UNIVERSITI TEKNOLOGI MARA

SYNTHESIS OF 3D HYDROGEL GRAPHENE QUANTUM DOTS NANOCOMPOSITES MODIFIED FIBRE OPTIC FOR BIOSENSORS APPLICATION

MUHAMMAD HAZIQ BIN NOOR AKASHAH

Thesis submitted in fulfillment
of the requirements for the degree of
Master of Science
(Mechanical Engineering)

College of Engineering

December 2022

ABSTRACT

In this study, a novel fibre optic sensor towards biosensing application was developed by introducing polymeric supporting membranes of hybrid hydrogels (HH) and nanomaterials of graphene quantum dots (GQDs) as coating materials. HH was formed by introducing chitosan (CH) as an additive that would further aid in introducing the amide group that was lacking in agarose (AG) hydrogel. GQDs were synthesised using a chemical reduction method transforming the precursor graphene oxide (GO) into GQDs nanostructures. GO was synthesised prior to that using a modified Hummers' method. GQDs were optimised by varying the parameters in the chemical reduction method with the optimum condition for synthesis ~2 nm of GQDs was obtained by using 25 µL of dispersed in 5 mL of sulfuric acid, 0.014 g of potassium permanganate and 0.1 mL of hydrogen peroxide. The optimum GQDs were found to be having an average size of 2.481 ± 0.649 nm as confirmed by transmission electron microscopy, subtle x-ray diffraction peak at around 2θ=26°, loaded with hydroxyl (OH), carbonyl (C=O) and carbon bonds found from Fourier transform infrared analysis reflect that GQDs have many oxygenated functional groups making them soluble in water and stable fluorescence stability for up to 90 days with a quantum yield of 48%. A conventional fibre optic cable was used and exposed its core region made from silica by using mechanical etching via a fibre optic cable stripper. Before that, signal validation for the uncladded region (1, 2 and 3 cm) was performed with 2 cm of uncladded and was found to give the signal of 1660.74 a.u. and efficient uncladded length to be spliced. The fibre optic core coated HH was then let to be coated with the optimum coating thickness of HH on fibre optic was found at 122.18 ± 0.3 µm by 2% total concentration of HH (2% w/v of AG with 2% v/v of CH) with confirmation by signal validation from the spectrometer with laser as a light source. After that, the coated HH (2% w/v of AG with 2% v/v of CH) probe was further coated with 2 nm GQDs via the drop-cast method and let to dry overnight. In enhancing the surface binding between the coating materials to the sensor probe, the unclad probe was further treated with sodium hydroxide, chromic acid, silane or ethanol prior coated to HH-GQDs. These coated probes were further tested for their sensing performance in the detection of refractive index (RI) changes (Δn) using glycerol solutions. Fibre optic pre-treated with chromic acid coated to HH-GQDs was found to obtain the lowest baseline error of 0.026% which improves the adhesive force between HH and the fibre optic core surface. The sensitivity was then calculated at 17358.0±0.21 RIU^{-1} with a coefficient of determination; R^2 of 0.94. As a proof of concept for biosensor applications, the protein of *streptavidin* (0.1 mg/mL) was successfully immobilised by incubation for 15 minutes on the fibre optic core treated with chromic acid and coated with HH-GQDs. 10 mg/mL of *bovine serum albumin* (BSA) was used as a blocking agent after incubation for 10 minutes on the HH-GQDs matrix. The sensor was successfully able to detect the *biotin* (0.1 mg/mL) after incubation for 15 minutes and washing with PBS. The binding affinity in respect of the change in peak wavelength (Δnm) of HH-GQDs matrix to the protein was measured at 90.445 $a.u.nm^{-1}$ with R^2 of 0.933. The sensor also shows excellent specificity, hence holding a bright potential as a highly sensitive biosensor.

ACKNOWLEDGEMENT

Ever since the start of my higher education phase, learning has always been meaningless and something I viewed as just a reason to get a job. As I am in my halfway through degree, I have flunked in my final year project (FYP) 1 and pretty much knocked me out and realising that I can only rely on myself and leave the rest to Allah. Learned a lot, reflect myself a lot too and keep on learning still. Be it in my struggle to keep my financials stable, to balance between part-time work and study, family has always been my top priorities. They have never been the obstruction or the reason holding me back, especially my mother who always keen on seeing her sons and daughters to pursue their studies. Moreover, the fact that knowing my mother is a single mother raising her six children, I would use any kind of resources to help her get through it be it in the form of my time and energy.

As I further my study to master's level, she still be encouraging me and still would be fascinated by how I explained every single detail on my lab work, writing skills, or even any online workshop or webinar that I have joined. She never had the opportunity. Regardless, I am here to fill her with every detail, the backbone of my will to live day to day. Throughout this journey, ups and downs moment are countless. Deep down, I know I am not the best student a teacher may have. Yet my strive for delivering good results has always been the fundamental of myself. Challenges are normal, if they are not there, I am practically not living and probably still be in my comfort zone. Many thanks can never be said enough to those who supported me morally especially Aiza, friends (Nad, Edlan, Kwan, Luke), my lab partner since day 1, Rafal and not to forget Lili too.

Most I would be thanking is my main supervisor Dr Siti Rabizah who have been in patience with me throughout my studies ever since my undergraduate study. Pushed me in a way that I would not know my potential. Joining innovation competitions, presenting in conference, and even drafting for Q1 paper has never been the A-list of my life. Not to forget Dr Rozina also who adhere relentless with my master's journey persistently and all the technical staff who helped and gave their best cooperation. Professor Dr Khairunisak from USM whom with her privilege I am able to characterise all of my TEM samples, much appreciated and Dr Patricia J. Scully who I have never met officially but I believe been giving all the moral support she can from 11069 KM away. Hereby, I thank you all and sending my pray to all of those who supported me to gain a prosperous, successful, and joyous life.

Last but not least, I want to thank me, for believing in me, for doing all this hard work, for having no days off, for never quitting, thank you and congratulations dearself for completing this study!

TABLE OF CONTENTS