



New conjugates of superoxide dismutase/catalase mimetics with cyclodextrins

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ABSTRACT

Mimetics of antioxidant enzymes such as superoxide dismutases (SOD) or catalases are reported as potential new drugs able to reduce oxidative stress damage. In particular, manganese(III) complexes of salen-type ligands have been studied as both SOD and catalase mimetics. In this paper, we report the synthesis of two novel conjugates of salen-type ligands with the β -cyclodextrin, the 6-deoxy-6-[(5-cyanoamidopropyl)(1,2-diamino)(*N,N'*-bis(salicylidene))]- β -cyclodextrin and the 6-deoxy-6-[(5-cyanoamidopropyl)(1,2-diamino)(*N,N'*-bis(3-methoxysalicylidene))]- β -cyclodextrin, their spectroscopic characterization, and the synthesis and the characterization of their manganese(III) complexes. The SOD-like activity of the metal complexes was investigated by the indirect Fridovich method. The catalase like activity was tested using a Clark-type oxygen electrode. The peroxidase activity was tested using the ABTS (2,2'-azino-bis-(3-ethylbenzothiazoline-6-sulfonic acid)) assay. The glycoconjugation of salen-manganese(III) complexes yields compounds with enhanced SOD activity. These complexes also show catalase and peroxidase activities higher than the simple salen complexes (EUK 813 and EUK 108).

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1. Introduction

Antioxidant compounds have been considered as therapeutic agents for a variety of disorders associated with the oxidative stress. In fact, oxidative stress processes have been implicated in a variety of detrimental health conditions including ischemia-reperfusion injury [1], chronic inflammation [2], diabetes [3], neurodegenerative disease [4,5], cancer [6] and aging [7].

In this perspective, mimetics of antioxidant enzymes such as superoxide dismutases (SODs) or catalases are presented as potential new drugs to prevent oxidative stress damages [2,8,9]. Among these systems, metal complexes of manganese(II) or (III) seem to be among the most widely investigated molecules [10]. Particularly, the manganese(III) complexes of [15]aneN₆ (1,4,7,10,13-pentaza-cyclopentadecane) type ligands [10–13] are reported in the literature as SOD mimetics and the manganese (III) complexes of porphyrins [14,15] or salen (H₂salen = *N,N'*-bis(salicylidene)-ethylenediamine) type ligands [9,16–18] have been reported as both SOD and catalase mimetics.

The molecular mechanism by which the manganese complexes work is still not entirely understood, but a number of different hypotheses based on experiments *in vivo* have been made. The SOD and catalase activities of these molecules have been considered as the main reason for their tested pharmacological activity. Investigations have been carried out in order to understand the action mechanism [19].

As regards the salen-manganese(III) complexes, generally the SOD-like activity, investigated *in vitro*, is considered to be the operative mechanism *in vivo* [8,20,21]. It has been shown that salen complexes exert beneficial effects in several *in vitro* and *in vivo* animal models of human diseases which involve oxidative stress ranging from Alzheimer's [22,23], Parkinson's [24], Prion Disease [25] and motor neuron disease to multiple sclerosis and excitotoxic neuronal injury [26]. The fluoranion compounds have also been shown to afford protection from ischemia-reperfusion injury of the brain [20], heart [27] and kidney [28,29] in animal models.

In the case of Mn(III) to salen derivatives, the mechanism for the dismutation of O₂^{•−} involves the reduction of Mn(III) to Mn(II) by O₂^{•−}, which is oxidized to O₂. The Mn(II) is oxidized back to Mn(III) by another molecule of O₂^{•−} yielding H₂O₂ [10]:



The mechanism by which the Mn-salen acts as a catalase mimetic involves the oxidation of Mn(III) to oxomanganese by H₂O₂, releasing water [20]. The oxomanganese is then reduced to Mn(III) by another hydrogen peroxide molecule to form oxygen and water:



Starting from the most simple system [Mn(salen)(X)] (X = Cl[−] or CH₃COO[−], code name EUK 8 and EUK 108, respectively), other modified salen complexes have been investigated (Chart 1). The water solubility of this class of molecules is not very high, as has been reported by several authors [10].

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