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Cross-linked chitosan polymers as generic adsorbents for simultaneous adsorption of multiple mycotoxins

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ABSTRACT

The primary objective of this study was to synthesize three types of cross-linked chitosan polymers and further investigate their adsorption capability for multiple mycotoxins, including aflatoxin B_1 (AFB_1), ochratoxin A (OTA), zearalenone (ZEN), fumonisin B_1 (FB_1), deoxynivalenol (DON) and T-2 toxin (T2). Among these synthetic adsorbents, cross-linked chitosan-glutaraldehyde complex presented the highest adsorption capability for AFB_1 (73%), OTA (97%), ZEN (94%) and FB_1 (99%), but no obvious adsorption for DON and T2 (<30%). The effect of various incubation conditions (contact time, dosage and pH) was also studied. Subsequently, the experimental data were fitted to Langmuir, Freundlich and Hill models. The best fitting model to describe AFB_1 and FB_1 adsorption was Langmuir model ($R^2 \geq 0.99$), with the theoretical maximum adsorption amounts of 5.67 mg/g for AFB_1 and 15.7 mg/g for FB_1. The Hill model was the best model for OTA and ZEN adsorption ($R^2 > 0.98$), with the predicted maximum adsorption amounts were 24.8 mg/g for OTA and 9.18 mg/g for ZEN. In addition, the adsorption capability of adsorbent for the simultaneous presence of multiple mycotoxins was also evaluated in buffer system and simulated gastrointestinal condition. The results indicated that the coexisted multiple mycotoxins didn't affected the adsorption capability of adsorbent, whereas the adsorption amounts of toxins were decreased by some gastrointestinal components. The findings of this research suggest that chitosan–glutaraldehyde complex has the potential to be applied as multitoxin adsorbent material for reducing the combined adverse effect of multiple mycotoxins on humans and animals.

1. Introduction

Mycotoxin contamination in cereal grains and feedstuffs has evoked global concern due to its potential health risk on humans and animals (Hussein & Brasel, 2001; Zaki, El-Midany, Shaheen, & Rizzi, 2012). Currently, more than 300 mycotoxins have been identified worldwide (Streit et al., 2012; Zaki et al., 2012). The most frequently monitored mycotoxins are aflatoxins, ochratoxins, zearalenone, trichothecenes and fumonisins (Armando et al., 2012; Hussein & Brasel, 2001; Zaki et al., 2012). These contaminants have carcinogenic, teratogenic, nephrotoxic and hepatotoxic effects on humans and animals (Hussein & Brasel, 2001). In addition, it has been demonstrated that the combined intake of multiple mycotoxins presents additive and synergetic toxic effects (Speijers & Speijers, 2004). To reduce mycotoxin production and avoid the resulting adverse effect, several types of management measures have been carried out, such as good agricultural practices, ideal storage conditions and the establishment of legislative limits in cereal grains and feed (Jouany, 2007; Streit et al., 2012). In spite of the substantial efforts, complete avoidance of mycotoxin contamination in agricultural commodities is impossible. Thus, it is necessary to develop practical and effective decontamination techniques.

The known strategies for the detoxification of mycotoxin-contaminated food and feed include chemical, biological and

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physical methods (Armando et al., 2012; Jouany, 2007; Vanara, Reyneri, & Blandino, 2009). Among these approaches, the addition of non-nutrition adsorbents into food and feed is regarded as the most effective and economical procedure to reduce the bioaccessibility of mycotoxin in gastrointestinal tract. So far, a large variety of adsorbents have been investigated for their capability to sequester mycotoxin *in vitro* and *in vivo*, such as inorganic adsorbents and their modified complexes (Daković, Tomašević-Čanović, Rottinghaus, Matijašević, & Sekulić, 2007; Mine Kurtbay, Bekçi, Merdivan, & Yurdakoç, 2008; Yue et al., 2013), organic adsorbents (Avantaggiato, Greco, Damascelli, Solfrizzo, & Visconti, 2014; Meca, Meneghelli, Ritieni, Mañes, & Font, 2012; Mine Kurtbay et al., 2008), and macromolecular polymers (Ramos, Hernandez, Pla-Delfina, & Merino, 1996; Solfrizzo, Visconti, Avantaggiato, Torres, & Chulze, 2001). However, most of these adsorbents only sequester single or two toxins while presenting poor adsorption capability for other toxins. Given the frequent co-occurrence of multiple mycotoxins in cereals and feedstuffs (Storm, Rasmussen, & Rasmussen, 2014; Zachariasova et al., 2014), the most desirable adsorbents must be able to simultaneously bind different types of mycotoxins.

Chitosan is a natural-based polyaminosaccharide and usually is obtained from waste biomass (shrimp, crabs and seashell) during seafood process (Crini, 2006) (Fig. 1). The biopolymer is non-toxic, biocompatible and biodegradable. It has been widely used as a promising biosorbent for the removal of various heavy metal ions and dyes. However, chitosan is very sensitive to pH and easily is dissolved in acid media (pH < 3), which limits its wide application in many fields. The chemical modification of chitosan with epichlorohydrin, tripolyphosphate and glutaraldehyde is regarded as an alternative to overcome the weakness (Varma, Deshpande, & Kennedy, 2004; Yong, Bolan, Lombi, Skinner, & Guibal, 2013). The cross-linked chitosan polymers not only enhance chitosan's resistance to acid media but also improve its adsorption capability (Crini, 2006; Yong et al., 2013).

Currently, only few studies are reported regarding the use of chitosan as adsorbent to sequester mycotoxin. For instance, chitosan bead could efficiently bind OTA in wine and improve wine safety (Bornet & Teissedre, 2008; Mine Kurtbay et al., 2008; Quintela, Villaran, De Armentia, & Elejalde, 2012). Chitosans with different molecular weights reduced the bioaccessibility of beauvericin in the simulated gastrointestinal fluid (Meca et al., 2012). Besides, a commercial adsorbent consisting of chitin, chitosan and chitosan oligosaccharides could provide protection against the combined toxicity of aflatoxins (AFs) and ZEN in duck (Khajarern, Khajarern, Moon, & Lee, 2003). However, to the best of our knowledge, there is no research carried out on the utilization of cross-linked chitosan polymers for simultaneous removal of multiple mycotoxins. Therefore, the aim of our work was to synthesize cross-linked chitosan polymers and assess their adsorption capability for the most frequently found mycotoxins in cereals and feeds, including AFB_1, OTA, ZEN, FB_1, DON and T2.

2. Materials and methods

2.1. Chemicals and reagents

Low-molecular-weight ($<2.0 \times 10^5$), medium-molecular-weight (2.0×10^5–2.5×10^5) and high-molecular-weight ($>5.0 \times 10^5$)

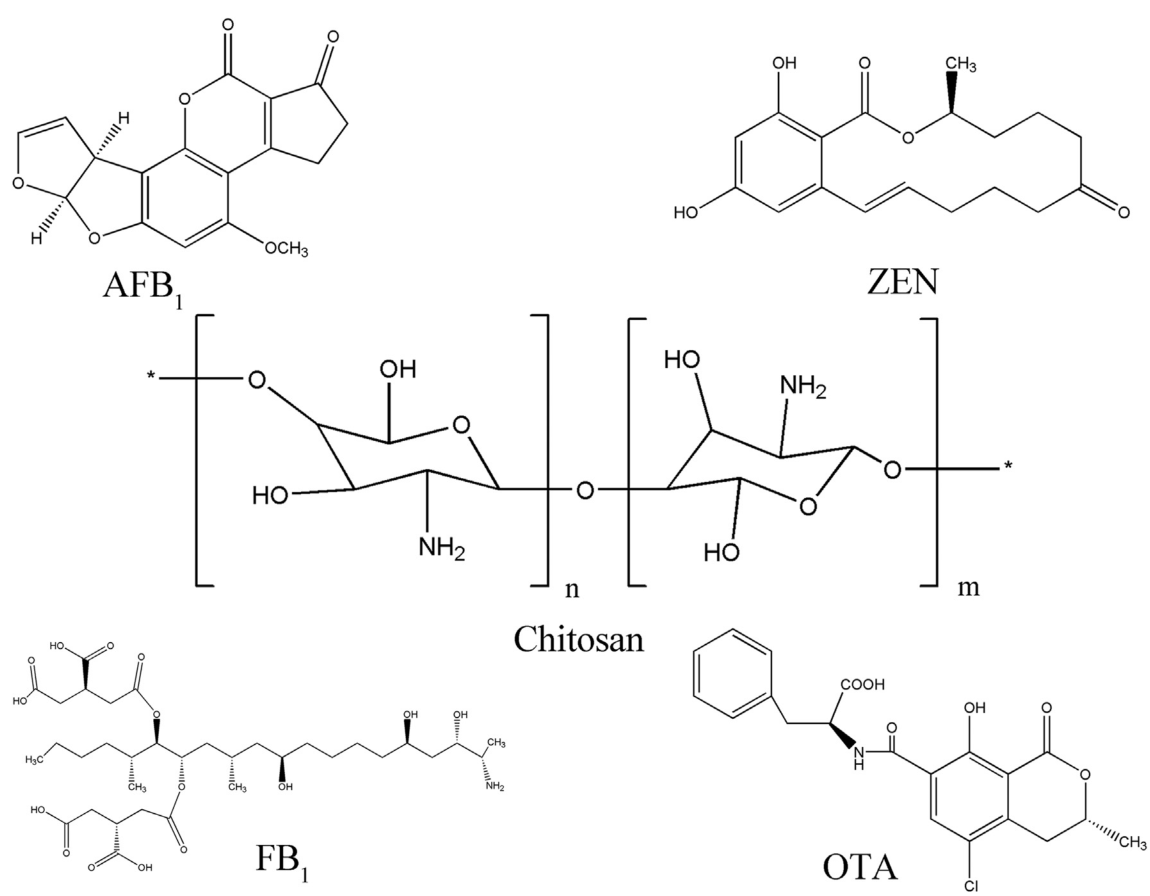

Fig. 1. Chemical structures of AFB_1, ZEN, FB_1, OTA and chitosan.