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Strain and damage-sensing performance of biocompatible smart CNT/UHMWPE nanocomposites

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ABSTRACT

Herein, we report strain- and damage-sensing performance of biocompatible smart CNT/UHMWPE nanocomposites for the first time. CNT/UHMWPE nanocomposites are fabricated by solution mixing followed by compression molding. The surface morphology, microstructural properties, thermal decomposition and stability, glass transition temperature and thermal conductivity of the nanocomposites are characterized. The degree of crystallinity of CNT/UHMWPE nanocomposites is found to have a maximum value of 52% at 0.1 wt% CNT loading. The degree of crystallinity influences the mechanical properties of the CNT/UHMWPE nanocomposites. The electrical percolation threshold is achieved at 0.05 wt% of CNT and it follows a two dimensional conductive network according to percolation theory. The piezoresistive response of CNT/UHMWPE nanocomposites is demonstrated with a gauge factor of ~2.0 in linear elastic regime and that in the range of 3.8–96.0 in inelastic regimes for 0.05 wt% of CNT loading. A simple theoretical model is also developed to predict the resistivity evolution in both elastic and inelastic regimes. High sensitivity of CNT/UHMWPE nanocomposites coupled with linear piezoresistive response up to 100% strain demonstrates their potential for application in artificial implants as a self-sensing material.

1. Introduction

Total joint replacement (TJR) is a surgical procedure in which the joint damaged as a consequence of diseases such as osteoarthritis or post-traumatic arthritis is replaced by an artificial implant [1]. TJR represents the largest segment within orthopaedic surgery, with over 2 million procedures performed globally each year [2–4]. However, failure of the surgery in the short and long term, necessitating revision, is not uncommon. Revision surgeries pose a significant risk to patient health, and place an enormous economic stress on the healthcare system. While survivorship of joint replacements are generally good through the first decade of implantation (90–95% survivorship at 10-15 years [5, 6]), these procedures are increasingly being performed in younger patients, which imposes significantly greater demand on the replaced joint. Therefore, there continues to be a significant need to improve long-term success of these procedures, which is related to the complex interplay between surgical technique, patient characteristics, and implant design.

To improve long term outcomes we need to: (1) understand how these factors affect implant performance within the body; (2) detect early signs of failure to enable early intervention, and (3) develop improved implant materials. Currently we have no way of measuring device performance once it is implanted in the patient. Ultra-high-molecular-weight-polyethylene (UHMWPE) continues to be the primary bearing material used in TJR implants, due to its cost-efficiency [7], excellent mechanical, tribological and chemical properties such as high impact strength, abrasion resistance, low friction coefficient, chemical inertness, and biocompatibility [8, 9]. Substantial advances have been made in improving wear performance of UHMWPE through radiation cross-linking coupled with stabilization against oxidative damage via melting, annealing and addition of antioxidants such as vitamin-E. However, while radiation cross-linking dramatically enhances wear performance, it has the disadvantage of reducing the mechanical strength and fatigue resistance of UHMWPE. Consequently as we look to enhance the life of TJR devices into the second and third decade of in vivo use, continual improvement in mechanical performance of

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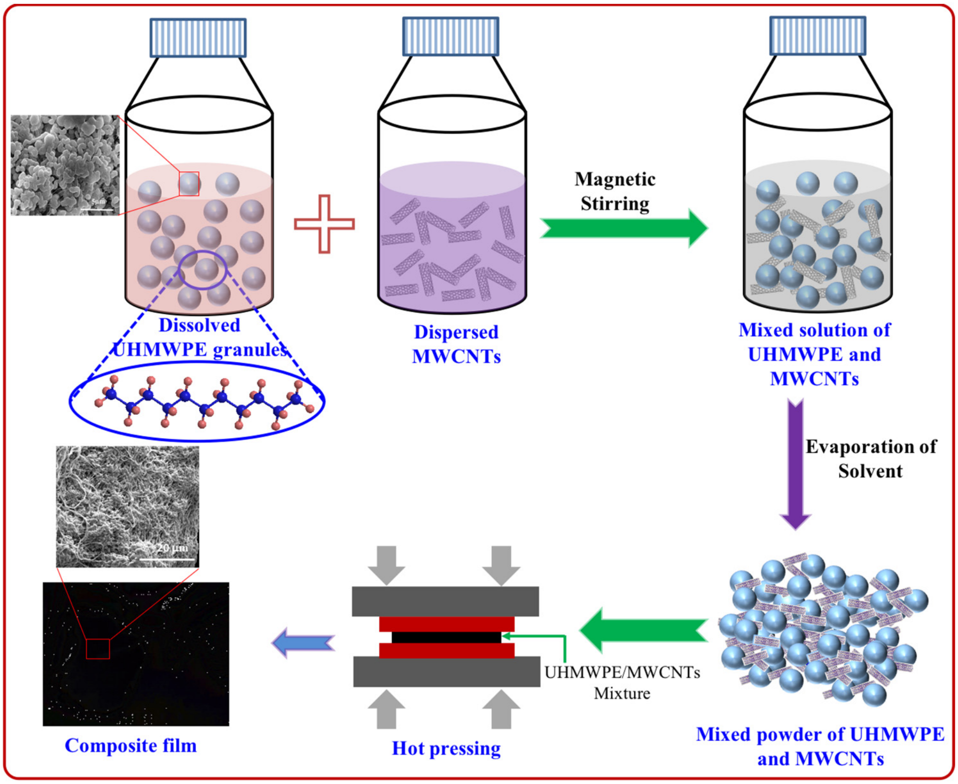

Fig. 1. Schematic of the CNT/UHMWPE fabrication process.

UHMWPE and the ability to measure in vivo device performance is required [10].

With the advent of nanocomposites [11–16], we now have a way to modify the implant material, to not only improve existing properties but also impart new ones. The addition of nano-fillers could be used to create orthopaedic biomaterials that can sense in vivo environment and measure in vivo device performance, without need for incorporating separate sensors into orthopaedic implants. At the same time, these nano-fillers could improve fracture toughness and fatigue resistance of current high-crosslinked UHMPWE materials. Amongst the various nano-fillers evaluated by researchers to be used for polymer composites [17], carbon nanotubes (CNTs) are particularly a promising carbon-based nano-fillers owing to their excellent mechanical and transport properties [18–21]. Although few researchers have explored the use of CNTs in UHMWPE, these studies indicate potential for substantial improvement not only in Young's modulus and yield strength, but also fracture toughness of the composite [22]. Martinez et al. [23] found fracture toughness of neat UHMWPE to reduce by 40% following 90 kGy irradiation [24]. In contrast, fracture toughness remained constant for 1-5 wt% CNT/UHMWPE composites under same irradiation dose [25]. Sreekanth et al. [26] used chemically functionalized CNTs and found the improvement in fracture strain and toughness of 70% and 176% respectively for 2.0 wt% CNT/UHMWPE composite relative to neat UHMWPE [27]. However, properties of CNT/UHMWPE composites are highly sensitive to synthesis parameters, and there is paucity of research in this area [28]. Therefore, it is unclear what the optimal process parameters are, and if the positive findings of the above studies are replicable [29].

The inclusion of CNTs in polymer matrix also leads to the modification of electrical resistivity, which changes as a function of applied mechanical stress/strain (piezoresistive response) [30]. This property could be used for self-sensing applications. For example, Mohiuddin et al. [31] showed that electrical resistance of CNT-PEEK composites changes as a function of applied pressure, with greater sensitivity for lower CNT concentration. With the exception of one recent study by Do et al. the self-sensing response of UHMWPE nanocomposites has not been reported. Do et al. reported the piezoresistive behavior of MWCNT/UHMWPE composite at 0.3 and 0.5 wt% of MWCNT loading under quasi-static compressive loading [32]. Therefore, the goal of the present work is to conduct detailed investigation of the mechanical and piezoresistive behaviour of UHMWPE nanocomposites at low CNT loadings.

2. Materials & methods

The materials used in the present work included multi-walled carbon nanotubes (MWCNTs) and ultrahigh molecular weight polyethylene (UHMWPE). The MWCNTs were purchased from Applied Nanostructured Solutions, LLC. CNTs have an average diameter of 30 nm, length of a few hundred microns and aspect ratio > 3000. The polymer matrix used in this study was medical grade GUR 1020 UHMWPE powder procured from Celanese, USA with density of 935 kg/m^3 and molecular weight of 3.6×10^6 g/mol.

UHMWPE nanocomposites with different wt% of CNT loadings were fabricated via solution mixing followed by compression molding technique [33]. These nanocomposites were prepared in two consecutive steps. Initially, the CNTs were added to ethanol and then dispersion was achieved by intense ultra-sonication at 20% amplitude for 2 h [34]. UHMWPE powder was poured into ethanol separately and magnetically stirred for two hours at a speed of 400 rpm at temperature 110 °C. The stable CNT/ethanol suspension was then transferred into the UHMWPE/ethanol solution and the final solution was stirred at the same speed of 400 rpm and at about 110 °C to completely evaporate the solvent. Finally, the dry powders of different wt% of CNT loaded UHMWPE were compression molded using a hot press at an operational force of about 500 N for 15 min, at 145 °C, to obtain a rectangular film of approximately 1 mm thickness. In the same way, neat UHMWPE sample was also prepared for comparison. The schematic of the fabrication process is shown in Fig. 1.

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