

***IN VITRO BIOACTIVITY OF NOVEL CHITOSAN/GELATIN/HALLOYSITE  
NANOSTRUCTURED COATINGS ON ANODISED TITANIUM VIA  
ELECTROPHORESATION FOR BONE IMPLANT***

**JANIFAL ALIPAL**

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This thesis is dedicated to my parents. May your souls rest in peace, Alipal and Sabaidah, who have always loved me unconditionally and taught me to work hard for the things I aspire to accomplish.



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## ABSTRACT

Electrophoretic deposition (EPD) involves coating densification via matrix micro/nano-filling on a template-assisted substrate. This mechanical interlock technique has recently been used to avoid coating cracking and delamination. This thesis reports that EPD organic-inorganic nanostructured coatings containing chitosan/gelatin hydrogel and halloysite nanotubes (HNTs) produce an ideal mechanical interlock. In the proposed bio-composite, HNTs are used to densify the coating's mixture. The mechanical interlocking of the proposed bio-composite coating defines its net mechanistic bioactivity. Prior to EPD, the study goes through a substrate pre-processing step in which the cp-Ti surface is modified using micro-arc anodic oxidation (MAO) in a CaP-based electrolyte (a mixture of  $\beta$ -glycerophosphate and calcium acetate). The study discovered that anodised titanium (MAT) in donut-shaped morphology (MAO 350 V) has better mechanical stability and osteogenic cellular response compared to the needle-like structure. The findings determined that the donut-shaped MAT microstructure is the best next choice for the EPD substrate in the coating mechanical interlock study. Despite the fact that the EPD processing parameters were varied (10-30 V; 5-20 min), the mechanically interlock nanostructured coating (template-assisted EPD) significantly improved coating adhesion and osteogenic development in this study. In coating fabrication, the weight fraction of HNTs in the hydrogel is critical, and this study determined that the optimal composition of a steric stabilised organic-inorganic EPD suspension for chitosan/gelatin/HNTs is 6:14:12 g/L. Modifying implant surfaces using novel techniques such as varying substrate morphology and/or degraded coatings has become a popular method for improving implant osseointegration. This recent study established that specific surface features influence how bone cells interact with a material and which specific surface features result in optimal bone integration. In this thesis, MAT is designed to be a highly bioactive EPD substrate, resulting not only in a highly stable coating structure but also in improved osteogenic development, specifically osteoblast mineralisation and differentiation.

## ABSTRAK

Pemendapan elektroforetik (EPD) berkapasiti untuk menumpatkan salutan pada permukaan biometalik melalui mendapan mikro/nano matriks bahan berbantu acuan. Teknik ini dikenali sebagai pelapis mekanikal saling berkait dan ia digunakan untuk mengelakkan keretakan dan salutan tertanggal pada permukaan bahan. Laporan kajian ini menunjukkan bahawa salutan berstruktur nano unsur-unsur organik dan bukan organik hasil campuran hidrogel kitosan/gelatin dan tiub nano halloysite (HNTs) memiliki jalinan mekanikal yang ideal. Dalam biokomposit yang dicadangkan, HNT menumpatkan campuran salutan. Jalinan mekanikal salutan biokomposit ini secara fizikal dinilai sebagai penentu kepada bioaktiviti permukaan bahan. Sebelum EPD, kajian ini melalui langkah pra-pemprosesan substrat yang mengubah permukaan cp-Ti menggunakan pengoksidaan anodik mikro-arka (MAO) dalam elektrolit campuran  $\beta$ -gliseroftosfat dan kalsium asetat. Kajian mendapati permukaan titanium yang teroksida (MAT) dalam morfologi berbentuk donat (MAO 350 V) mempunyai kestabilan mekanikal dan tindak balas sel osteogenik yang baik berbanding dengan struktur MAT morfologi berbentuk jarum. Oleh itu, MAT berbentuk donat menjadi pilihan untuk substrat EPD dalam kajian pelapis mekanikal saling berkait. Walaupun parameter pemprosesan EPD bervariasi (10-30 V; 5-20 min), salutan nanostruktur secara mekanikal menjadi faktor utama dalam lekatan lapisan dan pengembangan osteogenik dalam kajian ini. Dalam fabrikasi lapisan, pecahan berat HNTs dalam hidrogel adalah penting, dan kajian ini mendapati komposisi optimum EPD organik-anorganik koloid yang stabil untuk kitosan/gelatin/HNT adalah 6:14:12 g/L. Modifikasi morfologi substrat yang berbeza dan/atau lapisan yang terdegradasi telah menjadi kaedah popular dalam meningkatkan osseointegration implan. Kajian baharu ini membuktikan bahawa ciri permukaan tertentu mempengaruhi interaksi sel tulang terhadap bahan salutan, lalu menghasilkan integrasi tulang yang optimum. Dalam tesis ini, MAT menjadi substrat EPD yang sangat bioaktif, yang bukan sahaja menghasilkan struktur lapisan EPD yang sangat stabil tetapi juga dalam pengembangan osteogenik yang lebih baik, khususnya mineralisasi dan pembezaan osteoblast.

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