ScienceDirect

Journal of the Mechanical Behavior of Biomedical Materials

Volume 54, February 2016, Pages 268-282

Research Paper

Compressive cyclic ratcheting and fatigue of synthetic, soft biomedical polymers in solution

Andrew T. Miller [a], David L. Safranski [d], Kathryn E. Smith [d], Robert E. Guldberg [a b], Ken Gall [c d]

Show more

Share Cite

https://doi.org/10.1016/j.jmbbm.2015.09.034

Get rights and content

Highlights

- We tested several soft polymers in cyclic compression in saline to study behavior.
- DSC and DMA were used to explain the cyclic loading behavior.
- Polymers devoid of dissipation, as shown by tan delta, exhibited fatigue fracture.
- Polymers with dissipation remained largely undamaged but showed cyclic ratcheting.
- Some behave similar to meniscus tissue, suggesting potential as artificial tissue.

Abstract

The use of soft, synthetic materials for the replacement of soft, load-bearing tissues has been largely unsuccessful due to a lack of materials with sufficient fatigue and wear properties, as well as a lack of fundamental understanding on the relationship between material structure and behavior under cyclic loads. In this study, we investigated the response of several soft, biomedical polymers to cyclic compressive stresses under aqueous conditions and utilized dynamic mechanical analysis and differential scanning calorimetry to evaluate the role of thermo-mechanical transitions on such behavior. Studied materials include: polycarbonate urethane, polydimethylsiloxane, four acrylate copolymers with systematically varied thermo-mechanical transitions, as well as bovine meniscal tissue for comparison. Materials showed compressive moduli between 2.3 and 1900 MPa, with polycarbonate urethane (27.3 MPa) matching closest to meniscal tissue (37.0 MPa), and also demonstrated a variety of thermo-mechanical transition behaviors. Cyclic testing resulted in distinct fatigue-life curves, with failure defined as either classic fatigue fracture or a defined increased in maximum strain due to ratcheting. Our study found that polymers with sufficient dissipation mechanisms at the testing temperature, as evidenced by tan delta values, were generally tougher than those with less dissipation and exhibited ratcheting rather than fatigue fracture much like meniscal tissue. Strain recovery tests indicated that, for some toughened polymers, the residual strain following our cyclic loading protocol could be fully recovered. The similarity in ratcheting behavior, and lack of fatigue fracture, between the meniscal tissue and toughened polymers indicates that such polymers may have potential as artificial soft tissue.

Introduction

Within the biomedical industry, there has been considerable interest for decades in developing suitable synthetic replacements for soft load-bearing tissues such as intervertebral disc, meniscus, and articular cartilage. These tissues are particularly prone to degeneration as an individual ages which can lead to significant impairment and morbidity (Urban and Roberts, 2003, An et al., 2004, Luoma et al., 2000, Hugle et al., 2012, Bieleman et al., 2010). Unfortunately, current treatment options such as spinal fusion, total disc replacement, and total knee replacement are associated with a host of complications and negative side effects which are often attributed to the altered biomechanics of the joint due to a mismatch in mechanical properties between the implant materials and the tissues they replace (Han et al., 2013, Noble et al., 2005). For example, the loss of a flexible joint following spinal fusion can lead to adjacent-segment degeneration in a significant percentage of cases (Hilibrand et al., 1999). Total disc replacement has been proposed as an alternative; however, the unnatural motion of current devices, as well as wear debris, is known to lead to facet joint degeneration and even heterotopic ossification, causing fusion of the joint (André van et al., 2003, Christoph, 2006, Punt et al., 2008). Likewise, total knee replacement has issues which can also be in part attributed to implant materials, including: bone resorption due to stress shielding effects, osteolysis due to bearing surface wear particles, and an overall functional deficit (Noble et al., 2005, Agarwal et al., 2014, Au et al., 2007). For applications such as these, it seems a potentially superior approach could be developed in which joint properties and mechanics are restored utilizing a softer material which more closely mimics the natural tissue properties. In general, the development of a durable, soft material of this type would have many applications in the field of soft tissue replacement. Due to the load-bearing nature of many of the soft tissues in the body, an important consideration when developing replacement materials is the response to cyclic loading and fatigue behavior. Not surprisingly, such behavior of current common implant materials such as cobalt-chrome, stainless steel, titanium alloys, and ultra-high molecular weight polyethylene (UHMWPE) have been extensively studied. This includes the effects of various microstructures, surface treatments, and fabrications methods, as well as the effects of a physiological environment (Antunes and de Oliveira, 2012, Cahoon and Holte, 1981, Dharme et al., 2013, Fleck and Eifler, 2010, Gencur et al., 2006, Kurtz et al., 1998, Niinomi, 1998, Niinomi et al., 2001, Papakyriacou et al., 2000, Pruitt, 2005, Sauer et al., 1996, Semlitsch and Willert, 1980, Simis et al., 2006, Taira and Lautenschlager, 1992, Teoh, 2000).

There have been significantly fewer clinically successful devices based on soft materials. For example, as recently as the late 90s, silicone was touted as a potentially suitable material for the replacement of arthritic joints (Swanson and Peltier, 1997). However numerous studies since have documented cases of failed implants when silicone is used in hand, foot, wrist, and cardiac lead applications. The failure of these implants is generally attributed to fracture, compressive deformation, and wear leading to inflammation, arthritis, foreign body response, and loss of function (Chan et al., 1998, De Lurgio et al., 1997, Trepman and Ewald, 1991, Vanderwilde et al., 1994, Rosenthal et al., 1983). Similar issues with silicone were encountered in the development of the Acroflex lumbar disc replacement. Mechanical failure of the silicone core was observed six months after implantation, ending the clinical trial and prompting a return to the original design utilizing a polyolefin rubber core. Despite the promising results from preliminary fatigue and biocompatibility testing, clinical trials were ultimately stopped with results including: mechanical failure of the polyolefin rubber core, osteolysis attributed to the presence of wear particles, as well as periannular ossification (Cunningham et al., 2001, Fraser et al., 2004, Meir et al., 2013, Serhan et al., 2002). Learning from the issues that plagued prior compliant devices, a newer generation of soft orthopedic devices has emerged recently utilizing polycarbonate urethane, including the LP-ESP total disc replacement. The LP-ESP utilizes an outer annulus of polycarbonate urethane surrounding an inner core of silicone with microvoids, and preclinical tests demonstrated promising results for the device in terms of fatigue, wear, and biostability. Since 2005, the device has been used in over 2000 cases in Europe, and literature has yet to report a complication due to material wear or fatigue (Lazennec et al., 2013a, Lazennec et al., 2013b, Lazennec et al., 2013c, Lazennec, 2011, Lazennec et al., 2014). In addition to the LP-ESP, promising results have also been seen with similar polycarbonate urethane based devices including: Total Posterior Spine (Anekstein et al., 2015, McAfee et al., 2007), Bryan Cervical Disc (Cao et al., 2015, Quan et al., 2011, Rao et al., 2015), M6 cervical and lumbar disks (Lauryssen et al., 2012, Reyes-Sanchez et al., 2010, Thomas et al., 2013), Freedom Lumbar Disc (Rischke et al., 2011), acetabular cups (Scholes et al., 2006, Kurtz, 2008, Smith et al., 2000, John and Gupta, 2012, Elsner et al., 2011, Elsner et al., 2010), meniscal implant (Elsner et al., 2012, Shemesh, 2014), a bearing surface in osteochondral implants (Cook et al., 2014), as well as heart valve replacements (Daebritz et al., 2004, Daebritz et al., 2003). While short-term trials for these devices have been relatively successful, long-term results are largely missing as the push for the use of polycarbonate urethane in orthopedic devices has been a rather recent development (Treharne and Greene, 2008, John, 2014). Besides the preclinical testing of devices, little work has been done to characterize and understand the fundamental fatigue responses of synthetic, soft polymers in a physiological environment. Careful examination of the response to cyclic loading across multiple types of soft materials could provide valuable insights into the design and development of such materials for orthopedic applications.

When it comes to predicting fatigue performance of engineered components, the approaches typically used for metals and stiff polymers (stress-life, linear elastic fracture mechanics) are generally not always applicable for soft or elastomeric components. The discrepancy is due primarily to the nonlinear behavior and relatively large strains that most elastomeric materials exhibit. Fatigue approaches for elastomeric materials typically rely on strain and strain energy functions (Mars and Fatemi, 2002, Papadopoulos, 2006, Thomas, 1994, Zarrin-Ghalami and Fatemi, 2012). Further complicating the matter is the fact that the fatigue performance of elastomers, and most polymers in general, is especially susceptible to common factors well studied in metals such as load history, *R*-ratio, strain rate, geometry, frequency, temperature, material formulation, and environment (Zarrin-Ghalami and Fatemi, 2012, Legorju-Jago and Bathias, 2002, Mars and Fatemi, 2004). There has been a fair amount of work on the fatigue of elastomers and rubbers with most focused on natural rubbers for the tire industry. Effects such as minimum stress, strain crystallization, variable amplitudes, molecular weight, fillers, additives, and temperature have been studied with some papers proposing fatigue life models and investigating damage mechanisms (Fitzgerald et al., 1992, Fitzgerald et al., 1998, Flamm et al., 2011, Rodriguez et al., 1994, Harbour et al., 2008, Lemieux and Killgoar, 1984, Mars and Fatemi, 2006, Alshuth et al., 2002). While there has been considerable work on the fatigue of elastomers and rubbers in general, biocompatible elastomers tested in physiological conditions represent only a small subset. This is an important distinction when one considers the sensitivity of polymers to their environment. Currently known soft polymers suitable for biomedical applications generally fit into one of a few categories: polyurethanes, silicones, polyolefin rubbers, acrylates, and others (non-polyurethane thermoplastic elastomers, etc.) (McMillin, 2006, Yoda, 1998, Smith et al., 2009a). A review of the literature on the fatigue and high-cycle loading response of these polymers, in physiological conditions, turns up a relatively small body of scientific work.

Research on polyurethanes includes that of Chaffin and colleagues who tested various mechanical properties of two commercial PDMS and polyether urethanes, often used in pacemaker and defibrillator leads, after accelerated water exposure. They found a decrease in the molar mass related to the length of time spent in solution and subsequently reduced ultimate tensile strength, toughness, wear, and fatigue resistance properties. The molar mass reduction was attributed to hydrolytic cleavage of the polymer backbone, illustrating the dramatic effect a physiological environment can have on elastomers (Chaffin et al., 2012, Chaffin et al., 2013). In fact, the development of polyurethane materials for biomedical applications is testament to this effect. Initially polyester urethanes were used until it was discovered that the polyester segment was susceptible to substantial hydrolytic degradation *in vivo*. Polyether urethanes (PEUs) were then developed to improve upon the hydrolytic degradation, but were ultimately found to be susceptible to oxidation *in vivo*. Finally, polycarbonate urethanes and PDMS containing PEUs were developed that have proven superior to previous polyurethanes in terms of hydrolytic and oxidative resistance; however, both are still somewhat susceptible to *in vivo* degradation (Treharne and Greene, 2008, Chaffin et al., 2012, Chaffin et al., 2013, Mathur et al., 1997, Cipriani et al., 2013). Based on work from Wiggins et al, who examined the effect of strain rate and polyurethane soft segment chemistry on *in vitro* fatigue resistance of film type specimens, these degradative effects are exacerbated by mechanical loading (Wiggins et al., 2004, Wiggins et al., 2003, Wiggins et al., 2003). In addition to the studies above, several specialized and accelerated tests on polyurethanes, as well as polyolefin rubbers, have been conducted in physiological conditions for cardiovascular and blood pumping applications (Hayashi et al., 1984, Hayashi et al., 1985, Sevastianov and Parfeev, 1987, Hayashi, 1994, McMillin, 1983, Mcmillin, 1987, Murayama and McMillin, 1983, McKenna and Penn, 1980, Takahara et al., 1985).

In parallel with polyurethanes, cyclic testing in physiological conditions has also been investigated on a few non-polyurethane thermoplastic elastomers (NPUTPEs). The most notable is a polystyrene–polyisobutylene–polystyrene block copolymer often referred to as SIBS. SIBS, which resembles silicone, has proven to be an exceptionally biocompatible and biostable material and is often placed between silicone and polyurethane in terms of stiffness and tensile strength. The fatigue properties of SIBS and a few other NPUTPEs have been investigated using the so-called hysteresis method recently pioneered for polymers by

Renz, Ehrenstein, and Altstädt. The method involves tracking changes in hysteresis loops during cyclic loading which reveals changes in dynamic modulus and material damping. It is proposed that these parameters reveal more about irreversible structural changes and damage than the conventional fatigue testing methods. Using this method various compositions and architectures of SIBS and other NPUTPEs have been tested, in air as well as in physiological conditions, alongside polyurethane, polyester, and silicone. Their work provides important insights into the relationship between chemical structure, network architecture, chain interactions, hard and soft segment compositions and the resultant cyclic responses. While the results for SIBS seem promising, often outperforming silicone in terms of fatigue resistance, these block copolymers typically exhibit extensive permanent creep under cyclic load attributed to its' thermolabile physical crosslinks (El Fray et al., 2006a, El Fray et al., 2006b, Puskas and Chen, 2004, Puskas et al., 2009a, Puskas et al., 2009b, El Fray and Altstädt, 2003a, El Fray and Altstädt, 2003b, Fray and Altstädt, 2004, Götz et al., 2012, Götz et al., 2009, Pinchuk et al., 2008, Pavka, 2013). However, recent work by the same group has shown that carbon black reinforcing of dendritic polyisobutylene-based block copolymers not only improves tensile performance but also results in a reduction of cyclic creep to levels on par with silicone (Pavka, 2013, Götz et al., 2014). While the work is still ongoing, the results so far have provided interesting insights into the fundamental differences in fatigue responses between several materials.

In addition to the research on elastomers for cardiovascular applications, one other study was found which investigated the fatigue of a polyolefin rubber in physiological conditions for dental and orthopedic applications (Helsen et al., 1993). To the best of the authors′ knowledge, the previously mentioned papers represent the bulk of the literature regarding non-device fatigue and high-cycle testing of soft polymers in physiological conditions. Furthermore, within this body of work, the number of papers that sought to develop fundamental knowledge about the correlation between material chemistry, structure, etc. and cyclic loading or fatigue response represents an even smaller subset. As mentioned previously, acrylates have also been touted as potentially suitable polymers for the replacement of soft bodily tissues. While fatigue data in physiological conditions could not be found, their toughness in relation to their chemistry and structure have been well documented in such conditions (Smith et al., 2009a, Smith et al., 2011, Smith et al., 2009b, Safranski et al., 2011, Safranski et al., 2014, Safranski and Gall, 2008). In addition, their easily tailored nature makes them an attractive material for the investigation of fatigue properties in relation to polymer structure and thermo-mechanical transitions.

Knowing the susceptibility of polymers to factors such as environment, geometry, and *R*-ratio, it comes as no surprise that many of the fatigue testing standards for elastomers state that standardized test results do not provide correlation with service performance (for example, ISO 4666-1:2010 and ASTM D4482-11). It seems likely then that the preference towards device testing versus standardized elastomer testing is due in part to the difficulties in translating such standardized data to useful predictions for engineered components. Regardless, the testing of a broad range of soft polymers still allows for relative comparisons and the development of fundamental knowledge useful for materials optimization in fatigue-prone applications. In addition, as was seen in the case of the Acroflex lumbar disc, even seemingly conservative simulated fatigue tests of an actual device are imperfect as the Acroflex still failed *in vivo*. While the relatively new polycarbonate urethane based devices have clinical promise, the implantation history of elastomeric devices indicates that there is a clear need for fundamental knowledge of the fatigue properties of elastomers in physiological conditions. As such, the objectives of this paper are threefold:

1. Examine the fatigue properties and responses to cyclic loading of a range of synthetic polymers with systematically varied thermo-mechanical transitions to develop an understanding of the relationship between these transitions and responses.

2. Understand the response to cyclic loading (creep, recovery, damage accumulation) for the most promising materials from objective 1.

3. Compare the behavior of the synthetic polymers with soft biological tissues.

It is our hypothesis that the thermo-mechanical transition behaviors, which are indicative of various aspects of polymer structure and fundamental to classifying various polymer systems, will have implications on the fatigue performance of these materials. Through these objectives we hope to develop a better understanding of the link between polymer structure and response to cyclic compressive loading.

Section snippets

Materials

PDMS samples were created using the Sylgard 184 elastomer kit (Dow Corning). Methyl methacrylate (MMA), methyl acrylate (MA), butyl acrylate (BA), and 2,2-dimethoxy 2-phenylacetophenone (DMPA) were obtained from Sigma-Aldrich, and used as received. 1,12-dodecanediol dimethacrylate (DDDA) was obtained from Monomer–Polymer and Dajac Labs. Carbothane AC-4085A was obtained from Lubrizol. Bovine meniscus samples were obtained through Animal Technologies, Inc. (Tyler, Texas) and kept frozen until use.…

PBS absorption

The water content is reported for all synthetic materials in Fig. 1. With the exception of PCU, results indicated that the water content for all materials stabilizes within 24h of being placed in PBS. The acrylates showed a small increase in water content as the proportion of methyl acrylate increases relative to methyl methacrylate.…

Differential scanning calorimetry

Fig. 2 shows representative DSC curves for the tested materials, Table 1 shows tabulated glass transition temperatures for each material calculated from both DSC as …

Discussion

The fatigue of materials is a complex process that differs at macroscopic and microscopic levels depending on material type, loading profile, and environmental conditions. Native materials in the human body are subjected to continual cyclic stresses and depending on the function of the material and the level of the stresses, the human body can sometimes counteract cyclic loading and fatigue. Tissues such as bone and muscle are known to be very effective at repairing fatigue damage (Burr et al., …

Conclusions

We have investigated the fatigue properties and general response to cyclic loading of a range of soft, synthetic polymers with systematically varied thermo-mechanical transformation behavior. The following are the conclusions of the work.

1. Classic fatigue fracture under cyclic loading occurs when the polymer is devoid of a dissipation mechanism at the test temperature. Fatigue fracture was experienced by PDMS and 95BA, both of which have little dissipation on their thermo-mechanical transition…

…

Recommended articles

References (146)

A.G. Au
Contribution of loading conditions and material properties to stress shielding near the tibial component of total knee replacements
J. Biomech. (2007)

R.A. Antunes *et al.*
Corrosion fatigue of biomedical metallic alloys: mechanisms and mitigation
Acta Biomater. (2012)

D.B. Burr
Bone remodeling in response to in vivo fatigue microdamage
J. Biomech. (1985)

V. Bentolila
Intracortical remodeling in adult rat long bones after fatigue loading
Bone (1998)

K.A. Chaffin
Abrasion and fatigue resistance of PDMS containing multiblock polyurethanes after accelerated water exposure at elevated temperature
Biomaterials (2013)

E. Cipriani
In-vivo degradation of poly (carbonate–urethane) based spine implants
Polym. Degrad. Stab. (2013)

J.L. Coleman
Diurnal variations in articular cartilage thickness and strain in the human knee

J. Biomech. (2013)

D.B. De Lurgio
Incidence and implications of abrasion of implantable cardioverter-defibrillator leads
Am. J. Cardiol. (1997)

J.J. Elsner
Wear rate evaluation of a novel polycarbonate-urethane cushion form bearing for artificial hip joints
Acta Biomater. (2010)

M. El Fray *et al.*
Fatigue behaviour of multiblock thermoplastic elastomers. 1. Stepwise increasing load testing of poly(aliphatic/aromatic-ester) copolymers
Polymer (2003)

View more references

Cited by (17)

Use of a novel magnetically actuated compression system to study the temporal dynamics of axial and lateral strain in human osteochondral plugs
2024, Journal of Biomechanics

Show abstract

Lumbar Disk Arthroplasty for Degenerative Disk Disease: Literature Review
2018, World Neurosurgery

> *Citation Excerpt :*
>
> …The LP-ESP, or lumbar prosthetic elastic spine pad, was developed by FH Orthopedics and approved for clinical use in 2005.81 It differs from first-generation devices in its application of a polycarbonate urethane (PCU) annulus, designed to progressively resist motion passing through it, and reduce forces on facet joints, which surrounds a silicone rubber core with microvoids, which compress to mimic the shock absorption of a normal nucleus pulposus.82 It acts as a 1-piece viscoelastic disk, which provides 6 full degrees of motion with elastic return and has an adaptive physiologic centre of rotation similar to a natural disk—a design hoped to solve issues previously faced in articulation between the elastic component and endplates of first-generation devices.83…

Show abstract

3D porous polyurethanes featured by different mechanical properties: Characterization and interaction with skeletal muscle cells
2017, Journal of the Mechanical Behavior of Biomedical Materials

> *Citation Excerpt :*
>
> …Among them, silicones such as polydimethylsiloxane (PDMS), possess an excellent biostability and highly tunable elasticity. However, these materials have been mostly used for the fabrication of 2D films (Sun et al., 2013), and are featured by drawbacks such as a small resistance to fatigue (Miller et al., 2016). Another class of synthetic materials which deserves to be mentioned is the polyester-based one.…

Show abstract

Deformation and fatigue of tough 3D printed elastomer scaffolds processed by fused deposition modeling and continuous liquid interface production

2017, Journal of the Mechanical Behavior of Biomedical Materials

> *Citation Excerpt :*
>
> …In our previous work, we examined the toughness and fatigue performance of a particular PU, polycarbonate urethane (PCU), as a function of both hard segment content and processing via injection molding or 3D printing (fused deposition modeling). The material demonstrated considerable toughness in both compressive and tensile fatigue loading (Miller et al., 2016, 2017). In addition, solid 3D printed samples matched or exceeded the performance of injection molded counterparts in terms of tensile and compressive monotonic properties, shear strength, and tensile fatigue performance (Miller, 2017).…

Show abstract

Fatigue life of bovine meniscus under longitudinal and transverse tensile loading

2017, Journal of the Mechanical Behavior of Biomedical Materials

> *Citation Excerpt :*
>
> …An obstacle in the development of functional meniscus implants has been fatigue failure during long duration animal studies (Kelly et al., 2007; Hannink et al., 2011). To improve fatigue properties, researchers have begun to evaluate fatigue life of synthetic soft-tissue analogs, typically under compressive loading (Miller et al., 2016; Shemesh et al., 2014). The present study will allow research groups to directly compare the tensile fatigue properties of their replacement devices to the tensile fatigue properties of native meniscus.…

Show abstract

Fatigue of injection molded and 3D printed polycarbonate urethane in solution

2017, Polymer

> *Citation Excerpt :*
>
> …PCU has gained traction due to its relative biocompatibility and biostability as well as its stiffness and viscoelastic properties, which closely match that of many load-bearing, soft tissues. In addition, the material has proven to be durable through preclinical testing of many of the previously mentioned devices [11–28]. Despite this, caution is warranted as the long-term clinical results of PCU devices have yet to be seen.…

Show abstract

View all citing articles on Scopus ↗

View full text