

ALIEN PROPERTY CUSTODIAN

CONDENSATION PRODUCTS

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This invention relates to condensation products of 4,4'-diaminodiphenylsulphone and to a process of preparing the same.

4,4'-diaminodiphenylsulphone and 4,4'-diacetyldiamino-diphenylsulphone have a good activity against streptococci and pneumococci infections of the mouse, the utility of the 4,4'-diaminodiphenylsulphone for this purpose is however decreased by the fact that the said product gives rise to the formation of methemoglobin.

In accordance with the present invention new products are obtained which show a considerably increased activity when compared with the afore-mentioned products and which do not further give rise to the formation of methemoglobin. Applicants have found that the condensation products of 4,4'-diaminodiphenylsulphone with aldehydes are free from the disadvantages of the afore-mentioned compounds. The new condensation-products may contain 1 or 2 molecules of the aldehyde which are combined with one or both amino-groups of the 4,4'-diaminodiphenylsulphone. The condensation takes place while splitting off one molecule of water upon each molecule of aldehyde. In general applicants' new condensation products probably have the structure of the so called Schiff's bases, that is, in the condensation product the carbon atom of the aldehyde-group is combined with the amino-group(s) of the diaminodiphenylsulphone by means of a double bond.

In accordance with the present invention the new condensation products are obtainable by heating 4,4'-diaminodiphenylsulphone with an aldehyde. The most various aldehydes have proved useful for this process, for instance, aliphatic aldehydes, aromatic aldehydes, such as aldehydes of the benzene and naphthalene series, furthermore mixed aliphatic aromatic aldehydes and heterocyclic aldehydes. Depending on the activity of the aldehyde-group the condensation products contain one or two aldehyde radicals. Applicants have found that the condensation often may be facilitated by the presence of catalysts; 4,4'-diaminodiphenylsulphone-hydrohalides which sometimes are contained to a slight extent in the 4,4'-diaminodiphenylsulphone of technical manufacture, for instance, have proved operable as such catalysts, furthermore ammonium halides, hydrohalides of tertiary amine, such as triethylamine, pyridine. When using such catalysts often condensation takes place with 2 molecules of the aldehyde; 2 molecules of aldehyde are also condensed in general when the reaction mixture is heated to melting, for in-

stance, to a temperature of about 150 to about 180° C. In other cases it is advisable to heat the reaction mixture in the presence of a solvent, particularly in the presence of an alcohol, such as methyl, ethyl, isoamyl alcohols and the like. The condensation can also be effected in stages, for instance, by first using for the condensation a slightly active aldehyde, so that a mono-condensation product is formed which subsequently is subjected to further condensation with a more active aldehyde or under the more effective reaction conditions referred to above. In this manner also condensation products are obtainable containing two different aldehyde radicals.

The invention is further illustrated by the following examples without being restricted thereto:

Example 1.—24,8 grs of 4,4'-diaminodiphenylsulphone are heated to boiling with 30 grs of cinnamaldehyde in 300 ccms of alcohol during 15 minutes. The condensation product begins to separate already in the boiling heat. After cooling it is filtered with suction and washed with alcohol and ether. In this way the 4,4'-bis-(cinnamylidenamino)-diphenylsulphone is obtained. It forms light yellow leaflets melting at 236° C. In a corresponding manner there are obtained when using:

Propionaldehyde the 4,4'-bis-propylidenamino-diphenylsulphone melting at 246° C.

Benzaldehyde the 4-benzylidenamino-4'-aminodiphenylsulphone melting at 214° C.,

4-tolylaldehyde the 4,4'-bis-(4''-methylbenzylidenamino)-diphenylsulphone melting at 250° C.,

2-hydroxybenzaldehyde the 4,4'-bis-(2''-hydroxybenzylidenamino)-diphenylsulphone melting at 259° C.,

4-methoxybenzaldehyde the 4-(4''-methoxybenzylidenamino)-4'-amino-diphenylsulphone melting at 226° C.,

4-chlorobenzaldehyde the 4,4'-bis-(4''-chlorobenzylidenamino)-diphenylsulphone melting at 233° C.,

3,4-methylendioxybenzaldehyde the 4-(3'',4''-methylendioxybenzylidenamino)-4'-amino-diphenylsulphone melting at 227° C.

The melting points of the afore-mentioned condensation-products may be different corresponding to the state of purity of the starting material; a further purification of the products often is not possible because of their insolubility in organic solvents.

The condensation often is favourably influenced by using technical diaminodiphenylsulphone which still contains some small quantities

of its dihydrochloride. In this case, for instance, with 3,4-methylenedioxy-benzaldehyde directly the 4,4'-bis-(3'',4''-methylenedioxybenzylidenamino) - diphenylsulphone is obtained, melting at 231° C.

In the same manner also other aldehydes, for instance, β -hydroxypropionaldehyde, glycerolaldehyde, valerianaldehyde, butyraldehyde, chaulmoogrylaldehyde, nitrobenzaldehydes, vanillin, acetylaminobenzaldehyde, phenylacetaldehyde, resorcinolaldehyde, phloroglucinaldehyde, terephthalic aldehyde-acid, 1-hydroxynaphthalene-4-aldehyde, 2-hydroxynaphthalene - 1 - aldehyde, glucose, galactose, maltose, furfural, α -furyl-acrolein, pyridine-3-aldehyde, 2-methylindol-3-aldehyde, quinoline-2-aldehyde may be reacted.

Example 2.—10 grs of 4,4'-diaminodiphenylsulphone and 25 grs of 4-chlorobenzaldehyde are heated to 170–180° C. while slowly passing through a current of nitrogen until the water split off during the condensation is distilled off which takes about 10 minutes. The cold melt is triturated and boiled with ether whereby the 4,4'-bis-(4'' - chlorobenzylidenamino) - diphenylsul-

phone remains undissolved. It is sucked off; it melts at 233° C. The yield amounts to 90% of the theory.

In the same way when using benzaldehyde there is obtained the 4,4'-bis-benzylidenamino-diphenylsulphone melting at 236° C., when using 4-methoxybenzaldehyde the 4,4'-bis-(4''-methoxybenzylidenamino)-diphenylsulphone melting at 241° C.

Example 3.—10 grs of the 4-(4''-methoxybenzylidenamino)-4' - amino - diphenylsulphone melting at 226° C. obtained as per Example 1) are heated to 170–180° C. with 20 grs of 4-methoxybenzaldehyde while slowly passing through a current of nitrogen until the reaction-water is distilled off. The melt is poured into hot alcohol while stirring the precipitate filtered with suction and washed with alcohol and ether. The 4,4'-bis-(4''-methoxybenzylidenamino)-diphenylsulphone obtained forms colorless crystals melting at 240° C.

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