Conformational study of chitin and chitosan filaments in solution via Molecular Dynamics Simulation.

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Introduction

Chitin, a (1→4)-linked N-acetyl-β-D-glucosamine is widely distributed in nature as the main component in the exoskeleton of crustaceans, insects and fungi. The principle derivative of chitin is chitosan, generally produced by alkaline deacetylation of chitin[1].

These biopolymers have been widely developed for use as antimicrobials, biomedical materials, cosmetics, agricultural materials and principally as decontaminant of residual waters containing pesticides and heavy metals[1]. These various applications require information about the molecular conformation of chitin and chitosan[2].

Many experimental studies describe that chitin and chitosan properties can be affected by degree of acetylation (DA) of the biopolymer and by experimental conditions such as ionic strength (I), pH and solvent[3]. The goal of this work is to evaluate the influence ionic strength, pH and acetylation degree on the structural dynamics of chitin and chitosan fibers in aqueous solution.

Results and Discussion

In this study, a set of explicit-solvent 30-ns molecular dynamics simulations of chitin and chitosan fibers were performed at different ionic strength (0.0, 0.4 and 1.1 mol.Kg^{-1}), pH (basic, 6.5 and acid) and degrees of acetylation (0%, 20%, 40% and 100%), using full periodic boundary conditions at 300K and 1 atm. The GROMOS carbohydrate field[4] was used along with the GROMACS 3.1 package[5].

From the randomly generated initial configurations, the equilibrium structures converged to conformations in which their ϕ and φ dihedral angles are in agreement with the experimental data obtained by x-ray and electron diffraction[2]. A conformational analysis revealed that the chitin chains have preference for the extended 2-fold helix motif, and the chitosan filaments assume preferably a 5-fold and a relaxed 2-fold helix.

The variation of the ionic strength in all simulations only affected the convergence of the chitin filaments and not their conformation.

For chitosan, the variation in the degree of acetylation resulted in minor changes in solubility, conformation and electrostatic potential. On the other hand, pH variation, from basic to acid, leads to an increase in solubility and conformational changes as a result of the electrostatic attractive/repulsive interaction between the deprotonated/protonated amino groups. It is illustrated by the electrostatic potential surface, which was calculated by solving numerically the non-linear Poisson-Boltzmann equation and applying a finite-difference procedure using the APBS program[6] (Figure 1).

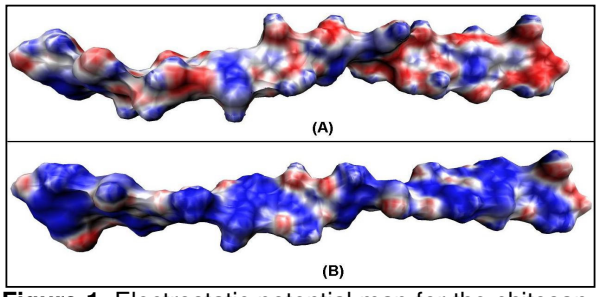

Figure 1. Electrostatic potential map for the chitosan in (A) basic and (B) acid media

Conclusions

The simulations suggest that variations of the ionic strength only affect the conformational kinetics chitin, while chitosan fibers are strongly affected by pH variations.

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