



Review article

Chitosan as an adhesive



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ABSTRACT

Chitosan is a well-known polysaccharide abundantly published during the last decades. This heteropolymer, composed of 2-acetamido-2-deoxy-D-glucopyranose and 2-amino-2-deoxy-D-glucopyranose, is obtained after alkaline deacetylation of chitin from crustaceans, fungi and other non-vegetable organisms. Soluble only at acidic pH, it is the unique polycationic polysaccharide extracted from bioresources. This characteristic gives to it original and specific properties finding some applications in several industrial fields but especially in the biomedical one because of its biocompatibility and its non-toxicity. Besides these traditional applications other ones begin actually to appear in the literature. They focus on the development of chitosan-based adhesives, binders or films. This review synthesizes the state of the art on this domain, but also deals with the assessment of chitosan environmental impact.

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1. Introduction

Chitin, deacetylated chitin (chitosan) and their derivatives are probably in the top 3 of the more published polysaccharides in the scientific literature, with cellulose and starch. This “old polysaccharide” sometimes called “animal cellulose” has also interested the field of industrial applications as a simple research in web site of the European Patent Office using the term “chitosan” finds 16,300 patents. However, despite this abundant literature and activity of patent deposition, chitin and chitosan have not known the same commercial success than cellulose even if these two polysaccharides are in competition for the most abundant natural organic compound on earth. The explanation could be in the structure of chitin which has some specificity compared to cellulose even if these two polysaccharides have basically a similar arrangement pattern. They are high molecular weight polymers of glucose linked by β -(1,4) glycosidic linkages. However, in the case of chitin, acetamido groups replace hydroxyl ones on the C-2 of glucose units leading to a polymer of N-acetyl glucosamines. The oxygen of this acetamido group can form hydrogen bonds with adjacent $-\text{NH}$ or $-\text{OH}$ functions reinforcing the insolubility of chitin. So, the degree of N-acetylation, i.e. the ratio of 2-acetamido-2-deoxy-D-glucopyranose to 2-amino-2-deoxy-D-glucopyranose structural units has a striking effect on chitin insolubility and limits its swelling possibilities in water compared to cellulose [153]. This main difference between these two polysaccharides can explain why the evolution has done the choice to conserve them for the same biological function, meaning the mechanical resistance and tissue isolation of organisms. Cellulose has been selected by plants which have to conserve some degrees of interactions with water, notably during the growth of their tissues, whereas chitin is employed by some marine organisms and insects which have to build an impermeable exoskeleton. Fungi have not done any choice having cell-walls with chitin and cellulose. Chitosan obtained after alkali deacetylation of chitin exhibit rates of deacetylation higher to 50% and can easily form quaternary nitrogen salts at low pH values where it is soluble. The expensive production of chitin and chitosan from marine organisms compared to cellulose obtained from terrestrial plants is probably at the origin of their under exploitation despite their huge potential. Current applications are usually concentrate on the biomedical fields such as tissue engineering, gene vectors, and drug carriers [43,187]. However, the increase of commercial availability of chitosans open the way to the development of materials notably in the field of films, binders and adhesives [50,142,153,176]. Most adhesives viz. polyvinyl acetate, epoxy adhesives, phenol/formaldehyde and polyurethane depend on non-renewable and depleting petrochemical resources. Moreover, numerous adhesives, films and binders are prepared with residual toxic chemi-

cals, such as formaldehyde and Volatile Organic Compounds (VOCs) that are injurious to health and environment [142]. The development of natural and renewable molecules exhibiting good bonding properties is actually an industrial challenge. The bio-based polymers need to match the properties of conventional compounds and/or introduce new valuable properties, preferably without any cost increase. Protein, tannin, lignin, and polysaccharides are examples of interesting bio-based polymers that have been suggested as good candidates for adhesives and binders developments [154]. Among them polysaccharides such as chitosan are an interesting group of polymers. Their numerous hydroxyl groups can interact with a lot of chemical functions and their high molar mass allows cohesive strength to materials. One of advantage of chitosan is its insolubility at neutral pH and its solubility at acidic ones. The combination of all these characteristics with its good mechanical properties, its biocompatibility and its biodegradability open the way to a lot of original applications in the adhesive and binder area. Note also that recently developed chitosan based nanomaterials have superior physical and chemical properties such as high surface area, porosity, tensile strength, conductivity, photoluminescent as well as increased mechanical properties compared to pure chitosan [182]. It appears then that a consolidation of data relating to this subject is required.

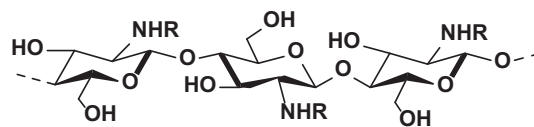
2. Chitosan: from the source to its characteristics

2.1. Origin

Chitin was the first polysaccharide identified by man from mushrooms preceding cellulose by 30 years [23,102]. After that this polymer was identified in the shells of insects and the exoskeletons of molluscs combined with minerals and proteins to harden these structures by cross-linking with polyphenols. In 1859, C. Rouget subjected first chitin to alkali treatment, which resulted in a substance that could be dissolved in acids. In 1894, the term “chitosan” was given to this deacetylated chitin by Hoppe-Seiler [187].

2.2. Structure

Chitins can be divided into α -, β -, and γ -chitins due to hydrogen bonds in their structures. α -Chitin comprises



(With R = H or COCH₃)

Fig. 1. Chemical structure of chitosan (%COCH₃ \leq 40%).

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