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C-Coordinated Chitosan Metal Complexes: Design, Synthesis and Antifungal Properties

Authors : Weixiang Liu, Yukun Qin, Song Liu, Pengcheng Li

Abstract : Plant diseases can cause the death of crops with great economic losses. Particularly, those diseases are usually caused by pathogenic fungi. Metal fungicides are a type of pesticide that has advantages of a low-cost, broad antimicrobial spectrum and strong sterilization effect. However, the frequent and wide application of traditional metal fungicides has caused serious problems such as environmental pollution, the outbreak of mites and phytotoxicity. Therefore, it is critically necessary to discover new organic metal fungicides alternatives that have a low metal content, low toxicity, and little influence on mites. Chitosan, the second most abundant natural polysaccharide next to cellulose, was proved to have broad-spectrum antifungal activity against a variety of fungi. However, the use of chitosan was limited due to its poor solubility and weaker antifungal activity compared with commercial fungicide. Therefore, in order to improve the water solubility and antifungal activity, many researchers grafted the active groups onto chitosan. The present work was to combine free metal ions with chitosan, to prepare more potent antifungal chitosan derivatives, thus, based on condensation reaction, chitosan derivative bearing amino pyridine group was prepared and subsequently followed by coordination with cupric ions, zinc ions and nickel ions to synthesize chitosan metal complexes. The calculations by density functional theory (DFT) show that the copper ions and nickel ions underwent dsp2 hybridization, the zinc ions underwent sp3 hybridization, and all of them are coordinated by the carbon atom in the p-π conjugate group and the oxygen atoms in the acetate ion. The antifungal properties of chitosan metal complexes against Phytophthora capsici (P. capsici), Gibberella zeae (G. zeae), Fusarium oxysporum (F. oxysporum) and Botrytis cinerea (B. cinerea) were also assayed. In addition, a plant toxicity experiment was carried out. The experiments indicated that the derivatives have significantly enhanced antifungal activity after metal ions complexation compared with the original chitosan. It was shown that 0.20 mg/mL of O-CSPX-Cu can 100% inhibit the growth of P. capsici and 0.20 mg/mL of O-CSPX-Ni can 87.5% inhibit the growth of B. cinerea. In general, their activities are better than the positive control oligosaccharides. The combination of the pyridine formyl groups seems to favor biological activity. Additionally, the ligand fashion was precisely analyzed, and the results revealed that the copper ions and nickel ions underwent dsp2 hybridization, the zinc ions underwent sp3 hybridization, and the carbon atoms of the p-π conjugate group and the oxygen atoms of acetate ion are involved in the coordination of metal ions. The phytotoxicity assay of O-CSPX-M was also conducted, unlike the traditional metal fungicides, the metal complexes were not significantly toxic to the leaves of wheat. O-CSPX-Zn can even increase chlorophyll content in wheat leaves at 0.40 mg/mL. This is mainly because chitosan itself promotes plant growth and counteracts the phytotoxicity of metal ions. The chitosan derivative described here may lend themselves to future applicative studies in crop protection.

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