ (loading the premade nanoparticles to the graphene surface) [22]. Jayabalan *et al.*, produced nickel oxide/reduce graphene oxide (NiO/RGO) and cobalt oxide/reduce graphene oxide ($\text{Co}_3\text{O}_4/\text{RGO}$) as catalyst in microbial electrolysis cell through ex-situ preparation. The nanocomposites were characterized using XRD, Raman spectra, SEM and EDX analysis. They found that the performance of NiO/RGO and $\text{Co}_3\text{O}_4/\text{RGO}$ gave a better result as catalyst when combining with the RGO [23]. Pang *et al.*, studied on producing zinc oxide/reduce graphene oxide (ZnO/RGO) nanocomposites for photo catalysis and they mentioned that ZnO alone had a weak activity as catalyst due to low surface

area. RGO was introduced to enhance the catalytic properties as it could minimize the recombination rate and produced high surface area with less aggregation for the photocatalytic study. Simplified impregnation method was used and the particle size ranging from 50 to 100 nm where the RGO acted as template to control the size of nanoparticles [24].

3. Copper/Graphene Based Materials Nanocomposites

Copper is a transition metal which has several oxidation states that make it special in term of its properties depending on its state. The surface plasmon resonance (SPR) of copper nanoparticles (Cu-NPs) also could help in improving the optical materials and devices due to its non-linear optical properties. It also has a high surface area to volume and low cost production compared to the noble metal such silver and gold [13]. Cu-NPs also known as a potential antibacterial agent as it has been used for centuries to disinfect microbial. However, the difficulties that need to be faced in producing Cu-NPs are (i) stabilization of particles against agglomeration, (ii) the achievement of monodisperse size distribution and (iii) easily oxidized when in contact with oxygen which could affect their potential application activity [25]. Cu-NPs is favourable in widespread of implementation including catalyst [26], sensor [27], anticancer [28] and antibacterial study [29].

In order to produce copper/graphene based material nanocomposites, the interaction of copper ions and graphene based materials occur through physisorption, cation- π interaction and electrostatic force interaction [30, 31]. Rios *et al.*, produced copper/reduce graphene oxide nanocomposites (Cu/RGO-NCs) as catalyst for ammonium perchlorate decomposition by in situ reduction and they also studied the interaction between the materials using *ab initio* calculation to reveal the important role of the functional groups in stabilizing metal nanoparticles [32]. Fahiminia *et al.*, reported a green synthesis of Cu/RGO-NCs using *Euphorbia cheiradenia* Boiss extract and it was used for reduction of organic dyes and 4-nitrophenol. The presence of phenolic compounds in the plant extract such as quercetin, kaemferol and rutin with sugar moieties acts as bio-reducer for the nanostructure formation. The size of the Cu-NPs synthesized was in the average size of 30 nm and spherical shape dispersed on the reduce graphene oxide sheet layer [12]. Some previous works of copper/graphene based material nanocomposites and their applications are shown as in Table 1.

Table 1. Some studies on copper/graphene based material nanocomposites and their applications

Copper/graphene based nanocomposites	Methodology	Size and Structure/ Shape	Application	Reference
Copper oxide-graphene nanocomposites	Wet chemical method	Less than 10 nm; spherical shape	Photocatalytic application	[33]
Copper oxide/graphene oxide nanocomposites	Ex-situ method	Less than 100 nm; cluster structure	Anticancer and catalytic application	[34]
Copper oxide/graphene oxide nanocomposites	Hydrothermal method	~ 10 nm; not stated	Catalytic application	[35]
Copper-functionalize/reduce graphene oxide nanocomposites	Chemical reduction	< 100 nm; not stated	Sensor application study	[36]

Copper/reduced graphene oxide nanocomposites	i. Ball milling ii. Electroless deposition method	Size not stated; porous structure	Tribological properties study	[37]
Copper /reduce graphene oxide nanocomposites	i. cyclic voltammetry ii. amperometry technique	< 100 nm; irregular shape	Sensor application study	[38]
Copper oxide/graphene oxide	Hydrothermal method	26 nm; irregular shape	Antibacterial and anticancer study	[39]
Copper/graphene oxide nanocomposites	In situ reduction method	~30 nm; not stated	Bone regeneration study	[40]

4. Antibacterial Applications of Copper/Graphene Based Nanocomposites

The growth of microbial resistance because of the bacteria that withstand to multiple antibiotics have been remain as a part of source for the morbidity, mortality and socioeconomic loss worldwide [41]. Due to the excellent antibacterial activity of copper, the US Environmental Protection Agency had registered about 300 copper-containing products as antibacterial agent back in 2008 and they were studied for biomedical application [42], crop disease control [43] and waste water treatment [44]. Cu-NPs has been a cheap and efficient antibacterial agent for centuries where it was discovered to exhibit broad antibacterial properties by oxidizing their proteins and lipids [45]. GO and RGO are toxic to both gram positive and gram negative bacteria [46]. However, the weak ability of the graphene based material to act as antibacterial agent leads to the aim in enhancing the antibacterial activity by hybridizing it with the Cu-NPs. Lately, green synthesis of graphene based materials attached with metal nanoparticles are favoured to utilize as biological agent since it is biocompatible and less hazardous [47]. Several factors are important for the nanocomposite to be used as biological agent such as size, shape, surface functionalization, stability and size distribution [48].

Table 2. A few previous literatures of the copper/graphene based materials nanocomposites for the antibacterial application.

Nanomaterials	Methodology	Type of Bacteria	Size and Structure/ Shape	Reference
Reduce graphene oxide/copper (I) oxide nanocomposites	Chemical reduction (toluene)	<i>P. aeruginosa</i> ¹ , <i>B. subtilis</i> ² and <i>E. coli</i> ³	70-90 nm; cubic structure	[51]
Reduce graphene oxide/copper (I) oxide nanocomposites	Thermal chemical vapour deposition method	<i>E. coli</i> ³	5-40 nm; crystal structure	[52]

Reduce graphene oxide/copper nanocomposites	In situ chemical reduction	<i>E. coli</i> ³	15–50 nm; cluster structure	[53]
Graphene oxide/copper oxide nanocomposites	Chemical reduction	<i>S. aureus</i> ⁴ and <i>E. coli</i> ³	3-10 nm; spherical shape	[54]
Graphene oxide/copper oxide nanocomposites	Thermal chemical reaction	<i>E. coli</i> ³ and <i>S. typhimurium</i> ⁵	190 ± 2.84 nm; not stated	[55]
Graphene oxide/copper oxide nanocomposites	Chemical reduction	<i>Pseudomonas syringae</i> pathovar tomato	21.28 nm; cluster structure	[56]

¹*P. aeruginosa*: *Pseudomonas aeruginosa*; ²*B. subtilis*: *Bacillus subtilis*; ³*E. coli*: *Escherichia coli*; ⁴*S. aureus*: *Staphylococcus aureus*; ⁵*S. typhimurium*: *Salmonella typhimurium*

Tu *et al.*, reported that antibacterial activity for copper/reduce graphene oxide (Cu/RGO-NCs) exhibit better antibacterial activity compared to the Cu-NPs alone [30]. Normally, the possible mechanism for copper/graphene based material nanocomposites is by the interaction of the positive charge of copper ions on RGO with the negatively charge surface of both gram positive and gram negative bacterial cells. The negative charge of gram positive bacteria is due to the presence of the teichoic acid with high amount of phospholipid groups that extend from the cell wall of the bacterial strain [49]. However, according to the Alayande *et al.*, the antibacterial activity of the nanocomposite was not related with the copper ion released nor the generation of reactive oxygen species but due to the electron transfer mechanism [50]. Table 2 shows a few previous literatures related to the copper/graphene based materials nanocomposites for the antibacterial application.

5. Conclusions

In conclusion, the usage of graphene based materials as supporting materials for metal nanoparticles including copper could lead to the enhancement of the nanomaterial properties and activities. Hence, copper/graphene based material has a good potential for the antibacterial agent which could be one of the options to combat the microbial resistance.

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References

- 1 Kumar, S.; Ojha, A.K.; Bhorolua, D.; Das, J.; Kumar, A.; Hazarika, A. Facile synthesis of CuO nanowires and Cu₂O nanospheres grown on rGO surface and exploiting its photocatalytic, antibacterial and supercapacitive properties. *Physica B Condens. Matter*, 2019, 558, 74-81, doi: 10.1016/j.physb.2019.01.040.
- 2 Mohamad Sukri, S.N.A.; Shameli, K.; Mei-Theng Wong, M.; Teow, S-Y.; Chew, J.; Ismail, N.A. Cytotoxicity and antibacterial activities of plant-mediated synthesized zinc oxide (ZnO) nanoparticles using *Punica granatum* (pomegranate) fruit peels extract. *J. Mol. Struct*, 2019, 1189, 57-65, doi: 10.1016/j.molstruc.2019.04.026.
- 3 Bashir, F.; Irfan, M.; Ahmad, T.; Iqbal, J.; Butt, M.T.; Sadeh, Y.; Umbreen, M.; Shaikh, I.A.; Moniruzzaman, M. Efficient utilization of low cost agro materials for incorporation of copper nanoparticles to scrutinize their antibacterial properties in drinking water. *Environ. Technol. Innov.*, 2021, 21, 101228, doi: 10.1016/j.eti.2020.101228.
- 4 Iliger, K.S.; Sofi, T.A.; Bhat, N.A.; Ahanger, F.A.; Sekhar, J.C.; Elhendi, A.Z.; Al-Huqail, A.A.; Khan, F. Copper nanoparticles: Green synthesis and managing fruit rot disease of chilli caused by *Colletotrichum capsici*. *Saudi J. Biol. Sci.*, 2021, 28(2), 1477-1486, doi: 10.1016/j.sjbs.2020.12.003.
- 5 Kumar, S.K.; Mamatha, G.; Muralidhara, H.; Anantha, M.; Yallappa, S.; Hungund, B.; Kumar, K.Y. Highly efficient multipurpose graphene oxide embedded with copper oxide nanohybrid for electrochemical sensors and biomedical applications. *J. Sci. Adv. Mater and Devices*, 2017, 2(4), 493-500, doi: 10.1016/j.jsamd.2017.08.003.
- 6 Yew, Y.P.; Shameli, K.; Miyake, M.; Ahmad Khairudin, N.B.; Mohamad, S.E.; Naiki, T.; Lee, K.X. Green biosynthesis of superparamagnetic magnetite Fe₃O₄ nanoparticles and biomedical applications in targeted anticancer drug delivery system: A review. *Arab. J. Chem.*, 2020, 13(1), 2287-2308, doi: 10.1016/j.arabjc.2018.04.013.
- 7 Nasrollahzadeh, M.; Issaabadi, Z.; Sajadi, S.M. Green synthesis of a Cu/MgO nanocomposite by *Cassia filiformis* L. extract and investigation of its catalytic activity in the reduction of methylene blue, congo red and nitro compounds in aqueous media. *RSC Adv.*, 2018, 8(7), 3723-3735, doi: 10.1039/c7ra13491f.
- 8 Pande, S.A. Green Synthesis of Biopolymer-Silver Nanocomposites for Gas Sensing. *Adv. Sci. Tech.*, 2016, 99, 54-60, doi: 10.4028/www.scientific.net/AST.99.54.
- 9 Bogdanovic, U.; Dimitrijevic, S.; Skapin, S.D.; Popovic, M. Rakocevic, Z.; Leskovac, A.; Petrovic, S.; Stoiljkovic, M.; Vodnik, V. Copper-polyaniline nanocomposite: Role of physicochemical properties on the antimicrobial activity and genotoxicity evaluation. *Mater Sci Eng C Mater Biol Appl*, 2018, 93, 49-60, doi: doi.org/10.1016/j.msec.2018.07.067.
- 10 Kumar, B.V.N.; Balla, P.K.; Chirauri, S.K.; Rao, T.K.V.; Ramakrishna, Y.; Rao, K.R. Synthesis and characterization of copper particles decorated reduced graphene oxide nano composites for the application of supercapacitors. *International Conference on Renewable Energy Research and Education (RERE-2018)*, India, 8-10 February 2018; Rao, K.R.; Suneetha, R.J.; Krishna, D.; American Institute of Physics: Andhra Pradesh, India, 2018; 040008,1-7, doi: 10.1063/1.5047973.
- 11 Aazadfar, P.; Solati, E.; Dorrarian, D. Properties of Au/Copper oxide nanocomposite prepared by green laser irradiation of the mixture of individual suspensions. *Opt. Mater.*, 2018, 78, 388-395, doi: 10.1016/j.optmat.2018.02.050.
- 12 Fahiminia, M.; Shamabadi, N.S.; Nasrollahzadeh, M.; Sajadi, S.M. Phytosynthesis of Cu/rGO using *Euphorbia cheiradenia* Boiss extract and study of its ability in the reduction of organic dyes and 4-nitrophenol in aqueous medium. *IET Nanobiotechnol*, 2019, 13(2), 202-213, doi: 10.1049/iet-nbt.2018.5175.
- 13 Kim, K.; Chaudhari, K.N.; Kim, S.; Kim, Y.; Shin, K.S. Facile single-step synthesis of Cu-rGO nanocomposite through simultaneous reduction process and its peroxidase mimic activity. *J. Ind Eng Chem*, 2021, 95, 388-396, doi: 10.1016/j.jiec.2021.01.013.
- 14 Karim, M.R.; Hayami, S. Chemical, thermal, and light-driven reduction of graphene oxide: Approach to obtain graphene and its functional hybrids. In: *Graphene Materials-Advanced Applications*, Kyzas, G.Z.; Mitropoulos, A.C.; IntechOpen Limited: London, 2017; pp. 1-17, doi: 10.5772/67808.

- 15 Yusoff, N. Synthesis of functionalized graphene/copper oxide (cuo) nanocomposites and their catalytic activity. Master Degree, Universiti Malaya, Malaysia, 2013.
- 16 Khan, M.S.; Yadav, R.; Vyas, R.; Sharma, A.; Banerjee, M.K.; Sachdev, K. Synthesis and evaluation of reduced graphene oxide for supercapacitor application. *Mater. Today: Proceedings*, 2020, 30, 153-156, doi: 10.1016/j.matpr.2020.05.403.
- 17 Shahriary, L.; Athawale, A.A. Graphene oxide synthesized by using modified hummers approach. *Int J Renew Energy Environ Eng*, 2014, 2(01), 58-63.
- 18 De Silva, K.K.H.; Huang, H.H.; Joshi, R.K.; Yoshimura, M. Chemical reduction of graphene oxide using green reductants. *Carbon*, 2017, 119, 190-199, doi: 10.1016/j.carbon.2017.04.025.
- 19 Gan, L.; Li, B.; Chen, Y.; Yu, B.; Chen, Z. Green synthesis of reduced graphene oxide using bagasse and its application in dye removal: A waste-to-resource supply chain. *Chemosphere*, 2019, 219, 148-154, doi: 10.1016/j.chemosphere.2018.11.181.
- 20 Dos Santos, P.L.; Katic, V.; Toledo, K.C.F; Bonacin, J.A. Photochemical one-pot synthesis of reduced graphene oxide/Prussian blue nanocomposite for simultaneous electrochemical detection of ascorbic acid, dopamine, and uric acid. *Sens. Actuators B Chem.*, 2018, 255, 2437-2447, doi: 10.1016/j.snb.2017.09.036.
- 21 Kang, X.; Teng, D.; Wu, S.; Tian, Z.; Liu, J.; Li, P.; Ma, Y.; Liang, C. Ultrafine copper nanoparticles anchored on reduced graphene oxide present excellent catalytic performance toward 4-nitrophenol reduction. *J Colloid Interface Sci*, 2020, 566, 265-270, doi: 10.1016/j.jcis.2020.01.097.
- 22 Yin, P.T.; Shah, S.; Chhowalla, M.; Lee, K-B. Design, synthesis, and characterization of graphene-nanoparticle hybrid materials for bioapplications. *Chem. Rev.*, 2015, 115(7), 2483-2531, doi: 10.1021/cr500537t.
- 23 Jayabalan, T.; Matheswaran, M.; Preethi, V.; Naina Mohamed, S. Enhancing biohydrogen production from sugar industry wastewater using metal oxide/graphene nanocomposite catalysts in microbial electrolysis cell. *Int. J. Hydrog. Energy*, 2020, 45(13), 7647-7655, doi: 10.1016/j.ijhydene.2019.09.068.
- 24 Pang, Y.L.; Tee, S.F.; Lim, S.; Abdullah, A.Z.; Ong, H.C.; Wu, C-H.; Chong, W.C.; Mohamadu, A.W.; Mahmoudi, E. Enhancement of photocatalytic degradation of organic dyes using ZnO decorated on reduced graphene oxide (rGO). *Desalination Water Treat*, 2018, 108, 311-321, doi: 10.5004/dwt.2018.21947.
- 25 Guo, M.; Zhao, Y.; Zhang, F.; Xu, L.; Yang, H.; Song, X.; Bu, Y. Reduced graphene oxide-stabilized copper nanocrystals with enhanced catalytic activity and SERS properties. *RSC Adv.*, 2016, 6(56), 50587-50594, doi: 10.1039/c6ra05186c.
- 26 Musa, A.A.M.; Hussein, M.Z.; Saiman, M.I.; Sani, H.A. Effect of gelatin-stabilized copper nanoparticles on catalytic reduction of methylene blue. *Nanoscale Res. Lett.*, 2016, 438(11), 1-13, doi: 10.1186/s11671-016-1656-6.
- 27 Manoj, D.; Saravanan, R.; Santhanalakshmi, J.; Agarwal, S.; Gupta, V.K.; Boukherroub, R. Towards green synthesis of monodisperse Cu nanoparticles: An efficient and high sensitive electrochemical nitrite sensor. *Sens. Actuators B Chem.*, 2018, 266, 873-882, doi: 10.1016/j.snb.2018.03.141.
- 28 Prasad, P.R.; Kanchi, S.; Naidoom E.B. In-vitro evaluation of copper nanoparticles cytotoxicity on prostate cancer cell lines and their antioxidant, sensing and catalytic activity: One-pot green approach. *J Photochem Photobiol B*, 2016, 161, 375-382, doi: 10.1016/j.jphotobiol.2016.06.008.
- 29 Varghese, B.; Kurian, M.; Krishna, S.; Athira, T.S. Biochemical synthesis of copper nanoparticles using *Zingiber officinalis* and *Curcuma longa*: Characterization and antibacterial activity study. *Mater. Today: Proceedings*, 2020, 25, 302-306, doi: 10.1016/j.matpr.2020.01.476.
- 30 Tu, Y.; Li, P.; Jiajia S.J.; Jiang, J.; Dai, F.; Yuanyan, W.G; Liang, C.; Guosheng, S.; Yanwen, T.; Haiping, F. Remarkable antibacterial activity of reduced graphene oxide functionalized by copper ions. In: *Advance Functional Materials*, Ritterbusch, J.; Vita, M.D.; Lenders, J.; Meskine, H. John Wiley & Sons, Inc.: Weinheim, 2021; pp. 1-16, doi: 10.1002/adfm.202008018.
- 31 Yang, Z.; Hao, X.; Chen, S.; Ma, Z.; Wang, W.; Wang, C.; Yue, L.; Sun, H.; Shao, Q.; Murugadoss, V.; Guo, Z. Long-term antibacterial stable reduced graphene oxide nanocomposites loaded with cuprous oxide nanoparticles. *J Colloid Interface Sci*, 2019, 533, 13-23, doi: 10.1016/j.jcis.2018.08.053.
- 32 Rios, P.L.; Povea, P.; Cerda-Cavieres, C.; Arroyo, J.L.; Morales-Verdejo, C.; Abarca, G.; Camarada, M.B. Novel in situ synthesis of copper nanoparticles supported on reduced graphene oxide and its application

- as a new catalyst for the decomposition of composite solid propellants. *RSC Adv.*, 2019, 9(15), 8480-8489, doi: 10.1039/c9ra00789j.
- 33 Prashanti, B.; Damodharam, T. Fabrication of graphene–cuo nanocomposite with improved photocatalytic degradation for palladium solution under solar light irradiation. *J. Nanosci. Technol.*, 2018, 4(5), 497-499, doi: 10.30799/jnst.149.18040509.
- 34 Ganesan, K.; Jothi, V.K.; Natarajan, A.; Rajaram, A.; Ravichandran, S.; Ramalingam, S. Green synthesis of copper oxide nanoparticles decorated with graphene oxide for anticancer activity and catalytic applications. *Arab. J. Chem.*, 2020, 13(8), 6802-6814, doi: 10.1016/j.arabjc.2020.06.033.
- 35 Zhang, K.; Suh, J.M.; Lee, T.H.; Cha, J.H.; Choi, J.W.; Jang, H.W.; Varma, R.S.; Shokouhimehr, M. Copper oxide-graphene oxide nanocomposite: efficient catalyst for hydrogenation of nitroaromatics in water. *Nano Convergen*, 2019, 6(1), 1-7, doi: 10.1186/s40580-019-0176-3.
- 36 Na, H.G.; Cho, H.Y.; Kwon, Y.J.; Kang, S.Y.; Lee, C.; Jung, T.K.; Lee, H-S.; Kim, H.W. Reduced graphene oxide functionalized with Cu nanoparticles: Fabrication, structure, and sensing properties. *Thin Solid Films*, 2015, 588, 11-18, doi: 10.1016/j.tsf.2015.03.078.
- 37 Pratik, A.; Biswal, S.K.; Haridoss, P. Impact of enhanced interfacial strength on physical, mechanical and tribological properties of copper/reduced graphene oxide composites: Microstructural investigation. *Ceram. Int.*, 2020, 46(14), 22539-22549, doi: 10.1016/j.ceramint.2020.06.014.
- 38 Moozarm Nia, P.; Woi, P.M.; Alias, Y. Facile one-step electrochemical deposition of copper nanoparticles and reduced graphene oxide as nonenzymatic hydrogen peroxide sensor. *Appl. Surf. Sci.*, 2017, 413, 56-65, doi: 10.1016/j.apsusc.2017.04.043.
- 39 Xu, X.; Shen, J.; Qin, J.; Duan, H.; He, G.; Chen, H. Cytotoxicity of bacteriostatic reduced graphene oxide-based copper oxide nanocomposites. *JOM*, 2019, 71(1), 294-300, doi: 10.1007/s11837-018-3197-1.
- 40 Zhang, W.; Chang, Q.; Xu, L.; Li, G.; Yang, G.; Ding, X.; Wang, X.; Cui, D.; Jiang, X. Graphene oxide-copper nanocomposite-coated porous cap scaffold for vascularized bone regeneration via activation of Hif-1alpha. *Adv. Healthc. Mater.*, 2016, 11(5), 1299-1309, doi: 10.1002/adhm.201500824.
- 41 Ghorbi, E.; Namavar, M.; Rashedi, V.; Farhadinejad, S.; Pilban Jahromi, S.; Zareian, M. Influence of nano-copper oxide concentration on bactericidal properties of silver–copper oxide nanocomposite. *Colloids Surf.*, 2019, 580, 123732, doi: 10.1016/j.colsurfa.2019.123732.
- 42 Ismail, N.A.; Shameli, K.; Wong, M.M.; Teow, S-Y.; Chew, J.; Mohamad Sukri S.N.A. Antibacterial and cytotoxic effect of honey mediated copper nanoparticles synthesized using ultrasonic assistance. *Mater Sci Eng C Mater Biol Appl*, 2019, 104, 109899, doi: 10.1016/j.msec.2019.109899.
- 43 Lopez-Lima, D.; Mtz-Enriquez, A.I.; Carrión, G.; Basurto-Cereceda, S.; Pariona, N. The bifunctional role of copper nanoparticles in tomato: Effective treatment for Fusarium wilt and plant growth promoter. *Sci. Hortic.*, 2021, 277, 109810, doi: 10.1016/j.scienta.2020.109810.
- 44 Raju, C.H.A.I.; Nooruddin, S.; Babu, K.S. Studies on leaf extract mediated synthesis of copper nanoparticles for the removal of bromo cresol green dye from synthetic waste waters. *Int. J. Sci. Eng. Technol. Res.*, 2017, 6(10), 1404-1411.
- 45 Liu, W.; Tao, Z.; Wang, D.; Liu, Q.; Zhang, Y.; Zhang, Y.; Dong, A. Immobilization of Cu (II) via a graphene oxide-supported strategy for antibacterial reutilization with long-term efficacy. *J Hazard Mater*, 2020, 124601, doi: 10.1016/j.jhazmat.2020.124601.
- 46 Alimardani, V.; Abolmaali, S.S.; Borandeh, S. Antifungal and antibacterial properties of graphene based nanomaterials: A mini-review. *J. Nanostruct*, 2019, 9(3), 402-413, doi: 10.22052/JNS.2019.03.002.
- 47 Navya Rani, M.; Murthy, M.; Shyla Shree, N.; Ananda, S.; Yogesh, S.; Dinesh, R. Cuprous oxide anchored reduced graphene oxide ceramic nanocomposite using *Tagetes erecta* flower extract and evaluation of its antibacterial activity and cytotoxicity. *Ceram. Int.*, 2019, 45(18), 25020-25026, doi: 10.1016/j.ceramint.2019.04.195.
- 48 Hegab, H.M.; ElMekawy, A.; Zou, L.; Mulcahy, D.; Saint, C.P.; Ginic-Markovic, M. The controversial antibacterial activity of graphene-based materials. *Carbon*, 2016, 105, 362-376, doi: 10.1016/j.carbon.2016.04.046.
- 49 Sanchez-Lopez, E.; Gomes, D.; Esteruelas, G.; Bonilla, L.; Lopez-Machado, A.L.; Galindo, R.; Cano, A.; Espina, M.; Ettcheto, M.; Camins, A.; Silva, A.M.; Durazzo, A.; Santini, A.; Garcia, M.L.; Sauto, E.B. Metal-

- based nanoparticles as antimicrobial agents: An overview. *Nanomaterials* (Basel), 2020, 10(2), 1-39, doi: 10.3390/nano10020292.
- 50 Alayandea, A.B.; Obaida, M.; Kima, I.S. Antimicrobial mechanism of reduced graphene oxide-copper oxide (rGO/CuO) nanocomposite films: The case of *Pseudomonas aeruginosa* PAO1. *Mater. Sci. Eng. C*, 2020, 109, 110596, doi: 10.1016/j.msec.2019.110596.
- 51 Selim, M.S.; Samak, N.A.; Hao, Z.; Xing, J. Facile design of reduced graphene oxide decorated with Cu₂O nanocube composite as antibiofilm active material. *Mater. Chem. Phys.*, 2020, 239, 122300, doi: 10.1016/j.matchemphys.2019.122300.
- 52 Chen, M.; Li, Z.; Chen, L. Highly antibacterial rGO/Cu₂O nanocomposite from a biomass precursor: Synthesis, performance, and mechanism. *Nano Materials Sci.*, 2020, 2(2), 172-179, doi: 10.1016/j.nanoms.2019.09.005.
- 53 Zhu, J.; Wang, J.; Uliana, A.A.; Tian, M.; Zhang, Y.; Zhang, Y.; Volodin, A.; Simoens, K.; Yuan, S.; Li, J.; Lin, J.; Bernaerts, K.; Van der Bruggen, B. Mussel-inspired architecture of high-flux loose nanofiltration membrane functionalized with antibacterial reduced graphene oxide-copper nanocomposites. *ACS Appl Mater Interfaces*, 2017, 9(34), 28990-29001, doi: 10.1021/acsami.7b05930.
- 54 Menazea, A.A.; Ahmed, M.K. Synthesis and antibacterial activity of graphene oxide decorated by silver and copper oxide nanoparticles. *J. Mol. Struct.*, 2020, 1218, 128536, doi: 10.1016/j.molstruc.2020.128536.
- 55 Rajapaksha, P.; Cheeseman, S.; Hombach, S.; Murdoch, B.J.; Gangadoo, S.; Blanch, E.W.; Truong, Y.; Cozzolino, D.; McConville, C.F.; Crawford, R.J.; Truong, V.K.; Elbourne, A.; Chapman, J. Antibacterial properties of graphene oxide-copper oxide nanoparticle nanocomposites. *ACS Appl Bio Mater*, 2019, 2(12), 5687-5696, doi: 10.1021/acsabm.9b00754.
- 56 Li, Y.; Yang, D.; Cui, J. Graphene oxide loaded with copper oxide nanoparticles as an antibacterial agent against *Pseudomonas syringae* pv. tomato. *RSC Adv.*, 2017, 7(62), 38853-38860, doi: 10.1039/c7ra05520j.