

# PATENT SPECIFICATION

NO DRAWINGS

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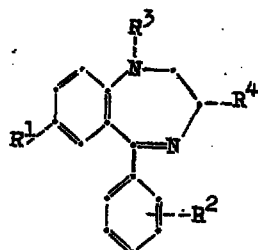
## COMPLETE SPECIFICATION

### Benzylideneamino Compounds and the Conversion thereof into Benzdiaz[1,4]Epine Derivatives

We, F. HOFFMANN-LA ROCHE & CO., AKTIENGESELLSCHAFT, a Swiss Company of 124—128 Grenzacherstrasse, Basle, Switzerland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to benzylideneamino compounds and the conversion thereof into benzdiaz[1,4]epine derivatives. More particularly, it is concerned with a process for the manufacture of benzdiaz[1,4]epine derivatives from benzylideneamino compounds, with said benzylideneamino compounds *per se* and with a process for the preparation thereof.

The benzdiaz[1,4]epine derivatives obtained according to this invention are compounds of the general formula



(I)

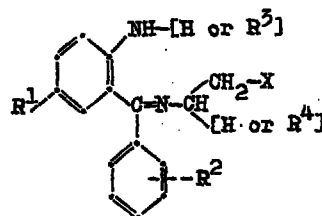
wherein R<sup>1</sup> represents a halogen atom (preferably a chlorine or bromine atom) or the nitro or trifluoromethyl group, R<sup>2</sup> represents a halogen atom and R<sup>3</sup> and R<sup>4</sup> each represent a lower-alkyl group (preferably a methyl group) and wherein the broken lines denote that the groups or atoms attached thereby are optional, and acid addition salts thereof.

It will be appreciated that the term "lower alkyl" is used in this description and in the claims appended hereto to mean both straight-

chain and branched-chain alkyl groups containing from one to seven carbon atoms (e.g. methyl, ethyl and the like). The term "halogen" means chlorine, bromine, fluorine and iodine.

One interesting class of benzdiaz[1,4]epine derivatives obtained by carrying out the present process comprises compounds of formula I in which R<sup>2</sup> and R<sup>4</sup> are absent, and acid addition salts thereof. A preferred class of benzdiaz[1,4]epine derivatives obtained by carrying out the present process comprises compounds of formula I in which R<sup>2</sup> and R<sup>4</sup> are absent and R<sup>1</sup> is present and represents a chlorine or bromine atom, and acid addition salts thereof. The most preferred benzdiaz[1,4]epine derivatives obtained by carrying out the present process are compounds of formula I in which R<sup>2</sup> and R<sup>4</sup> are absent, R<sup>1</sup> is present and represents a chlorine atom and R<sup>3</sup> (when present) represents the methyl group, and acid addition salts thereof.

According to the invention, the benzdiaz[1,4]epine derivatives aforesaid are manufactured by cyclising a benzylideneamino compound of the general formula



(II)

wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup> and the broken lines have the significance given earlier and X stands for a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group, and, if desired, converting the cyclisation product into an acid addition salt.

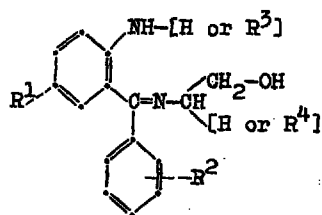
[Price 4s. 6d.]

In order to bring about cyclisation of a benzylideneamino compound of formula II to yield a corresponding benzdiaz[1,4]epine compound of formula I, the benzylideneamino compound may be heated, preferably at a temperature within an approximate range of from 50°C to 250°C. In one embodiment an acid acceptor is present when a benzylideneamino compound of formula II is heated. If an acid acceptor is used, an excess thereof may be provided whereby it can also serve as the medium in which the cyclisation occurs. By this simple expedient a single substance can be provided to serve a two-fold purpose; namely as acid acceptor and cyclisation medium. However, the cyclisation is preferably carried out by simply heating a benzylideneamino compound of formula II in the absence of a solvent medium and/or an acid acceptor, preferably with agitation. In another, but less preferred embodiment the cyclisation can be carried out in the presence of an inert organic solvent. Examples of inert organic solvents which can be used in this embodiment are xylene, toluene, halogenated aromatic hydrocarbons such as chlorobenzene, N,N - dimethylformamide, dimethylsulphoxide, quinoline, lower - alkanols such as methanol, ethers such as bis(2 - methoxy - ethyl) - ether and any other suitable inert organic solvent. The cyclisation is preferably carried out at a temperature within an approximate range of from 50°C to 250°C.

The benzylideneamino compounds of formula II occur in two isomeric forms; namely, the *syn* - form and the *anti* - form. They can be resolved into their isomeric forms and each of the so-obtained stereoisomers can be cyclised to the compounds of formula I as described earlier. Alternatively, without resolution, a mixture of both stereoisomers can be cyclised as described earlier.

The benzylideneamino compounds of formula II, which are useful in the manufacture of the pharmaceutically valuable benzdiaz[1,4]epine derivatives aforesaid, are novel and it will be appreciated that both they and their preparation also form part of the present invention.

The benzylideneamino compounds of formula II hereinbefore are prepared in accordance with the invention by replacing the hydroxyl group of a compound of the general formula



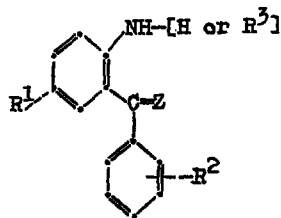
(III)

wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup> and the broken lines have the significance given earlier, by a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group by treatment with an agent which provides said atom or group.

Advantageously, X in the benzylideneamino compounds of formula II stands for a chlorine or bromine atom. The replacement of the terminal hydroxyl group of a compound of formula III to yield a benzylideneamino compound of formula II in which X stands for a bromine or chlorine atom is advantageously carried out by treating a compound of formula III with an agent such as thionyl chloride or phosphorus tribromide. This treatment is preferably carried out in the presence of an inert organic solvent such as methylene chloride and at elevated temperatures (e.g. at about the reflux temperature of the mixture). Other agents which can be used to replace the terminal hydroxy group of a compound of formula III by a bromine or chlorine atom are, for example, thionyl bromide, phosphorus oxychloride and phosphorus pentachloride.

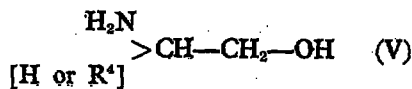
X in the benzylideneamino compounds of formula II can also stand for an alkylsulphonyloxy group (e.g. mesyloxy) or an arylsulphonyloxy group (e.g. tosyloxy). Suitably, agents such as *paratoluene* - sulphonyl chloride and methanesulphonyl chloride can be used to replace the hydroxyl group of a compound of formula III by the tosyloxy or mesyloxy group. It will be appreciated that when X in the benzylideneamino compounds of formula II is other than halogen, for example, mesyloxy or tosyloxy, this replacement, in practice, involves the prior formation of an alkali - metal salt of the compound of formula III in order that the site of the hydroxy group is preferentially acylated.

The compounds of formula III hereinbefore can, in turn, be obtained by reacting a compound of the general formula



(IV)

wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> and the broken lines have the significance given earlier and Z stands for an oxygen atom or an =NH or =N - (lower alkyl) group (preferably an oxygen atom or an =NH group), with an amino alcohol of the general formula



(V)

wherein  $R^4$  has the significance given earlier.

The reaction of a compound of formula IV with an amino - alcohol of formula V can be carried out at any suitable temperature and pressure and, thus, reaction conditions are not critical. However, in a preferred embodiment the reaction is carried out at elevated temperatures (e.g. at about the reflux temperature of the reaction mass). Thus, in one embodiment, a compound of formula IV above is heated with an amino - alcohol of formula V. The amino - alcohol of formula V can be used in excess whereby it can serve as the solvent medium. However, in a less preferred embodiment, the reaction is carried out in the presence of a solvent medium such as xylene or a higher - alkanol such as hexanol. Of course, the temperature employed in either embodiment should not be so high as to result in loss of the solvent medium and/or in decomposition of the reactants or resulting product.

The compounds of formula I hereinbefore and their acid addition salts are claimed *per se* in the specification of our Letters Patent No. 1,017,276.

The following Examples illustrate the invention:

#### EXAMPLE 1

A) The preparation of the starting material:  
60.0 g (0.258 mol) of 2 - amino - 5 - chlorobenzophenone in 400 ml (400 g, 6.66 mol) of ethanol amine were stirred and heated at reflux for 4 hours. The solution thus obtained was cooled and poured into 800 ml of water and the resulting precipitate was recovered by filtration. The solid was dissolved in methylene chloride and the solution was washed with water, dried over sodium sulphate and evaporated to give 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol as a mixture of brown gum and crystals. This mixture was extracted several times with hot hexane, then the insoluble residue was recrystallized from aqueous ethanol to give 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenylbenzylideneamino) - ethanol as tan-coloured crystals of melting point 118°—120°C.

A solution of 2.75 g (10 mmol) of 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol in 20 ml of methylene chloride and 1 drop of pyridine was stirred in an ice-bath and treated dropwise over a period of 1 hour, while protecting from atmospheric moisture, with a solution of 1.8 ml (3.0 g, 25 mmol) of thionyl chloride in 10 ml of methylene chloride. The mixture was then stirred and heated at reflux for 2 hours and subsequently evaporated *in vacuo*. The resulting yellow crystalline residue (3.79 g) was added carefully to a mixture of an excess of ice-cold 1-N sodium hydroxide solution and methylene chloride. The organic layer was washed with water, dried over sodium sulphate and evaporated to give a pale brown partially crystalline residue. Recrystallization from hexane and

from ether/pentane gave 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino)-ethyl chloride as yellow prisms of melting point 71°—73°C.

#### B) The process:

(i) 0.25 g of 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride contained in an open test-tube were immersed in an oil-bath at 150°C and heated, with stirring, for 20 minutes. The material melted and then re-solidified to give crude 7 - chloro - 5 - phenyl - 2,3 - dihydro - 1H - benzodiaz[1,4]epine hydrochloride as an orange solid. This hydrochloride was converted to the corresponding free base by treatment with dilute sodium hydroxide solution and extraction with methylene chloride, giving 7 - chloro - 5 - phenyl - 2,3 - dihydro - 1H - benzodiaz[1,4]epine as a yellow gum; trituration with acetone afforded yellow prisms of melting point 172°—173°C.

(ii) 0.29 g (1 mmol) of 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride in 5 ml of xylene were stirred and heated at reflux for 18.5 hours, then the solvent was evaporated *in vacuo*. The residue was extracted with a mixture of dilute hydrochloric acid and ether. The aqueous layer was made basic with dilute sodium hydroxide and extracted with methylene chloride. Evaporation of the methylene chloride extract gave crude 7 - chloro - 5 - phenyl - 2,3 - dihydro - 1H - benzodiaz[1,4]epine as a yellow gum which crystallized on trituration with acetone to give yellow prisms melting at 169°—171°C.

(iii) 0.29 g (1 mmol) of 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino)-ethyl chloride in 5 ml of quinoline was stirred and heated in an oil-bath at 150°C for 1.5 hours, then the mixture was poured into 50 ml of water. The crude product was recovered by extraction with methylene chloride and was obtained as a brown oil which was further purified in the manner described in part B(ii) of this Example to yield 7 - chloro - 5 - phenyl - 2,3 - dihydro - 1H - benzodiaz[1,4]epine of melting point 170°—172°C.

(iv) 0.29 g (1 mol) of 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride in 10 ml of ethanol was treated with 0.2 g (1.9 mmol) of sodium carbonate and the resulting mixture was stirred and heated at reflux for 18 hours. The product was isolated in the manner described in part B(ii) of this Example and was obtained as a yellow gum. Trituration with acetone afforded 7 - chloro - 5 - phenyl - 2,3 - dihydro - 1H - benzodiaz[1,4]epine of melting point 169°—172°C.

#### EXAMPLE 2

A) The preparation of the starting material:  
20.0 g (0.813 mol) of 2 - methylamino - 5 - chlorobenzophenone in 200 ml (200 g, 3.33 mol) of ethanol amine were stirred and heated

at reflux for 4 hours, then cooled and poured into 2 litres of water. The resulting oily precipitate was washed with water several times by decantation and was then dissolved in methylene chloride. The solution was washed with water, dried over sodium sulphate and evaporated to give 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol as a very viscous yellow oil.

10.77 g (37.3 mmol) of 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol (as a yellow oil) in 100 ml of methylene chloride and one drop of pyridine were treated with a solution of 6.65 ml (11.0 g, 93 mmol) of thionyl chloride in 25 ml of methylene chloride. 2 - (2 - Methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride was isolated by a procedure analogous to that described in the second paragraph of Example 1A for the isolation of 2 - 2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride. The crude product was obtained as a dark - coloured oil. This oil was dissolved in benzene and the solution was filtered through a column of 200 g of neutral alumina (WOELM, activity I). Evaporation of the filtrate gave 2 - (2-methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride which was obtained as yellow prisms of melting point 75°—76°C after recrystallization from hexane and from ether/pentane.

#### B) The process:

2 - (2 - Methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride was heated, with stirring, in a large test-tube immersed in an oil-bath at 150°—155°C until the melt largely solidified (after 10 minutes) to give crude 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine hydrochloride as an orange solid. This hydrochloride was converted to the corresponding free base by treatment with dilute aqueous sodium hydroxide and extraction with methylene chloride giving crude 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine as a light brown gum. The crude product was purified by filtration of a benzene solution thereof through 10 g of neutral alumina (WOELM, activity III). Evaporation of the filtrate gave an orange gum which was recrystallized from hexane to give purified 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine as pale yellow prisms of melting point 96°—98°C.

#### EXAMPLE 3

##### A) The preparation of the starting material:

3.45 g (14.1 mmol) of 2 - methylamino - 5 - chlorobenzophenone imine in 20 ml (20 g, 328 mmol) of ethanol amine were stirred and heated at reflux for 2 hours. The mixture was then cooled and poured into water. The resulting sticky precipitate was washed with

water by decantation and dissolved in methylene chloride. The solution was washed with water, dried over magnesium sulphate and evaporated to give 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol as a viscous yellow oil. This free base was treated with methanolic hydrogen chloride according to conventional techniques to give the corresponding hydrochloride as orange prisms of melting point 212°—214°C.

The conversion of 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol into 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride may be effected as described in the second paragraph of Examples 1B and 2B.

#### B) The process:

(i) 1 g (3.26 mmol) of 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride in 10 ml of N,N - dimethylformamide was heated in an oil-bath at 120°C for 17 hours and then at 150°C for 20 minutes. The resulting mixture was cooled, poured into dilute sodium carbonate solution and extracted with methylene chloride. The extract was evaporated and the residue extracted with a mixture of dilute hydrochloric acid and ether, which was allowed to stand overnight at room temperature to hydrolyse any unreacted starting material. The aqueous layer was made basic with dilute sodium hydroxide solution, and extraction with ether afforded the product as a dark-brown gum. This gum was dissolved in benzene and the solution was filtered through a column of 3 g of neutral alumina (WOELM, activity III). Evaporation of the filtrate gave crude 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine as an orange gum. Trituration with hexane afforded orange crystals of 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine of melting point 94°—98°C.

(ii) The procedure described in part B(i) of this Example was repeated using 10 ml of dimethyl sulphoxide instead of N,N-dimethylformamide. Isolation and purification of the product in the same manner as that described in part B(i) of this Example gave 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz[1,4]epine as an orange brown gum. Trituration with hexane containing a small amount of ether afforded orange-tan coloured crystals of melting point 95°—98°C.

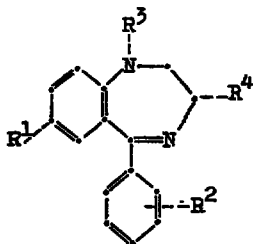
(iii) 0.69 g (6.5 mmol) of sodium carbonate was added to a solution of 2 g (6.5 mmol) of 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride in 20 ml of bis(2 - methoxy - ethyl) ether and the resulting mixture was stirred and heated at reflux for 42 hours. Isolation and purification of the product in the manner described in part B(i) of this Example gave 7 - chloro - 1 - methyl - 5 - phenyl - 2,3 - dihydro - 1H - benzdiaz

[1,4]epine as a dark red oil. Recrystallization from hexane afforded yellow prisms of melting point 96°—98°C.

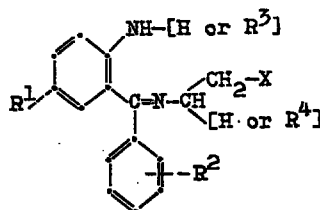
- 5 Having regard to the provision of Section 9 of the Patents Act, 1949, attention is directed to the claims of Patents Nos. 1,056,423 and 1,117,062.

WHAT WE CLAIM IS:—

- 10 1) A process for the manufacture of compounds of the general formula



- 15 wherein R¹ represents a halogen atom or the nitro or trifluoromethyl group, R² represents a halogen atom and R³ and R⁴ each represent a lower-alkyl group and wherein the broken lines denote that the atoms or groups attached thereby are optional, and acid addition salts thereof, which process comprises cyclising a benzylideneamino compound of the general formula



- 25 wherein R¹, R², R³, R⁴ and the broken lines have the significance given earlier in this claim and X stands for a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group and, if desired, converting the cyclisation product into an acid addition salt.

- 30 2) A process as set forth in claim 1, wherein the cyclisation is effected by the application of heat.

- 3) A process as set forth in claim 2, wherein the cyclisation is effected by heating the benzylideneamino compound to a temperature between 50°C and 250°C.

- 35 4) A process as set forth in claim 1, claim 2 or claim 3, wherein a benzylideneamino compound in which X stands for a chlorine or bromine atom is used as the starting material.

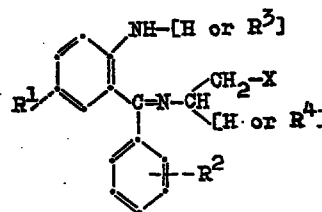
- 40 5) A process as set forth in claim 4, wherein 2 - (2 - methylamino - 5 - chloro - α - phenyl - benzylideneamino) - ethyl chloride is used as the starting material.

- 6) A process as set forth in claim 4, wherein 2 - (2 - amino - 5 - chloro - α - phenyl - benzylideneamino) - ethyl chloride is used as the starting material.

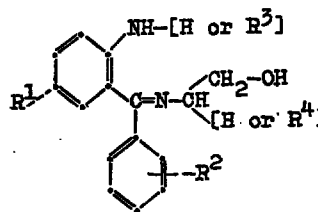
- 7) A process for the manufacture of the benzdiaz[1,4]epine derivatives set forth in claim 1, substantially as described with reference to part B of the foregoing Examples.

- 8) Benzdiaz[1,4]epine derivatives as set forth in claim 1, when manufactured by the process claimed in any one of claims 1 to 7 inclusive.

- 9) A process for the preparation of benzylideneamino compounds of the general formula



- wherein R¹, R², R³, R⁴, X and the broken lines have the significance given in claim 1, which process comprises replacing the hydroxyl group of a compound of the general formula



- wherein R¹, R², R³, R⁴ and the broken lines have the significance given in claim 1, by a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group by treatment with an agent which provides said atom or group.

- 10) A process as set forth in claim 9, wherein said agent is thionyl chloride, thionyl bromide, phosphorus oxychloride, phosphorus pentachloride, phosphorus tribromide, *para*-toluene - sulphonyl chloride or methanesulphonyl chloride.

- 11) A process as set forth in claim 9, wherein the hydroxyl group is replaced by a chlorine or bromine atom.

- 12) A process as set forth in claim 9, wherein 2 - (2 - methylamino - 5 - chloro - α - phenyl - benzylideneamino) - ethanol is used as the starting material.

- 13) A process as set forth in claim 9, wherein 2 - (2 - amino - 5 - chloro - α - phenyl - benzylideneamino) - ethanol is used as the starting material.

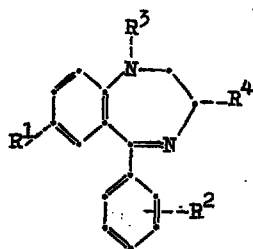
- 14) A process for the preparation of the

[1,4]epine as a dark red oil. Recrystallization from hexane afforded yellow prisms of melting point 96°—98°C.

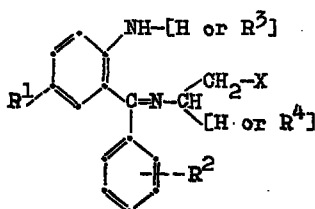
Having regard to the provision of Section 9 of the Patents Act, 1949, attention is directed to the claims of Patents Nos. 1,056,423 and 1,117,062.

WHAT WE CLAIM IS:—

1) A process for the manufacture of compounds of the general formula



wherein R<sup>1</sup> represents a halogen atom or the nitro or trifluoromethyl group, R<sup>2</sup> represents a halogen atom and R<sup>3</sup> and R<sup>4</sup> each represent a lower-alkyl group and wherein the broken lines denote that the atoms or groups attached thereby are optional, and acid addition salts thereof, which process comprises cyclising a benzylideneamino compound of the general formula



wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup> and the broken lines have the significance given earlier in this claim and X stands for a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group and, if desired, converting the cyclisation product into an acid addition salt.

2) A process as set forth in claim 1, wherein the cyclisation is effected by the application of heat.

3) A process as set forth in claim 2, wherein the cyclisation is effected by heating the benzylideneamino compound to a temperature between 50°C and 250°C.

4) A process as set forth in claim 1, claim 2 or claim 3, wherein a benzylideneamino compound in which X stands for a chlorine or bromine atom is used as the starting material.

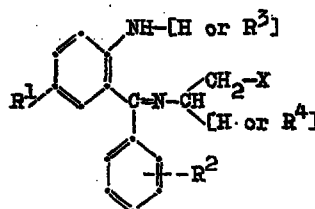
5) A process as set forth in claim 4, wherein 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride is used as the starting material.

6) A process as set forth in claim 4, wherein 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride is used as the starting material.

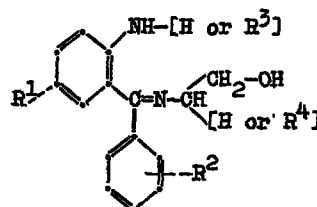
7) A process for the manufacture of the benzdiaz[1,4]epine derivatives set forth in claim 1, substantially as described with reference to part B of the foregoing Examples.

8) Benzdiaz[1,4]epine derivatives as set forth in claim 1, when manufactured by the process claimed in any one of claims 1 to 7 inclusive.

9) A process for the preparation of benzylideneamino compounds of the general formula



wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup>, X and the broken lines have the significance given in claim 1, which process comprises replacing the hydroxyl group of a compound of the general formula



wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup> and the broken lines have the significance given in claim 1, by a chlorine or bromine atom or an alkylsulphonyloxy or arylsulphonyloxy group by treatment with an agent which provides said atom or group.

10) A process as set forth in claim 9, wherein said agent is thionyl chloride, thionyl bromide, phosphorus oxychloride, phosphorus pentachloride, phosphorus tribromide, *para*-toluene - sulphonyl chloride or methanesulphonyl chloride.

11) A process as set forth in claim 9, wherein the hydroxyl group is replaced by a chlorine or bromine atom.

12) A process as set forth in claim 9, wherein 2 - (2 - methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol is used as the starting material.

13) A process as set forth in claim 9, wherein 2 - (2 - amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethanol is used as the starting material.

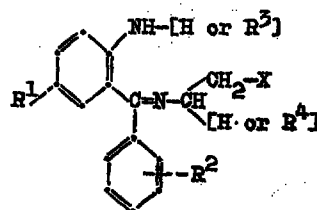
14) A process for the preparation of the



benzylideneamino compounds of the formula first written in claim 9, substantially as described with reference to part A of the Examples.

- 5 15) Benzylideneamino compounds of the formula first written in claim 9, when prepared by the process claimed in any one of claims 9 to 14 inclusive.

- 10 16) A benzylideneamino compound of the general formula



wherein  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $X$  and the broken lines have the significance given in claim 1.

- 17) A benzylideneamino compound as formulated in claim 16, wherein  $R^1$  is present and represents a chlorine or bromine atom.

- 18) A benzylideneamino compound as formulated in claim 16 or as set forth in claim 17, wherein  $R^3$  and  $R^4$  (when present) each represent a methyl group.

- 19) A benzylideneamino compound as formulated in claim 16 or as set forth in claim 17 or claim 18, wherein  $X$  stands for a chlorine or bromine atom.

- 20) 2 - (2 - Methylamino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride.

- 21) 2 - (2 - Amino - 5 - chloro -  $\alpha$  - phenyl - benzylideneamino) - ethyl chloride.

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