

Electrochemical Impedance Spectroscopy (EIS) Study of Modified Type-316L Stainless Steel (SS) as an Effective Biomaterial for Orthopedic Implant Applications

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For centuries, metals and alloys have been used in substitution for anatomical structures owing to their extraordinary mechanical properties [1]. Type 316L Stainless Steel (SS) is known to be widely used for implantation purposes in orthopedic surgery due to its corrosion resistance, effective cost and superior mechanical properties [1,2]. However, several incidences of failures involving type 316L SS have been occurred which released the metal ions into tissues [1] and blood stream surrounding the implants, thus causing histological changes of the local tissue by direct toxic effects or local hypersensitivity reaction [3]. It is highly probable that electrophoretically deposited Multiwall Carbon Nanotubes (MWCNTs) and Hydroxyapatite (HA) on type 316L SS could minimize this limitation, and further investigation of its corrosion behavior using *in vitro* electrochemical techniques [1,4] may confirm the acceptability of modified SS as an effective biomaterial to be used as orthopedic implants.

An ideal orthopedic implant should possess biocompatible, osteoconductivity [5], superior mechanical properties [6-8] and excellent corrosion resistance to physiological environments of the human body [1,9]. Different electrochemical techniques for evaluation of corrosion behavior have been studied extensively included Electrochemical Impedance Spectroscopy (EIS), cyclic potentiodynamics polarization and Open Circuit Potential (OCP) studies [2]. EIS is a useful approach to monitor in situ electrochemical changes [10] and to develop understanding of the physical process occurring at the electrode-electrolyte interface [11-13]. EIS study was carried out based on an appropriate circuit model in order to fit the experimental data to equivalent circuit model parameters, whereas each parameter would represents macroscopic physical quantity which contributing to interface impedance [11]. An equivalent circuit was selected to represent the electrochemical behavior of metals implants that have been coated with an unsealed porous film [14]. The equivalent circuit consists of solution resistance of the electrolyte (experimental human body fluid solution), R_s , capacitance of non-defective coating layer, C_p , charge transfer resistance of electrolyte's penetration through pinholes, R_p , polarization resistance of substrate, R_b and electrical double-layer capacitance at substrate/electrolyte interface, C_b [4]. This physical model of the electrochemical reactions that occurred at the electrodes is important to interpret the electrochemical behavior of the metal implants from EIS spectra [15].

Certain amount of current density is necessary to generate activity in stimulation process which resulted in interface impedance of the metal implants [4]. The equivalent model comprises an interface capacitance, shunted by charge transfer resistance and in series with solution resistance [16]. Then, a theoretical equation has been used to determine the interface capacitance impedance and solution resistance in order to confirm the results that correspond well with the fitted parameter values, hence validating the model equations [4]. Impedance behaviors of the metals implants could be expressed either in a Bode plot of logarithm of impedance modulus ($|Z|$) and phase angle (Φ) as a function of logarithm of frequency or in a Nyquist plot of Z''

(ω) as a function of $Z'(\omega)$ [14,17]. The Bode plots of logarithm total impedance $|Z|$ vs. frequency for uncoated SS and coated samples of SS/MWCNT-OH are shown in Figure 1(a) and for uncoated SS and

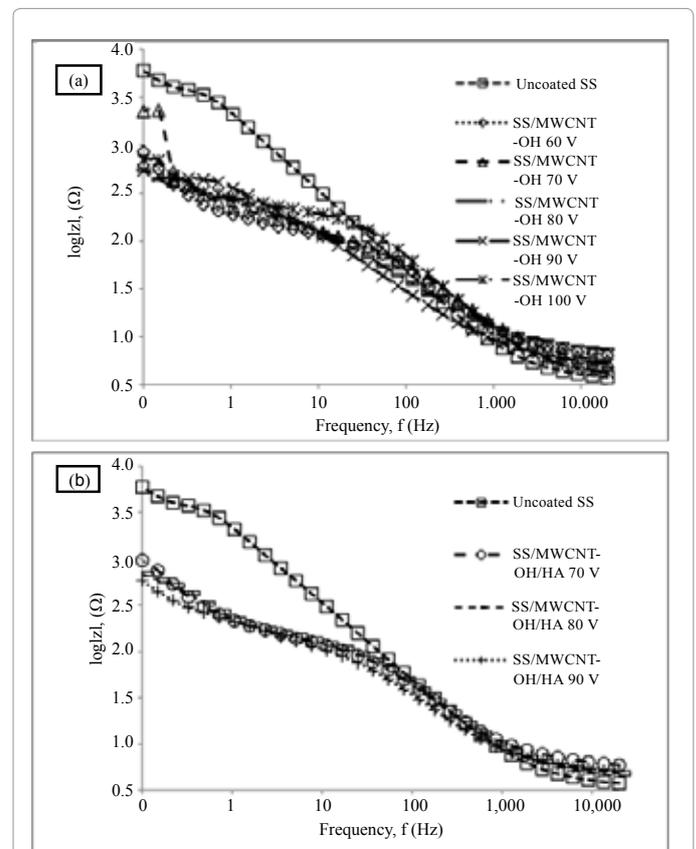


Figure 1: Bode plots of logarithm total impedance $|Z|$ vs. frequency of (a) uncoated SS and coated samples of SS/MWCNT-OH and (b) uncoated SS and coated samples of SS/MWCNT-OH/HA.

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coated samples of SS/MWCNT-OH/HA as in Figure 1(b). Bode plots were used to estimate the impedance parameters in equivalent circuit model, thereby indicates the corrosion behavior of modified SS after immersed in Phosphate Buffer Saline (PBS) solution. The decrease of polarization resistance and coating layer capacitance imply that the coating layers of MWCNT-OH and HA tend to protect SS by reducing the penetrations of anions from PBS solution [1,14], hence improving corrosion resistance of the modified SS [1].

Cyclic potentiodynamic polarization study is carried out when the susceptibility of metal implants to localized corrosion need to be addressed [18]. Pitting potential, E_b and pit protection potential, E_{prot} could be determined from cyclic potentiodynamic polarization curve [19] as these parameters are needed to evaluate pitting corrosion of metal implants [2]. Direct measurement of pit propagation mechanisms is proportional to the area under cyclic potentiodynamic polarization curve [2,19], as larger area of hysteresis loop indicates higher rate of pits formation [1]. OCP measurement provides simple evaluation of corrosion behavior including oxide film formation and metal implants passivation in experimental body fluid environment as a function of time [2,20]. The increasing potential in positive direction achieving steady state indicates the metal implants remains protective [17,20] while potential drops toward negative direction implies no film formation or break in the film [17] due to dissolution of carbon, ferum and oxides particles from metal implants surface when immersed in experimental body fluid solution [4]. Cyclic potentiodynamic polarization and OCP studies are able to provide useful evaluation of corrosion behavior [20] and pitting corrosion resistance [19] of metal implants, however, these electrochemical techniques provide less information of adsorption behavior and interactions of body fluid environment to metal implant surface [21], thus leaving EIS study as suitable technique to investigate the acceptability of modified SS to be used as orthopedic implants.

So, is the corrosion behavior study of the modified SS by EIS is enough to confirm its acceptability as an effective orthopedic implants? While *in vitro* EIS study have indeed able to show lower polarization resistance and coating layer capacitance of modified SS, more quantitative determination of corrosion products need to be performed in order to investigate their adverse effects on tissues surrounding the implants [19]. Hence, both *in vitro* cell-culture experiments and clinical retrieval studies [22] in conjunction with *in vivo* biological investigations of modified SS need to be investigated to ensure its continuous clinical applications in biomedical field.

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