Preparation and characterization of composited PVA-H

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Hydrogel is defined as a polymer having a three-dimensional network structure. Compared to conventional biomaterials such as ceramics and metals, hydrogels possess better biocompatibility, owing to their unique capability to hold water. Among all the hydrogels, poly (vinyl alcohol) hydrogels (PVA-H) are most widely used in artificial cartilage because it has a similar structure to cartilage, possess good biocompatibility, and is easy to prepare or modify. However, PVA-H is known to be a bioinert material and is therefore extremely difficult to adhere and affix to the surface of a living joint. In addition, PVA-H is usually made by low temperature crystallization (LTC) method using dimethyl sulfoxide (DMSO) as a cryoprotectant, which is harmful to our body. In this study, a novel hot pressing (HP) method was used to prepare PVA-H, in which no organic solvent was used. Graphene oxide (GO) and inorganic salts were used as composite materials, aiming to reinforce the mechanical strength and improve the cell adhesion of PVA-H.

First, the conventional LTC method was used to prepare GO composited PVA-H. GO was found to have a reinforcing effect on Young's modulus. The addition of GO increased the roughness and hydrophobicity of gel surfaces. In addition, cell attachment was improved by the addition of GO, and it seemed that a rough structure is beneficial to cell adherence.

Subsequently, GO composited PVA-H was prepared by the novel HP method for better biocompatibility. Cell attachment was also found to improve by the addition of GO, which might be due to the increased hydrophobicity and higher protein affinity. Unexpectedly, however, GO was found to non-uniformly disperse in the gel and aggregate a lot. As a result, the tensile strength of the gels decreased significantly.

Since the gelation was not conducted in a water environment by the HP method, composite materials such as GO might have some problems in dispersing in the gel. Therefore, I tried to use inorganic salt as a composite material. A number of salts (LiCl, LiBr, $CaCl_2$, $MgCl_2$, etc.) were successfully composited to PVA-H by the HP method. It was found that the addition of salt impeded the combination of PVA and water molecules, thus affecting the crystallinity of gels. Young's modulus, glass transition temperature, and melting point decreased with the addition of Li, Mg, Mn salt, confirming the feasibility of application as a plasticizer. $CaCl_2$ composited PVA-H, on the other hand, was found to have a bridging structure formed by Ca^{2+} and O atoms. Therefore, Young's modulus and T_g increased at low salt concentration. Taking into account that salt in gels may elute in the solvent over time, which was not beneficial for biomaterials, a desalting treatment was carried out and salt in each gel was completely removed. Furthermore, all of the desalted gels were found to have no toxicity and showed higher protein and cell affinity than pure PVA-H, which is beneficial for artificial cartilage materials.

Keywords: hot pressing method, PVA, hydrogel, GO, salt