

Title	Studies on the reduction products of streptomycin
Sub Title	
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Publisher	慶應義塾大学藤原記念工学部
Publication year	1970
Jtitle	Proceedings of the fujihara memorial f aculty of engineering keio university Vol.23, No.96 (1970.) ,p.174- 174
JaLC DOI	
Abstract	
Notes	Summaries of Doctoral Theses
Genre	Departmental Bulletin Paper
URL	https://koara.lib.keio.ac.jp/xoonips/modules/xoonips/detail.php?koara_id=KO50001004-00230096-0174

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Studies on the Reduction Products of Streptomycin

Kenji SHIROYANAGI (白柳健治)

1) The author has found that the reduction of streptomycin with amalgamated aluminum gives a new, medically useful compound, namely, dihydrodesoxystreptomycin, which is structurally different from the known dihydrostreptomycin obtained by catalytic hydrogenation of streptomycin. This finding was ascertained by the clear difference of the melting points of the new compound and of its derivatives as well as UV absorption spectrum of a decomposition product.

2) Since the hydrolysis of the new reduction product with hydrochloric acid gave streptidine and N-methyl-L-glucosamine, it became clear that a moiety structurally different from the dihydrostreptomycin exists on the streptose residue. Methanolysis of the new reduction compound gave a derivative which corresponds to dihydrostreptobiosaminide. The derivative was acetylated and then converted into a thiosugar with ethylmercaptane and hydrochloric acid. The thioethoxy group was removed from the thiosugar with mercuric chloride to give a product which corresponds to free dihydrostreptobiosamine. Oxidation of the product with bromine followed by hydrolysis with hydrochloric acid gave a monolactone compound. The product was proved to be 3, 5-dideoxy-3-C-hydroxymethyl-L-lyxono-r-lactone on the basis of NMR, IR and other data. The above-mentioned results led to the conclusion that the new product obtained by aluminum amalgam reduction is dihydrodesoxystreptomycin.

3) Reaction conditions for the production of dihydrodesoxystreptomycin by reduction of streptomycin with amalgamated aluminum was studied in detail. It has been found that pH of the reaction medium was important factor and that the highest yield of dihydrodesoxystreptomycin was given by reducing the streptomycin at a pH range of 2.2~2.4.

4) As a result of detailed studies on reaction to reduce streptomycin with amalgamated aluminum in acidic solution, it has been found that the highly pure dihydrodesoxystreptomycin was obtained by keeping the medium at pH 2.2~2.5 in the early stage and at pH 2.7~3.5 in the late stage at which the reaction was vigorous. Accordingly, it was not necessary to maintain the medium at pH 2.2~2.4 throughout the reaction. It was also found to be advantageous to carry out the reduction in an aqueous solution of aluminum sulfate. Further experiments on reaction showed that the best industrial procedure for producing the dihydrodesoxystreptomycin was to reduce the streptomycin sesquisulfate in an aqueous solution with amalgamated aluminum, adjusting the pH of the medium with phosphoric acid as described above and followed by removal of the excess phosphoric acid by neutralization with calcium hydroxide to pH 7.5 and further removal of a part of sulfuric acid by addition of barium hydroxide to pH 10.0 to afford a crystalline monosulfate of dihydrodesoxystreptomycin.