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Methacrylated chitosan with improved mucoadhesiveness for transmucosal application

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Background: The continued relevance and biomedical application of chitosan are due to its safety and mucoadhesiveness. Mucoadhesive delivery systems are desirable to extend the bladder residence time of loaded drugs. In recent years, maleimide and acrylate-functionalised delivery systems are being explored for transmucosal application due to their superior mucoadhesive features relative to thiolated drug carriers. Methacrylated drug carriers have not been explored for enhanced mucoadhesiveness. In this work we have synthesised methacrylated chitosan with a variable degree of modification (LMeCHI and HMeCHI) and evaluated their pH sensitivity, mucoadhesiveness, and safety in comparison to chitosan (CHI), for intravesical use.

Methods: Products were characterised using 1H Nuclear Magnetic Resonance (1H NMR), Fourier Transform- Infrared and UV-Vis spectroscopy. 1H NMR and ninhydrin test quantified the methacrylate grafting density on chitosan. Turbidimetric analysis of samples evaluated their resistance to pH changes in the biological fluid. The mucoadhesiveness of fluorescein sodium in the presence of the mucoadhesive polymers was evaluated using artificial urine flow-through techniques and fluorescence microscopy. MTT assay was used to study their UMUC3 malignant cell antiproliferative features.

Results: There was a broad correlation in the methacrylation extent of LMeCHI and HMeCHI obtained with both methods. Turbidimetric analysis (λ = 400 nm) revealed that the turbidity of HMeCHI solution remained unchanged from pH 3 to 9 while that of CHI and LMeCHI increased rapidly after pH 6, inferring that the stability of the drug carriers in biological fluid may be improved by methacrylation. The degree of methacrylate conjugation had a profound influence on their mucoadhesiveness. The polymers are presented in order of increasing mucoadhesion: FITC-Dextran < FS/CHI< FS/LMeCHI < FS/HMeCHI. Based on MTT assay, the UMUC3 cell antiproliferative effect of the unmodified and modified chitosan solutions (6.25-200 µg mL-1) was not significantly different, confirming the biocompatibility of our novel mucoadhesive polymers. Methacrylation of chitosan did not compromise its biocompatibility with bladder cancer cells.

Conclusions: Methacrylated chitosan is a safe drug carrier for intravesical delivery with superior mucoadhesiveness relative to chitosan. This result suggested that the degree of methacrylation can be tailored for desirable physicochemical properties of methacrylated chitosan. HMeCHI appears the most promising for intravesical delivery of bladder cancer chemotherapeutics

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