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Mechanical and tribological properties of a novel hydrogel composite reinforced by three-dimensional woven textiles as a functional synthetic cartilage

Mohammadreza Arjmandi[a,*], Maziar Ramezani[a], Tim Bolle[b], Gesine Köppe[b], Thomas Gries[b], Thomas Neitzert[a]

[a] *Department of Mechanical Engineering, Auckland University of Technology, Auckland 1010, New Zealand*
[b] *Institute für Textiltechnik, RWTH Aachen University, Aachen 52074, Germany*

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ABSTRACT

Hydrogels have been studied extensively as a potential cartilage replacement candidate as they are biocompatible, and can mimic the lubrication mechanisms found in cartilage tissue. As for the mechanical properties, there is still room for improvement. In the current research, two different three-dimensional weave patterns were developed and produced out of biocompatible polymers to reinforce the hydrogel matrix. Inspired by the articular cartilage tissue, the woven preforms featured a through-the-thickness stiffness gradient, and could resist delamination. Experiments were conducted on the hydrogel composites. Infiltration of hydrogel into the woven fabric led to a decrease in surface roughness when compared to neat textiles, and an increase in load-to-failure capacity. The wear rate and friction coefficient of the reinforced hydrogel matrix were greatly reduced under the range of applied loads and sliding velocities. These promising results are attributed to the synergistic interaction between the fiber phase and the hydrogel matrix.

1. Introduction

In the past decades, the advent of orthopedic prostheses and their widespread applications have helped millions of patients worldwide to be relieved from pain and gain their mobility. Despite the great advancements in design and manufacturing of such implants, knee and hip prostheses are still not suitable for young or middle-aged patients, suffering from localized cartilage damage. This is due to the limited life span of these load bearing prostheses which might lead to more invasive revision surgeries. All available remedies for those patients, such as medication, marrow stimulation and cartilage transplantation are temporary and might result in a regenerated tissue with different properties to the existing one, hence limited durability [1,2]. Therefore, an alternative way should be investigated to prevent further tissue degeneration through replacing damaged regions of the tissue, hence preserving the remaining healthy portion, prolonging the tissue functionality, and further postponing the total joint replacement. As a potential candidate, three-dimensional (3D) woven textiles, infiltrated with hydrogels were suggested as a scaffold in tissue engineering of cartilage reconstruction in the past [3,4]. The rationale was to benefit from both energy-dissipative hybrid hydrogel and the rigid, yet flexible fiber reinforcement network providing strength, similar to fibrous tissues [5,6]. Relatively good integration of these structures with the neighboring cartilage tissue arises from the close resemblance in terms of mechanical characteristics the woven textiles could offer, including anisotropy, inhomogeneity and viscoelasticity [3]. Furthermore, with the incorporation of the 3D textiles in the implant structure, fixation and anchorage is feasible as the woven form of a biomaterial promoting bone ingrowth, such as Ti6Al4V [7], could be utilized at the implant base, adjacent to the bone, to affix to the porous trabecular bone after the subchondral bone layer was removed. This would also provide structural strength to the prosthesis. Moreover, through-the-thickness material transition is accessible by interweaving polymer fibers to the base metallic structure, allowing a stiffness gradient similar to the native cartilage tissue. Polymer weaves also provide resilience and soft elastohydrodynamic lubrication (EHL) similar to the articular cartilage [8].

Compared to laminated composites, 3D woven composites offer improved out-of-plane properties, due to the binding yarns, introduced in the normal direction to weft and warp yarns, resulting in an enhanced delamination resistance [9,10]. It was shown in another study that 3D woven preforms were able to absorb twice the impact energy

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[*] Corresponding author.
E-mail address: mohammadreza.arjmandi@aut.ac.nz (M. Arjmandi).

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compared with 2D woven constructs, without any sign of fiber damage, suggesting their potential capacity for load-bearing applications [11]. A compression after impact test showing damage initiation and further propagation was performed and 3D woven textiles showed up to 79% higher performance compared to 2D woven fabrics [12]. Other properties observed in these materials, including high interlaminar toughness [12] and high strain to failure, an indication of damage-tolerance [9,13], suggest 3D woven preforms as an efficient structural component for load-bearing applications.

3D networks of hydrophilic polymers swelling in water, also known as hydrogels, were studied as a potential cartilage replacement candidate, due to their biocompatibility and tunable mechanical properties [14–16]. Any improvements in their mechanical response of the soft hydrogels is highly desirable, thus fiber-reinforcement was suggested in recent studies [17,18]. However, only tough hydrogels could be considered as the matrix for such composite materials, as those with low toughness could not sustain the fiber-matrix interaction once being loaded, and the composite fails as soon as fibers cut through the hydrogel matrix [19]. Interestingly, a similar form of composition is found in the articular cartilage itself, where proteoglycan matrix gel is reinforced by collagen fibers, and although containing over 70 wt% water, the tissue is stiff and tough [20,21]. Interpenetrating polymer network (IPN) (also known as hybrid) hydrogels, developed by covalent crosslinking of polyacrylamide (PAAm) and ionic crosslinking of alginate (ALG), were turned out to exhibit significantly higher stiffness and toughness values compared to the single networks [22]. Therefore, fiber-reinforcement is feasible due to sufficient matrix toughness of hybrid hydrogels, and hydrogel composites benefit from even more robust mechanical performance, becoming eligible for use in structural components. In this regard, the PAAm-ALG hydrogel was reinforced by steel wool and the mechanical properties were investigated, in addition to a failure mechanism characterization [17]. It was shown that under tension, the hydrogel matrix was capable of maintaining the steel fibers in place, preventing their original entanglement to be disrupted, and hence the composite could sustain large deformation to the point where fibers eventually turned towards the loading direction, and the composite failed due to fiber-matrix debonding. Moreover, the amount of dissipated strain energy could be engineered by changing the reinforcement concentration, suggesting an additional design parameter compared to unreinforced hydrogels.

In addition to achieving a structure with desired stiffness and toughness, friction and wear behaviors in the presence of biological lubrication are other crucial factors determining the component life span and its reliability. It is worth mentioning that the tribological layers of articular cartilage are located at superficial and middle zones, in which the former contribute to boundary lubrication, while the latter develop fluid film support [23,24]. These layers also benefit from semipermeable medium, facilitating interstitial fluid pressurization and boundary lubrication, known as the major mechanisms associated with ultra-low friction coefficient of the tissue [25]. Similarly, hydrogels are biphasic and under load, could exude their fluid phase and contribute to lubrication, a significant advantage over dense materials. To simulate the same interstitial fluid pressurization of cartilage in a hydrogel, its permeability must be controlled, which in turn is a function of pore size and interconnectivity [26]. This suggests that using woven scaffolds without a hydrogel matrix is not an efficient way to achieve a high load-carrying capacity once the structure is loaded. Instead, infusing the hydrogel into the woven fabric, not only enhances the mechanical properties of the composite, but the fluid phase is efficiently retained within the structure and contributes to load support as well as lubrication. The fluid load support also accounts for stress relaxation observed in hydrogels [27].

In the past, various hydrogels were evaluated against mechanical and tribological criteria required for synthetic cartilage [14,28,29]. 3D woven hydrogel composites were studied as synthetic cartilage, although the same yarn size was utilized for weaving the whole fabric and the stiffness gradient found in cartilage was not considered in the weave [3,4]. The individual fibers to be used in 3D woven fabrics were assessed with microscale screening methods using atomic force microscopy in a previous study [30]. Polyester weaves produced in 3D woven form were also evaluated against sliding wear [31]. Furthermore, a unit-cell-based finite element model was developed for the simulation of sliding wear of 3D woven fabrics based on Archard's wear law in another study [32]. In the current study, we developed various novel 3D woven textiles with through-the-thickness stiffness gradients, inspired by the articular cartilage tissue, and infused it with hybrid hydrogel to produce hydrogel composites with superior mechanical properties. As an initial attempt, only the load-bearing layer was produced from polymer fibers and infused with hydrogel. Mechanical tests such as compression and indentation were carried out to characterize the elastic properties of composites, making the comparison with the reference un-reinforced hydrogel possible, while stress relaxation tests were conducted to study the effect of reinforcement design on the viscoelastic behavior of the hydrogel composite. Furthermore, friction and wear responses were investigated using reciprocating sliding motion against an alumina ceramic ball, allowing the effect of various parameters such as load, slip rate and biological lubricant to be identified. Results were analyzed to find out any interactions between mechanical properties and the frictional response of the hydrogel composite.

2. Experimental

2.1. Materials

2.1.1. 3D woven textile production and analysis

Medical-grade monofilament polyvinylidene fluoride (PVDF) yarns were purchased in 3 different sizes, called USP-2.0 (Ø0.315 mm), USP-3.0 (Ø0.23 mm), and USP-5.0 (Ø0.134 mm) from G. Krahmer GmbH, Buchholz, Germany. The weaving design and manufacturing process were conducted at the Institute für Textiltechnik, RWTH Aachen University, Germany [33]. A needle loom (Type NH2, Jakob Müller AG, Frick, Switzerland) was utilized for the weaving process. Three distinct layers were incorporated into the woven preform, each having a different warp thread size as schematically shown in Fig. 1(a) to mimic the stiffness gradient feature of the articular cartilage. The weft threads were selected from individual monofilament size USP-3.0 and double insertion was accomplished by custom setting of the weaving loom. For the binding of the multilayered woven structure, medical-grade polyethylene terephthalate (PET) multifilament yarns were selected which passed through the layers (z-direction, Fig. 1(b)) in a continuous fashion. Weaving trials revealed weft density (weft thread picks per unit length of the fabric) would affect porosity and total thickness of the 3D textile. Starting from 15 picks/cm, the thickness was found insufficient and the structure seemed unstable. With a gradual weft density increase, the combination of 55 picks/cm for every first and 15 picks/cm for the subsequent 7 weft insertion in a row seemed efficient as a uniform textile and was produced with the thickness and porosity close to articular cartilage (2.6 cm [34]). The weave was called *Type-A*. The second weave pattern *Type-B* was developed so that each single layer is interlocked with one another, as schematically shown in Fig. 1(c), to achieve more flexibility in thickness and higher stability and less relative movements of fibers. A weft density of 35 picks/cm was considered proper for the second pattern.

Various view angles of each pattern are also illustrated in Fig. 1(b) and (c), using gold-sputtered specimens in scanning electron microscopy (SEM, Hitachi SU-70). Dissimilar fiber configurations at the top and bottom layers, and different fiber sizes implemented at each layer could be clearly distinguished from this inspection. Furthermore, double weft insertion could be seen from side view images, attributed to the weaving mechanism of needle loom. It should be noted that the stiffness gradient through the thickness was achieved by using fibers