Improving Fatigue and Fracture Resistance of 45S5 Bioactive Glass/Polyurethane Biocomposites for Repairing Bone Defects

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Statement of Purpose: Injectable, settable bone grafts that possess dynamic mechanical strength exceeding that of host bone and maintain mechanical competence comparable to bone while remodeling could improve the clinical management of a number of orthopaedic conditions, such as repair of open tibial plateau fractures, screw augmentation, and vertebroplasty. Injectable polyurethane (PUR) biocomposites are an attractive alternative to calcium phosphate cements due to their tough mechanical properties and ability to facilitate faster remodeling [1]. 45S5 bioactive glass (BG) has widely been used for bone regeneration purposes due to its osteoconductivity and bioactivity. Because of its rigid brittle nature, it is best delivered as a composite[2]. Although physiological loads are generally cyclic, fatigue and corresponding fracture toughness properties of biomaterials utilized in load-bearing applications are rarely reported. In this study, we compared the dynamic compressive and fracture toughness properties of unmodified and surface-modified BG/PUR biocomposites [3] to those of a commercially available bone cement (calcium sulfate and phosphate, CaP cement). We hypothesized that a low-porosity BG/PUR biocomposite incorporating surface-modified BG would possess a longer fatigue life at physiologically relevant maximum stress levels compared to the CaP cement.

Methods: Prior to reaction with the PUR binder, BG particles were functionalized with the silane-coupling agent 3-aminopropyl-triethoxysilane followed by surface grafting of polycaprolactone (PCL). Biocomposites were prepared from a lysine triisocyanate– poly(ethylene glycol) prepolymer, triethylene diamine catalyst, polyester triol (70% caprolactone, 20% glycolide, 10% lactide polyol, Mn ~300 g mol^{-1}), and BG (56.7 volume %). The biocomposites and bone cement were injected into molds to generate cylindrical dowels (6 mm diameter X 12 mm length) and rectangular beam specimens (ASTM E1820) for fatigue and fracture toughness testing, respectively. Specimens for all testing were pre-conditioned in phosphate buffer solution for 24 h. Cyclic loading of dowels was performed using sinusoidal waveform in force control at a frequency of 5 Hz generating physiologically relevant maximum compressive stress levels ranging from 5 MPa to 30 MPa (minimum stress was near zero for all levels). Throughout the entire testing period, specimens were hydrated with water via a drip-system and strain was tracked using an extensometer. The fatigue life (N_f) was defined as the number of cycles achieved before mechanical failure in which failure was independently defined as: 1) 10% decrease in secant modulus, 2) 1% creep, or 3) 3% maximum displacement. For fracture toughness testing, a single edge-notched beam was loaded in three-point bending using a resistance-curve (r-curve) approach (ASTM E1820).

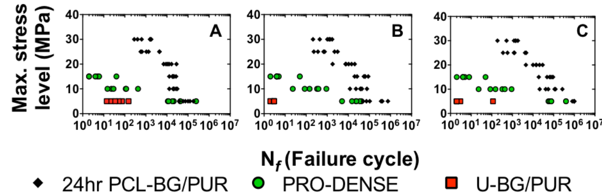

Figure 1. S-N curves for compressive fatigue testing of unmodified (U-BG/PUR) and surface-modified BG (24hr PCL-BG/PUR) polyurethane biocomposites, and bone cement (PRO-DENSE). Mechanical failure: A) modulus, B) creep, and C) displacement.

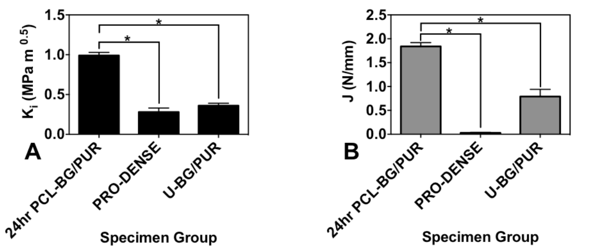

Figure 2. Fracture toughness testing parameter, A) plane-strain fracture toughness and B) J-integral, results for U-BG/PUR and 24hr PCL-BG/PUR biocomposites, and bone cement (PRO-DENSE®).

Results: At a maximum stress level of 5 MPa, fatigue life was similar between surface-modified BG composites and the CaP cement when failure was defined as a 10% loss in modulus (Fig. 1A), but the composite had a higher fatigue life than CaP cement for displacement and creep based failures (Fig. 1B-C). The difference in mean fatigue life (displacement failure) between these biomaterials increased as the maximum stress level increased. Surface-modified BG composite had a longer fatigue life (102,666 ± 88,917 cycles and 62,833 ± 26,581 cycles) vs. CaP cement (321 ± 326 and 12 ± 19) at 10 MPa and 15 MPa, respectively. PUR composite with unmodified BG had inferior fatigue life such that specimens could not resist compressive stress levels above 5 MPa. The surface-modified BG/PUR biocomposite also showed significantly higher crack initiation toughness (K_i) and non-linear strain energy dissipation during crack growth (J-integral) than both the U-BG/PUR biocomposite and CaP cement (Fig 2A-B).

Conclusions: Surface-modified BG/polyurethane biocomposite displayed significantly longer fatigue life for each of the three definitions of failure, based on modulus, creep, and displacement, and at each comparative maximum stress level tested. Additionally, it was able to withstand higher cyclic stress loads than the CaP. Lastly, the surface-modified BG/PUR composites showed significantly superior fracture toughness properties testing, which in part may explain the longer compressive fatigue life.

References: [1]J.E Dumas, et al (2010) Acta Biomaterialia 6:2394-2406. [2] A.R Boccaccini, et al (2005) Expert Rev Med Devic 2:303-317. [3]A.J Harmata et al (2014) J Mater Res. 254-64.