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Nanocoating Strategies for Enhancing Traditional PET Anterior Cruciate Ligament Grafts

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Abstract:

Anterior cruciate ligament (ACL) injuries are becoming increasingly common nowadays. Anterior cruciate ligament replacement (ACLR) is the surgery people turn to in most cases. Autografts are often the first choice for ACLR, but harvesting autografts prolongs recovery time and damage the healthy tendon in the other area. For artificial ligaments, polyethylene terephthalate (PET) ACL is mostly used; they do not have autograft' s drawbacks but are constrained by their lack of bone integration, low wear resistance, and infection risk. Recent advances in nanocoatings provide insight into improving the PET ACL surface's properties. Nanocoatings made of bone morphogenetic protein (BMP)-2-loaded ligament, triple-nano-coated ligament, and hydroxyapatite (HAp) have shown significant promise in improving bone integration. The nano-TiN composite exhibits excellent wear resistance. Metal-based and carbon-based nanocoatings have also proven to be effective in preventing infection risks. Despite its advantages, there are still issues, such as the toxicity of nanoparticles, the gap between humans and small animals, and the neglect of soft tissue integration. This review suggests nanocoating techniques for enhancing conventional PET ACLs and provides direction for future ACLR development.

Keywords: Anterior cruciate ligament reconstruction; Polyethylene terephthalate; Artificial ligament; Nanocoating.

1. Introduction

The anterior cruciate ligament (ACL) is located in the knee joint, connecting the thighbone and the shinbone and maintaining knee stability. ACL injuries are becoming common nowadays, especially occurring most commonly in football, soccer, and basketball athletes when the knee is twisted, displaced, or traumatically impacted. ACL injuries can cause meniscal degradation, arthritis, unstable knees, and decreased athletic performance if left untreated. Conventional treatment is anterior cruciate ligament reconstruction

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(ACLR). The choice of the graft material is a crucial factor. In most cases, patients choose autografts-harvesting tissues from their own body. Bone-patellar tendon-bone (BPTB) autograft, the hamstring tendon (HT) autograft, and the quadriceps tendon (QT) autograft are the most frequently used autografts in ACLR. Despite harvesting tissues from the human body slowing down the rehabilitation process, impacting the donor site, this approach is still continuously favored. Another graft choice is allografts, which include a high risk of infection.

Artificial ligaments are also an option in ACLR. Polyethylene terephthalate (PET) is the most widely used material for artificial ACLs. Using artificial ligaments can avoid donor site damage and hasten the rehabilitation process. PET grafts exhibit several shortcomings. First, they lack osseointegration with the surrounding bone; next, long-term friction may results in wear debris [1]; and finally, their surface property provides a favorable environment for bacteria, which raises the risk of infection [2]. Even when extracting autografts damages the patient‘s healthy tissue parts, clinical practices are reluctant to use artificial ligaments, and autografts are their first option overall.

Nanotechnology manipulates materials with 1-100 nm (1 nm = 10^{-9} m). Nanotechnology is increasingly used to improve a number of medical challenges, including drug discovery, disease prevention, and disease diagnosis. The nanocoating process is one of the most promising uses for improving surface properties. Nanocoating is a nanoscale thin layer deposited on the surface of materials. Their special qualities make them a great option to enhance conventional artificial ligaments since they can successfully encourage cell adhesion and biocompatibility, prevent bone debris wear, and offer antibacterial capabilities.

This review focuses on the nanocoating techniques to overcome the drawbacks of traditional PET ACL material, aiming to provide insight for the future development of ACLR.

2. Limitations of Artificial PET ACL

PET is the most used material in PET ACLs due to its superior tensile strength, excellent dimensional stability, great thermal resistance, and outstanding chemical resistance[3]. However, PET has several shortcomings when it comes to coating applications, including a lack of bone integration, risk of wear debris, and infection issues.

2.1 Bone Integration

The PET surface is inert, and this property makes PET integrate poorly with the bone. PET has low biactivity, which can eventually lead to bone tunnel resorption, lowering the ligament’ s stability. PET’ s hydrophobic surface is difficult for cells to attach to, reducing effective osseointegration [2].

2.2 Risk of Long-Term Wear

Long-term friction and stress can cause PET wear, limiting the lifespan of the PET ACL. In most cases, complication rates are high. For example, research shows that patients implanted PET ACLs after an average of 16.5 years: 66% of them experienced complications, including 20% synovitis and 24% graft failures; approximately half of the patients had reoperation [4]. In another study, a mean of 7.5 years following surgery was associated with a 22% ACL rupture rate [5]. These studies indicate that high mechanical failure rate and wear rate are major concerns for the long-term use of PET ACL.

2.3 Risk of Infection

The PET surface provides a favorable site for bacterial colonization, especially for Staphylococcus epidermidis [6]. In ACLR, bacterial colonization of PET grafts can lead to infection and inflammation, and even graft failure. Thus, the plan of the PET ACL is often held back.

3. Application of Nanocoatings for Improving PET ACL

In recent years, nanocoatings have demonstrated promise in enhancing bone integration, fostering wear resistance, and reducing infection risk in PET ACLs.

3.1 Improving Osseointegration

One of the best ways to prevent surface corrosion and improve biocompatibility for PET ACL is to apply a protective coating. Hydroxyapatite (HAp), dicalcium phosphate dihydrate, and octacalcium phosphate on its surface can be deposited on the surface of PET grafts to improve osseointegration[7, 8]. Researchers have shown that HAp-coated ligaments can promote bone cell development in the ligament in ACLR. Because HAp releases osteogenic substances, such as calcium ions, to enhance cell adhesion. These characteristics have long made HAp used in ligament and bone treatments [9]. A great variety of approaches can be applied to obtain nano-scale coatings, including sol-gel, hydrothermal, plasma spray, and biomimetic [8].

Li et al. conducted coating tests for PET ACLs using an organic and layer-by-layer self-assembled template made of chitosan and hyaluronic acid, two recognized polymers for their biocompatibility. The layer-by-layer assembly technique enables precise control over coating thickness, ensuring uniformity of the PET surface coating. The in vitro experiments confirm that this coating enhances transplanted cell compatibility and stimulates osteoblast proliferation compared to uncoated ones; in vivo studies, researchers observed bone formation at the interface between the graft and the bone. Meanwhile, both osseointegration and stability of the ligaments were enhanced, thereby reducing the risk of graft failure [10].

Increasing hydrophilicity will result in an increase in osseointegration, since wettability can enhance cell adhesion by laying a solid foundation. For example, by encouraging both in vitro and in vivo cell attachment, it has recently been demonstrated that strontium-substituted hydroxyapatite (SrHA) coatings increase the hydrophilicity of PET ACLs; therefore, boosting bone integration. Furthermore, Li et al. focused on bone morphogenetic protein (BMP)-2-loaded ligament and triple-nano-coated ligament chondroitin sulfate (CS) coating technique. BMP-2 can stimulate the formation of bone, and CS mimics the structure of the human body. The combination has two outperforming functions: First, it may regulate cytokine release; second, it can guide the differentiation of stem cells towards bone cells. Together, stimulate the synthesis of osteocytes and enhance the stability of the PET ACL [11].

Zinc oxide nanoparticles (ZnO-NPs)-coated implants have biocompatibility and antibacterial properties, which improve bone-soft tissue integration, thereby strengthening implant fixation. In addition, ZnO-NPs, as osteoconductive nanoparticles, interact with bone to attach to the implants, stimulating osteoblast adhesion. [7].

3.2 Promotion of Wear Resistance

Nano-sized HAp is broadly used as a coating material to prevent debris wear since it has excellent mechanical properties-high hardness and high wear resistance-which is an ideal option for PET ACL coating to prevent wear debris [12]. Hank' s solution was used to measure the nano-TiN composite' s wear performance under a 10N stress. In addition, the ISO-10993-4 standard was used to assess the biocompatibility. The nano-TiN composite structure has been shown to greatly enhance strength, hardness, and anti-wear performance in Hank's solution, which is an ideal choice for improving the wear performance of PET ACLs [13].

3.3 Antibacterial properties

ZnO-NPs can reduce infection risk by inhibiting bacterial adhesion, creating a more favorable environment for ACL healing [7].

Nanostructured HAp layers are antibacterial, thus reducing bacterial colonization. In addition, metal-based or carbon-based nanomaterial coatings have been widely used in the biomedical field due to their antibacterial effects. These coatings can inhibit bacterial growth through ion release. Graphene-based modifications can disrupt microbial metabolism, thereby preventing bacterial growth [14]. The multifunctional coatings provide defense against infection, making them optimal to prevent PET ACL grafts' infection risks.

Some metal-based nanomaterials have antibacterial properties, such as silver ions (Ag^+), zinc oxide (ZnO), and copper nanoparticles (Cu-NPs), and they can be incorporated into implant surfaces, inhibiting bacterial adhesion and maintaining biocompatibility. In Wang et al.'s experiment, silver nanoparticles (Ag-NPs) were coated on the PET ACL surface between the polydopamine layer and Ag^+. Cu-NPs and Ag-NPs show outstanding antibacterial activity against Escherichia coli. After ten washes, the majority of the Cu-NPs and Ag-NPs still adhered to the PET ACL, indicating high durability. These metal coatings are reliable choices for surface modification and for preventing bacterial colonization [14].

4. Challenges

Concerns remain on nanocoatings for PET ACL grafts. First, most experimental studies have been performed on small animals such as rabbits, which are different from humans in size, capacity, and healing. To bridge the clinical gap, future studies should be done on larger animals whose biological responses are closer to humans, providing more reliable outcomes [11]. Second, the ACL is connected to two interfaces: one side is attached to the bone, and the other side is connected to soft tissue. Most studies focus on "osseointegration" at the bone–ligament interface, and neglect the soft tissue side. To improve both sides' integration, research should develop dual-functional coatings that simultaneously promote bone ingrowth and soft tissue attachment. Third, while nanocoatings offer mechanical benefits, they may result in toxicity. After mechanical wear, particles may be shed from the coating surface. These released nanoparticles will accumulate, leading to oxidative stress, inflammatory reactions, or DNA damage. Before widespread clinical application, toxicological studies must be conducted to evaluate the effects of wear particles. To ensure that nanocoatings enhance wear resistance without introducing safety issues.

5. Conclusion

PET has become the most widely used material in ACLR due to its excellent mechanical strength and chemical stability. Autografts are preferred in surgeries because of the weakness in the PET ACL, even if the donor location

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loses healthy ligament tissue. Nanocoating offers an innovative solution: HAp, SrHA nanocoatings, and triple-layer BMP-2 nanocoatings enhance osteocytes' adhesion and osseointegration. Nano-TiN composite reduces bone debris. Nanocoatings made of metal and carbon, such as copper and silver, have antibacterial properties. Nanocoated PET is a promising approach to improving artificial ACL. However, the discrepancies between small animal models and humans, the lack of soft tissue integration, and nanomaterial toxicity are holding the strategy back. Future research should include studies in large animal models, control for toxic release, and develop coatings that combine osteointegration and soft tissue integration. Once the efficacy of PET has been confirmed, PET material could replace autografts, reducing complications at the donor site, shortening recovery time, and improving the results of ACLR.

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