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Studies on the Preparation of Bioactive Oligomerstilbene by Oxidative Coupling Reaction (I) -Preparation of Shegansu B using Silver Oxide as Oxidant

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Abstract: The oxidative coupling reaction of isorhapontigenin using sliver oxide as oxidant afforded a major product, named shegansu B (2), which was isolated from the roots of *Belamcanda chinensis* (L.) DC. Both the natural and synthetic Shegansu B have the same potent antagonism activities of leukotriene B4, D4 receptor .

Keywords: Isorhapontigenin, oligomerstilbene**,** shegansu B, oxidative coupling reaction, silver oxide.

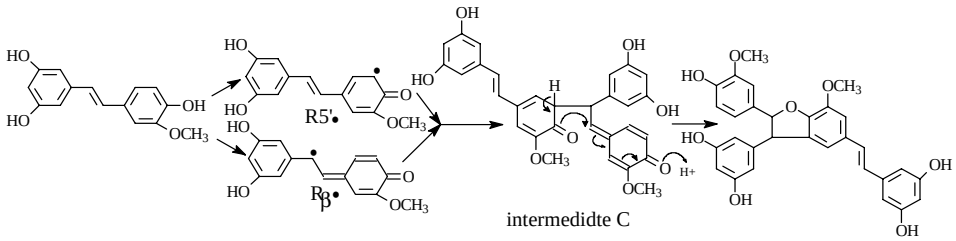
In previous paper[1], we reported a new dimer of isorhapontigenin, named shegansu B (2), which was isolated from the acetone soluble fraction of the ethanolic extract of the roots of *Belamcanda chinensis* (L.) DC. It showed potent antagonism activities for leukotriene B_4, D_4 receptor. Because shegansu B was obtained in small amount, to confirm its structure and pharmacological activities was difficult, thus we conceived that shegansu B might be obtained by oxidative coupling reaction with isorhapontigenin as starting material.

For the synthesis of shegansu B, we presumed that the route based on mimetic biosynthesis would be the most convenient. According to Melini's [2] and Baba's [3] methods, we assumed that shegansu B might be an oxidative dimerization product of isorhapontigenin using various oxidizing agents, such as iron(III) chlorid , potassium hexacyanoferrate-sodium acetate, silver oxide , hydrogen peroxide and peroxidase. In this paper we report the synthesis of shegansu B by oxidative coupling using silver oxide and isorhapontigenin as starting material. Isorhapontigenin was presumably converted into phenoxy radicals by oxidant (in **Scheme 1**), the intermediate C was obtained by coupling between the two radicals Rβ and R5', nucleophilic attack of C=O onto the quinous methine system of the intermediate C, would afford our target compound **2**.

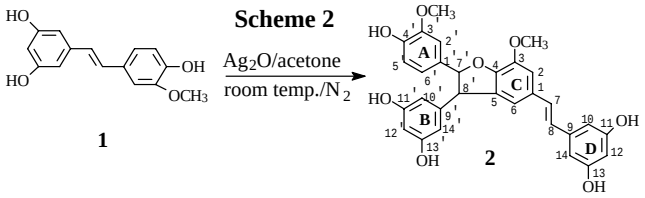
We now report the proceeding of the mimetic biosynthetic scheme. Isorhapontigenin (1) (159mg, 0.78mmol) was dissolved in aqueous acetone (10mL) and stirred with silver oxide (150mg, 0.58mmol) under N_2 at room temperature for 8 h, the

suspension was filtered, the filtrate was evaporated to dryness and the residue was chromatographed on preparative TLC ($CHCl_3$: MeOH: n-hexane: EtOAc: H_2O 7.5: 1.1: 1: 0.5:0.08) giving compound **2** as yellowish amorphous powder in a 30% yield. Its UV spectrum absorption maxima at 328 nm, indicated the characteristics of the hydroxystilbene; The molecular formula $C_{30}H_{26}O_8$ was determined by EI-MS (*m/z* 514 M+); ^{1}HNMR spectrum revealed the presence of two aromatic methoxyls at δ 3.82 (3H, s), 3.92 (3H, s); two methine groups at δ4.48 (1H, d, J=8.6Hz), 5.48 (1H, d, J=8.6 Hz); two *trans* coupling olefinic protons at δ6.89 (1H, d, J=16.3Hz), 7.00 (1H,d, J=16.3Hz); eleven aromatic protons at δ6.26-7.16, in which two signals of protons H-2 and H-6 (δ7.16 and 6.79) were attributed to *meta* coupling on ring C; the signals of protons H-5', H-6' , H-2' (δ6.81, 6.83, 7.02) belonged to an ABX system on ring A, the remaining six protons [H-12, H-10, H-14 (δ6.26, 6.51) and 12', 10', 14'(δ6.20, 6.23)] were attributed to two sets of AB_2 system (J=2 Hz) on ring D and B, respectively.

Scheme 1



Scheme 2



The above spectral data were completely identical with shegansu B from the natural source except optical value $[\alpha]_D^{25}$ +21 (c 0.048, EtOH), they have the same potent antagonism activities of leukotriene B4, D4 receptor.

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