



Full Length Article

Optimizing grafting thickness of zwitterionic sulfobetaine polymer on cross-linked polyethylene surface to reduce friction coefficient

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ABSTRACT

The purpose of this study involves reducing the friction coefficient of cross-linked polyethylene (CLPE) by optimizing the grafting thickness of zwitterionic poly(2-(methacryloyloxy)ethyl dimethyl-(3-sulfopropyl)ammonium hydroxide) (PMEDSAH) via a photo-induced free radical polymerization process. Cross-sectional TEM images and XPS spectra indicated that the PMEDSAH layers were successfully grafted on the CLPE. The grafting thickness increased when UV-irradiation time, UV-irradiation energy, and MEDSAH monomer concentration increased. The contact angle data indicated that the PMEDSAH layers successfully imparted hydrophilicity (below 20°) on the CLPE surface. The friction coefficient test and the resulting 3-D confocal images confirmed a significant decrease in the friction coefficient with increases in the grafting thickness. A minimum grafting thickness of 200 nm PMEDSAH was required to minimize the friction coefficient in an aqueous medium. However, a PMEDSAH-grafting thickness of approximately 40 nm was sufficient to reveal the minimized friction coefficient in simulated body fluids (ionic buffer solution) due to the salt-induced hydration effect. The results defined the minimum required thickness of PMEDSAH grafting to minimize the friction coefficient on CLPE surface that can potentially be applied for artificial hip joints.

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1. Introduction

Cross-linked polyethylene (CLPE) is widely used in various engineering fields due to its conducive mechanical properties, good formability, and reasonably low cost [1]. Recently, CLPE is also attracting significant attention in biomedical applications due to its mechanical durability and good formability. The hemisphere liner for femoral head in total hip arthroplasty (THA) is one of the most frequently used biomedical applications of CLPE [2]. Hip injuries continuously increase with respect to the aging population, and thus the resulting THA using CLPE also increased in recent decades [3]. However, several disadvantages of CLPE that are mainly caused by its hydrophobic surface nature must be overcome for its stable and safe application as an artificial joint *in vivo*. First, the hydrophobic surface of CLPE induces nonspecific protein adsorption resulting in undesirable biological responses such as foreign body reactions or inflammatory reactions [4]. A more significant disadvantage of using hydrophobic CLPE in artificial hip-joint is the generation of wear debris induced by a high friction coefficient. Generally, most PEs used in THA are irradiated

by gamma-rays to decrease wear [5]. Nevertheless, it does not completely prevent wear in a constant friction environment. Hydrophobic surface nature is known to accelerate this wear process in an aqueous medium due to the lack of hydrated lubrication effect [6]. Therefore, a large amount of wear debris spreads around the artificial joint surface, and this wear debris is the main cause of osteolysis that results in the loosening of the artificial joint [7,8]. Therefore, a reduction in the wear debris on the artificial hip-joint is an extremely important issue in THA.

The surfaces of a natural synovial joint are covered with articular cartilage that protects the joint surfaces from mechanical frictions [9]. In healthy cartilage, chondrocytes create and preserve the cartilaginous matrix that mainly consists of collagen and proteoglycans [10]. Furthermore, Proteoglycan 4 that is termed lubricin and surface-active phospholipid (SAPL) exists on the surface of articular cartilage as a boundary lubricant to reduce the friction coefficient [11,12]. Due to the electrical charge of these molecules, water molecules are easily trapped on the surface of articular cartilage, and this hydrated layer plays an important role in the smooth motion of the joint [9,13]. In order to overcome the limitations of the artificial hip-joint composed of hydrophobic CLPE, various studies focused on effectively forming the hydration layer on the CLPE surface by mimicking the natural synovial joint. The most

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successful example corresponds to surface grafting of cell-membrane mimicking 2-methacryloyloxyethyl phosphorylcholine (MPC) polymer on a CLPE surface to reduce wear debris [14,15]. Ishihara et al. first invented a poly(MPC) (PMPC)-grafted CLPE surface to form an extremely hydrophilic artificial hip-joint surface [16]. They reported a successful reduction in the friction coefficient, and this was the first report on the usefulness of hydrophilic polymer grafting to prevent wear debris in simulated friction test [17–19]. Although several studies investigated optimizing the lubrication effect of the grafted MPC polymer, significant effort is required to determine various materials that are potentially applicable on the surface of artificial joint from the cost effectiveness viewpoint. The extremely hydrophilic nature of MPC polymer is potentially attributed to its characteristic zwitterionic molecular structure [14,20]. However, only a few studies focused on the effect of surface grafting of other types of zwitterionic materials on reducing friction coefficient on the CLPE surface. Specifically, 2-(Methacryloyloxy)ethyl dimethyl-(3-sulfopropyl)ammonium hydroxide (MEDSAH) is a very well verified zwitterionic materials in terms of its hydrophilic and anti-biofouling properties [21–25]. Furthermore, the synthetic route of MEDSAH is very simple, and the cost effectiveness of the use of MEDSAH is considered as relatively good when compared to other types of zwitterionic materials. Although the contribution of the MPC polymer in this field is overwhelming, it is scientifically valuable to confirm the feasibility of application of other types of zwitterionic polymers, and required conditions for stable use of the alternative zwitterionic polymers to reduce friction coefficient on the CLPE surface.

In the present study, optimal conditions for coating poly(MEDSAH) (PMEDSAH) on CLPE surface were explored to effectively reduce friction coefficient of CLPE in an aqueous medium. The relationship between hydrophilicity, friction coefficient, and the thickness of PMEDSAH on the CLPE surface prepared in different conditions was analyzed to determine the optimal coating conditions. In addition, comparative study of PMPC- and PMEDSAH-grafted surface was conducted to confirm the difference in grafting condition and feasibility of using PMEDSAH to reduce friction coefficient of the CLPE surface in water and simulated body fluids (SBF).

2. Materials and methods

2.1. Materials

In the experiment, 2-(methacryloyloxy)ethyl dimethyl-(3-sulfo propyl)ammonium hydroxide (MEDSAH) monomer and sodium dodecyl sulfate (SDS) were purchased from Sigma–Aldrich (St. Louis, MO, USA). Additionally, 2-methacryloyloxyethyl phosphorylcholine (MPC) was purchased from KCI. CO., Ltd. (Seoul, Korea). Benzophenone was purchased from TOKYO CHEMICAL IND. CO., Ltd. (Tokyo, Japan), and synovial body fluid (SBF) solution was purchased from Biosesang CO., Ltd. (Gyeonggi-Do, Korea). All the organic solvents used in the study were purchased from SAMCHUN PURE CHEMICAL IND. CO., Ltd. (Gyeonggi-Do, Korea) and used as received.

2.2. PMEDSAH and PMPC grafting using free radical polymerization under different conditions

Specifically, CLPE with a gamma-ray level corresponding to 75-kGy according to ASTM F2102-06e1 and ASTM F648-14 standard for cross-linking process was purchased from Orthoplastics Ltd. (Bacup, UK).

The CLPE samples were sequentially sterilized by using an ultrasonic cleaner with 1wt% SDS solution and pure water. Subsequently, they were washed with ethanol and dried *in vacuo* at

room temperature to remove the residual solvent. This was followed by soaking the CLPE samples with acetone solution containing 10 mg/mL benzophenone for 30 s and drying the same *in vacuo* at room temperature to remove the residual acetone. MEDSAH monomer was dissolved in distilled degassed pure water for a 0.25–0.5 mol/L monomer solution, and the CLPE samples coated with benzophenone were soaked in the solution. The samples were then irradiated by a UV curing machine (LZ-UVM-H141AL with a UV transmission filter of 365 nm ± 50 nm, LICHTZEN CO., Ltd., Gyeonggi-Do, Korea) with various intensities in the range of 10–40 mW/cm² at 60 °C for 0.5–2 h to conduct photo-induced grafting polymerization in different conditions. The following three main variables were controlled: UV-irradiation time was varied from 0.5 h to 2 h at a fixed energy (40 mW/cm²) and concentration (0.5 M), UV-irradiation energy was varied from 10 mW/cm² to 40 mW/cm² at a fixed time (1.5 h) and concentration (0.5 M), and monomer concentration was varied from 0.25 M to 0.5 M at a fixed time (1.5 h) and energy (40 mW/cm²). After the polymerization, the PMEDSAH-grafted CLPE samples were sequentially washed with distilled water and ethanol and dried *in vacuo*. Additionally, MPC grafting was performed under the same conditions at fixed time (1.5 h), energy (40 mW/cm²), and concentration (0.5 M) as per the previously reported method [14].

2.3. Transmission electron microscopy (TEM) observation for grafting thickness measurement

Cross-sectional area of CLPE samples prepared by various grafting conditions was observed using TEM. First, each sample was embedded in epoxy resin and stained with ruthenium oxide. Subsequently, the samples were sliced into ultrathin sized (approximately 50–100 nm) discs by using a microtoming machine (Ultracut S, Leica Microsystems, Wetzlar, Germany) for the TEM observation. A TEM microscope (Cryo Tecnai G2, FEI CO., Hillsboro, OR, USA) was used for the observation with an acceleration voltage of 100 kV. The thickness of each PMEDSAH layer was calculated by averaging 20 different points on more than three different TEM images.

2.4. Surface elemental analyses and contact angle measurement

The surface elements of the PMEDSAH-grafted CLPE samples were analyzed by using X-ray photoelectron spectroscopy (XPS) (X-TOOL, ULVAC-PHI, INC, Kanagawa, Japan). More than three different samples were analyzed for the elemental analysis.

The static water contact angles on the PMEDSAH-grafted CLPE and PMPC-grafted CLPE samples were measured by the sessile drop method using a contact angle goniometer (Phoenix 150, Surface Electro Optics, Ltd., Gyeonggi-Do, Korea) with 1 μL distilled water drops on the CLPE surfaces. Measurements were performed on three different points for each sample. The air bubble contact angle was also measured on the sample surface in pure water and SBF. The air bubble contact angle was subtracted from 180 °.

2.5. Friction tests

The dynamic friction coefficient was calculated from the friction force measured by using a tribometer (UMT, CETR, Inc., Campbell, CA, USA). The tribo-test was performed in the reciprocating mode for 100 cycles on the plate sample by using a 9 mm stainless steel ball at a sliding velocity of 2 mm/s. The friction test was performed in pure water or SBF solution at 3 N, the stroke distance was 10 mm, and three areas were evaluated for each sample. The friction coefficient of PMEDSAH-grafted CLPE and PMPC-grafted CLPE surfaces was obtained by averaging the friction force from the 100th cycle data.

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