Abstract

Cellulose nanofiber (CNF) is the most abundant and renewable natural polymer on the planet. Nowadays, since the environmental problems are becoming severe every year, the development of materials based on green materials such as CNF is of great significance. Because of its unique properties such as high specific surface area, high mechanical strength, reactive surface, biocompatibility, biodegradability, and non-toxicity, CNF has become a very attractive biological material. [1] The surface reactivity allows surface modification, hence the modification of cellulose to improve biocompatibility is a very attractive solution. In this dissertation I have used two methods to modify CNF to enhance its adhesion to cells and biodegradability and analyzed the principles of these two methods in order to achieve precise control.

The first method is the grafting of concentrated polymer brushes onto CNF. Polymer brush grafting is a practical surface grafting technique that can easily alter the physical and chemical properties of the surface, such as hydrophilicity, elasticity, and cell adsorption. Concentrated polymer brushes (CPBs), that is, grafted chains at very high concentrations which exhibit unique entropic properties, and CNF grafted with CPBs (CNF-CPBs) can flocculate with cells to form flocs, [2] which is promising as a 3D culture system. There is a possibility that this flocculation phenomenon can be explained by the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory that guides colloidal flocculation, but further proof is needed. According to the DLVO theory, the flocculation phenomenon is influenced by the surface charge and spatial location. Therefore, in Chapter 2, CPBs with different charges were introduced on the CNF surface and co-cultured with cells. The results demonstrated that larger flocs could be obtained at appropriate zeta potentials. In Chapter 3, CNF-CPBs with different fiber lengths were synthesized, and flocculation of the three cell types was observed. Larger flocs are formed in samples of CNF-CPBs with shorter fiber lengths. Equally important was the fact that all three cells exhibited the same flocculation phenomenon. The results proved that flocculation based on CNF-CPBs, and cells can be explained by the DLVO theory and has the potential to be applied to a broader range of cell lines. In conclusion, this is the first time the DLVO theory has been used to explain the flocculation phenomenon in cells and CNF-CPBs.

The degradability of materials is also an essential indicator of the adaptability of biomaterials. [3] In this study, the Malaprade reaction introduces dialdehyde into CNF, which can be degraded by the Maillard reaction triggered by the Schiff base reaction of dialdehyde CNF with amino acids in the body. Furthermore, the effect of reaction conditions on the reaction rate, CNF molecular weight, and crystalline index in this series of reactions was systematically investigated in order to better understand and control the degradation behavior of CNF. Briefly, the molecular weight decreased in both the oxidation and decomposition reactions. Crystalline index studies demonstrated that the Malaprade reaction occurred in crystalline region, which leads to a decrease in the

crystallization index. While the degradation reaction occurs mainly in the amorphous region, hence the increase in a crystalline index after the degradation reaction.

In this thesis, the biocompatibility of CNF was enhanced by surface grafting of CPBs and triggering the Maillard reaction for CNF based on oxidation of CNF, and the principles of the reaction were explored for precise control.

Keyword: *cellulose nanofiber; concentrated polymer brush; flocculation; DLVO theory; surface charge; fiber length; Malaprade reaction; Maillard reaction*

Reference

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