GREEN SYNTHESIS OF SILVER AND COPPER NANOPARTICLES USING HYDROXYETHYL CELLULOSE AND ITS ANTIBACTERIAL ACTIVITY

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MASTER OF SCIENCE
(ADVANCED MATERIALS)

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SUPERVISOR'S DECLARATION

We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science in Advanced Materials

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STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that this thesis has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institution.

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ABSTRAK

Disertasi ini memfokuskan pada pengenalan satu kaedah baharu mensintesis zarah-zarah nano argentum dan kuprum (AgNPs dan CuNPs) menggunakan hidroksietil selulosa (HEC). Kajian ini dilakukan melalui kaedah lestari untuk mengelakkan penggunaan bahan kimia yang berbahaya dan juga untuk menjimatkan kos pengeluaran. Disertasi ini juga menerangkan keadaan-keadaan optimum untuk sintesis zarah-zarah nano dengan menganalisis pelbagai parameter, seperti isipadu pelopor-pelopor argentum nitrat ($AgNO_3$) dan kuprum nitrat ($Cu(NO_3)_2$), kepekatan HEC, masa tindak balas serta suhu. Pada 100 °C, tindak balas AgNPs selesai dalam 30 minit manakala CuNPs pula dalam 3 minit. Kehadiran kedua-dua AgNPs dan CuNPs dibuktikan oleh spektroskopi ultraungu-tampak (UV-Vis), yang menunjukkan puncak resonans plasmon permukaan (SPR) masing-masing pada 410 - 430 nm dan 550 - 600 nm. Kehadiran zarah-zarah nano dan struktur kristal disahkan oleh difraktometri sinar-X (XRD) dan sinar-X sebaran tenaga (EDX). Pencirian struktur dan morfologi AgNPs dan CuNPs dilakukan menggunakan mikroskop elektron penghantaran (TEM) dan mikroskop elektron pengimbasan pancaran medan (FESEM). Aktiviti antibakteria zarah-zarah nano juga diuji melalui kaedah peresapan agar-agar, kepekatan perencatan minimum (MIC) dan kepekatan bakterisidal minimum (MBC). Kajian ini juga menguji kesan AgNPs, CuNPs dan campuran Ag-CuNPs terhadap bakteria Gram-positif dan Gram-negatif. Zarah-zarah nano menunjukkan aktiviti antibakteria yang baik terhadap *Bacillus subtilis* (*B. subtilis*), *Escherichia coli* (*E. coli*), *Pseudomonas aeruginosa* (*P. aeruginosa*), *Entercoccusfaecalis* (*E. faecalis*) dan *Staphylococcus aureus* (*S. aureus*). Zon perencatan bagi zarah-zarah nano ini berubah mengikut jenis bakteria. Zon perencatan yang lebih besar diperhatikan pada bakteria Gram-negatif (*E. coli*). Saiz zon perencatan tersebut ialah 19 mm untuk AgNPs, manakala 16 mm untuk CuNPs dan Ag-CuNPs. Oleh itu, disertasi ini menyimpulkan bahawa penemuan kaedah lestari yang sesuai untuk sintesis AgNPs dan CuNPs mempunyai kesan perencatan yang baik terhadap bakteria, justeru terdapat pelbagai aplikasi yang berpotensi untuk zarah-zarah nano ini.

ABSTRACT

This dissertation is mainly focused about the introduction of a new method of synthesizing silver and copper nanoparticles (AgNPs and CuNPs) using hydroxyethyl cellulose (HEC). The study was done via green chemistry method to avoid the usage of some hazardous chemicals and also to save the cost of production. This thesis describes the optimal conditions for the synthesis of nanoparticles by analysing various parameters, such as the volume of the precursors silver nitrate ($AgNO_3$) and copper nitrate ($Cu(NO_3)_2$), the concentration of HEC, reaction times and temperature. At 100 °C, the AgNPs reaction went to completion in 30 min while the CuNPs reaction about 3 min. The presence of both AgNPs and CuNPs were assured by ultraviolet visible spectroscopy (UV-Vis), which showed surface plasmon resonance (SPR) peaks at 410 - 430 nm and 550 - 600 nm respectively. The presence of the nanoparticles and the crystal structure were confirmed by X-ray diffractometry (XRD) and energy-dispersive X-ray (EDX). The structural and morphological characterisations of the AgNPs and CuNPs were performed using a transmission electron microscope (TEM) and field emission scanning electron microscope (FESEM). The antibacterial activities of the nanoparticles were also studied via the agar-well diffusion method, minimum inhibition concentration (MIC) and minimum bactericidal concentration (MBC). The research also tested the effects of AgNPs, CuNPs and Ag-CuNP mixtures on Gram-positive and Gram-negative bacteria. The nanoparticles showed good antibacterial activity against *Bacillus subtilis* (*B. subtilis), Escherichia coli* (*E. coli*), *Pseudomonas aeruginosa* (*P. aeruginosa*), *Entercoccus faecalis* (*E. faecalis*) and *Staphylococcus aureus* (*S. aureus*). The inhibition zones for these nanoparticles varied based on the type of bacteria. The larger inhibition zone was observed on Gram-negative bacteria (*E. coli*). The sizes of the said inhibition zones were 19 mm for AgNPs, 16 mm for CuNPs and Ag-CuNPs. Thus, it can be concluded that the invention of a feasible green method for the synthesis of AgNPs and CuNPs had a good inhibitory effect on the bacteria and hence, there are various potential applications for these nanoparticles.

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LIST OF SYMBOLS

+	Add
α	Alpha
Z	Atomic number
β	Beta
Cu^{2+}	Copper (II) ions
Cu^{0}	Copper particles
°	Degree
°C	Degree Celcius
$	Dollar
g	Gram
g/mol	Gram per mol
h	Hour
keV	Kilo electronvolt
Kg	Kilogram
kV	Kilovolts
L	Litre
µL	Microliter
mA	Milliampere
mL	Millilitre
mm	Millimetre
min	Minutes
M	Molarity
M_w	Molecular weight
nm	Nanometer
n	Number of mol
%	Percentage
pH	Potential of hydrogen
M_r	Relative molar mass
Ag^{+}	Silver ion
Ag^{0}	Silver particles
θ	Theta
V	Volume
λ	Wavelength
Wt%	Weight percentage

LIST OF ABBREVIATIONS

Al	Aluminium
ATCC	American Type Culture Collection
$C_6H_8O_6$	Ascorbic acid
$(C_{16}H_{18}N_2O_4SNa)$	Benzylpenicillin sodium
Co	Cobalt
CFU	Colony forming unit
Cu	Copper
$Cu(NO_3)_2$	Copper (II) nitrate
CuNPs	Copper nanoparticles
EDX	Energy dispersive X-ray
E-Test	Epsilometer test
Fcc	Face-centered cubic
Au	Gold
AuNPs	Gold nanoparticles
HTAB	Hexadecyltrimethylammonium bromide
HRTEM	High-resonance transmission electron microscopy
H_2O_2	Hydogen peroxide
HEC	Hydroxyethyl cellulose
OH	Hydroxy groups
HP	Hyperbranched polyurethane
FESEM	Field emission scanning electron microscopy
Fe	Iron
FeNPs	Iron nanoparticles
Fe_2O_3	Iron oxide
Mn	Manganese
MBC	Minimum bactericidal concentration
MIC	Minimum inhibition concentration
MRI	Magnetic resonance imaging
MHA	Mueller Hinton agar
MHB	Mueller Hinton broth
NPs	Nanoparticles
Ni	Nickel
PAN	Polyacrylonitrile
PTFE	Polytetrafluoroethylene
PVA	Polyvinyl alcohol
PVP	Polyvinylpyrolidone
rpm	Revolutions per minute
Si	Silicon
Ag	Silver
Ag-CuNPs	Silver-copper nanoparticles
AgNPs	Silver nanoparticles
$AgNO_3$	Silver nitrate
H_2SO_4	Sulphuric acid
ROS	Reactive oxygen species
R&D	Research & Development
SPR	Surface plasmon resonance
Ti	Titanium
TEM	Transmission electron microscopy
UV-Vis	Ultraviolet visible spectroscopy
USD	United State Dollar
H_2O	Water

XRD	X-ray diffraction
Zn	Zinc

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