Our Ref: 466943

663580
AUSTRALIA

P/00/001
Section 29

Patents Act 1990

PATENT REQUEST : STANDARD PATENT

I/We, being the person/s identified below as the Applicant, request the grant of a patent to the person/s indicated below as the Nominated Person/s, for an invention described in the accompanying standard complete specification.

Full application details follow.

[71] [70] Applicant/s and Nominated Person/s:
**Ciba-Geigy AG,**
**of Klybeckstrasse 141, CH-4002 BASLE, SWITZERLAND**

[54] Invention Title: Preparation of Iminostilbene by
~~**Novel**~~ **high-temperature dehydrogenation** ~~**process**~~

[72] Name/s of actual inventor/s: (optional)

[74] Address for service in Australia:
**DAVIES COLLISON CAVE**, Patent Attorneys
10 Barrack Street, SYDNEY NSW 2000 Attorney Code : CA

BASIC CONVENTION APPLICATION/S DETAILS:

| [31] Appln No.: | [33] Country: | Code: | [32] Date: |
|---|---|---|---|
| 1535/92-0 | SWITZERLAND | CH | 13 May 1992 |

DATED this TWENTY SECOND day of APRIL 1993

Ciba-Geigy AG
By Patent Attorneys
DAVIES COLLISON CAVE

Peter Stearne

Dr. PETER STEARNE, FIPAA

Fee: $ 341.00

AUSTRALIAN PATENT OFFICE

S 037002 220493

5010

P/00/008 28/5/91
Section 29(1)
Regulation 3.2(2)

AUSTRALIA

*Patents Act 1990*

**NOTICE OF ENTITLEMENT**

We, **CIBA-GEIGY AG** of Klybeckstrasse 141, 4002 Basle, Switzerland, the Applicant in respect of Australian Application No. 37111/93 state the following:

1. Ciba-Geigy AG is the Nominated Person in respect of the application.

2. The actual inventors of the invention, the subject of the application, are Beat M Aebli, Dr Daniel Monti and Milos Rusek.

3. The Nominated Person, Ciba-Geigy AG, is entitled to the grant of a patent in respect of the application because the said Nominated Person derived title to the invention from the actual inventors by Assignment.

4. The Nominated Person is entitled to claim priority from the basic application listed on the patent application form because (i) the Nominated Person is the applicant in respect of the basic application; and (ii) the basic application was the first application made in a convention country in respect of the invention the subject of the application.

DATED this 27th day of June, 1995.

**CIBA-GEIGY AG**
By Its Patent Attorneys,
DAVIES COLLISON CAVE

**JAMES G SIELY, FIPAA**

p:\wpdocs\grs\466943\jgs

AU9337111

(12) PATENT ABRIDGMENT (11) Document No. AU-B-37111/93
(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 663580

(54) Title
**PREPARATION OF IMINOSTILBENE BY HIGH-TEMPERATURE DEHYDROGENATION**

International Patent Classification(s)
(51)[5] C07D 233/22

(21) Application No. : **37111/93** (22) Application Date : **22.04.93**

(30) Priority Data

| (31) Number | (32) Date | (33) Country |
|---|---|---|
| 1535/92 | 13.05.92 | CH SWITZERLAND |

(43) Publication Date : **18.11.93**

(44) Publication Date of Accepted Application : **12.10.95**

(71) Applicant(s)
**CIBA-GEIGY AG**

(72) Inventor(s)
**BEAT M AEBLI; DR DANIEL MONTI; MILOS RUSEK**

(74) Attorney or Agent
**DAVIES COLLISON CAVE , GPO Box 3876, SYDNEY NSW 2001**

(56) Prior Art Documents
**US 3449324**
**US 3531466**
**GB 1077648**

(57) The invention relates to a novel process for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapor phase.

Iminostilbene, 5H-dibenz[b,f]azepine, is an important intermediate for the preparation of 5-carbamoyl-dibenz[b,f]azepine which under the generic name carbamazepine has gained great importance as an anti-convulsive active ingredient in medicaments. A number of processes are known for the conversion of iminostilbene into carbamazepine, which processes are based on the direct or stepwise introduction of the carbamoyl group in the 5-position of the azepine ring.

**Claim**

1. A process for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapour phase, wherein there is used an iron oxide/potassium salt contact catalyst comprising from 44 to 85 % by weight of an iron compound, calculated as $Fe_2O_3$, and from 7 to 35 % by weight of a potassium compound, calculated as $K_2O$, in addition to from 0.0 to 3.5 % by weight of a chromium compound, calculated as $Cr_2O_3$, and from 0.0 to 3.0 % of a calcium compound, calculated as CaO, and optionally customary promotors, and wherein the reaction is carried out at a temperature of 500° to 600° C.

Our Ref: 466943

P/00/011
Regulation 3:2

663580

AUSTRALIA

Patents Act 1990

**ORIGINAL**
**COMPLETE SPECIFICATION**
**STANDARD PATENT**

Applicant(s): Ciba-Geigy AG
Klybeckstrasse 141
CH-4002 BASLE
SWITZERLAND

Address for Service: DAVIES COLLISON CAVE
Patent & Trade Mark Attorneys
Level 10, 10 Barrack Street
SYDNEY NSW 2000

Invention Title: ~~Novel~~ Preparation of Iminostilbene by high-temperature dehydrogenation ~~process~~

The following statement is a full description of this invention, including the best method of performing it known to me:-

AUSTRALIAN PATENT OFFICE

5020

4-19088/A
Australia

Process for the Preparation of Iminostilbene by High-temperature Dehydrogenation of Iminodibenzyl

The invention relates to a novel process for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapor phase.

Iminostilbene, 5H-dibenz[b,f]azepine, is an important intermediate for the preparation of 5-carbamoyl-dibenz[b,f]azepine which under the generic name carbamazepine has gained great importance as an anti-convulsive active ingredient in medicaments. A number of processes are known for the conversion of iminostilbene into carbamazepine, which processes are based on the direct or stepwise introduction of the carbamoyl group in the 5-position of the azepine ring.

Numerous processes are available for the preparation of iminostilbene, amongst which high-temperature dehydrogenation of iminodibenzyl in the vapor phase on iron oxide contacts has achieved particular importance. According to the variant of that process having the greatest technical importance at present, the variant according to US-3,449,324, there is used a contact catalyst comprising "in addition to iron(III) oxide, which is best used in amounts of from 30 to 70 %, preferably also additions of, for example, from 1.5 to 3 % by weight $Cr_2O_3$, from 10 to 15 % by weight CaO and the remainder $K_2CO_3$" and the operation is carried out "at temperatures of from 300° to 500°C, preferably from 400° to 450°C". As expressly pointed out, and supported by numerical data for reaction temperatures of from 550° and 600°C, in US-3,449,324, temperatures higher than 500°C result "to an unacceptable extent or even predominantly" in undesired acridine secondary products.

Furthermore, the known process has the serious disadvantage expressly mentioned in US-3,449,324 that the activity of the catalyst gradually declines, so that according to US-3,449,324, Example 2, it would have to be regenerated after only about 30 minutes' use. This can be attributed to the fact that a not inconsiderable proportion of the iminodibenzyl used becomes resinated and is precipitated in the form of tar- or carbon-like deposits which have to be removed periodically by burning out the catalyst. The yields

AUSTRALIAN PATENT OFFICE

given in US-3,449,324 therefore cannot be obtained in practice, as shown especially by Example 21 in comparison with Comparison Examples 1 and 2. The resination of considerable proportions of the iminodibenzyl used not only reduces the yield of iminostilbene but also involves ecological and industrial safety risks resulting from the oxidative secondary reactions which are difficult to control. In particular, it is necessary to interrupt the process periodically, which renders continuous operation impossible.

The problem underlying the invention is, therefore, to provide an improved process for the preparation of iminostilbene by catalytic high-temperature dehydrogenation of iminodibenzyl in the vapor phase that avoids the above-described disadvantages and other shortcomings of the known process. The solution to this problem proposed according to the invention is based on the surprising finding that the periodic regeneration of the catalyst is unnecessary when the reaction is carried out at a temperature of 500° to 600° C and when there is used an iron oxide/potassium salt contact catalyst comprising from 44 to 85 % weight of an iron compound, calculated as $Fe_2O_3$, and from 7 to 35 % by weight of a potassium compound, calculated as $K_2O$, in addition to from 0.0 to 3.5 % by weight of a chromium compound, calculated as $Cr_2O_3$, and from 0.0 to 3.0 % of a calcium compound, calculated as CaO, and optionally customary promotors.

It is also surprising that the catalyst used according to the invention in the high-temperature dehydrogenation of a heterocyclic compound of such a high molecular weight as iminodibenzyl works autoregeneratively and exhibits excellent activity and selectivity.

It is especially surprising that, as will be seen from Examples 12 to 15, contrary to the prejudice arising from US-3,449,324according to the invention it is precisely at a reaction temperature of 550°C and above that the best conversion and the highest yield of iminostilbene are achieved.

As will be seen from Example 2, the contact catalyst used according to the invention advantageously achieves a service life of more than 1100 hours without significant amounts of iminodibenzyl being lost as a result of undesired oxidative secondary reactions. As a consequence, the invention opens up the possibility of continuous operation.

The process according to the invention for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapor phase is accordingly carried out by using an iron oxide/potassium salt

AUSTRALIAN PATENT OFFICE

contact catalyst comprising from 44 to 85 % by weight, of an iron compound, calculated as $Fe_2O_3$, and from 7 to 35 % by weight, for example from 9 to 31 % by weight, of a potassium compound, calculated as $K_2O$, in addition to from 0.0 to 3.5 %, more particularly 0.02 to 2.5%, by weight of a chromium compound, calculated as $Cr_2O_3$, and and 0.0 to 3.0 % of a calcium compound, calculated as CaO, and optionally customary promotors.

Suitable promotors are, for example, magnesium, calcium, cerium, molybdenum, cobalt, vanadium and tungsten compounds, for example in the form of oxides of the said metals, especially tungsten, cerium, vanadium and cobalt oxides.

Although promotors of the mentioned or similar type can increase the performance of the catalyst, they are not absolutely necessary, as will be seen from Examples 17 to 20. In general the quantitative ratios of the components are not critical provided that the proportions of iron and potassium remain within the scope of the definition given above. Particularly, the process may be carried out by using an iron/potassium salt contact catalyst comprising from 44 to 85 % by weight of an iron compound, calculated as $Fe_2O_3$, and form 7 to 35 % by weight, for example from 9 to 31 % by weight of a potassium compound, calculated as $K_2O_3$, in addition to from 0.02 to 2.0 % by weight of a tungsten compound, calculated as $WO_3$; from 1.0 to 3.0 % by weight of a calcium compound, calculated as CaO; from 0.5 to 5.0 % by weight of a cerium compound, calculated as $Ce_2O_3$; from 0.02 to 2.5 % by weight of a vanadium compound, calculated as $V_2O_5$; from 2.0 to 3.0 % by weight of a magnesium compound, calculated as MgO; from 0.05 to 2.5 % by weight of a molybdenum compound, calculated as $MoO_3$; and/or from 0.02 to 1.0 % by weight of a cobalt compound, calculated as $Co_2O_3$.

The contact catalyst used according to the invention is preferably, but not necessarily, brought into contact with iminodibenzyl vapors in an adiabatic fixed bed reactor. It is also possible, however, to use tube bundle, fluid bed and fluidised bed reactors, as well as other types of reactor in accordance with the prior art. The morphology of the catalyst used according to the invention should offer as little resistance as possible to the vapors flowing through the catalyst bed. It is preferable to use geometric, for example cylindrical, moulded bodies about 1 to 6 mm, especially 1 to 4 mm, in length and about 0.5 to 5 mm, especially 1 to 3 mm, in diameter, and also granules having an average particle size of about from 0.5 to 5 mm, especially about from 1 to 3 mm.

It is advantageous to dilute the iminodibenzyl vapors with an inert carrier gas, such as

with nitrogen or especially an approximately 50- to 200-fold, for example approximately 120- to 140-fold, molar amount of water vapor. It is advantageous to operate at normal pressure or at slightly reduced pressure, for example at about from 0.2 to 1.1 bar, especially about from 0.95 to 1.05 bar, (absolute) and to allow the reaction gases to cool to room temperature, the resulting iminostilbene and unreacted iminodibenzyl being obtained in a solid form suspended in the condensed transport water. The iminostilbene component can then be separated from the condensate and the unreacted iminodibenzyl can be isolated and recycled. The procedure to be used is known and can be found, for example, in US-3,449,324.

The following Examples serve to illustrate the invention; temperatures are given in degrees Celsius and pressures in bar absolute.

Example 1: The dehydrogenation of iminodibenzyl (IDB) to iminostilbene (IS) in the gaseous phase is carried out in a quartz glass tube reactor 31 mm in diameter and 1 m in length. 30 g of a catalyst having the following properties

| | | | |
|---|---|---|---|
| Composition: | 1.1 | % by weight | chromium oxide, calculated as $Cr_2O_3$ |
| | 22 | % by weight | potassium compound, calculated as $K_2O$ |
| | remainder | | iron oxide |

particle size: 1-2 mm

are introduced between two filler layers (corundum; particle size: 1-2 mm) in the centre of the reaction tube. The upper filler layer acts as a vaporisation zone for the iminodibenzyl. 27.7 g/h of water are vaporised in a separate oven and passed into the reactor from above. 6 g/h of iminodibenzyl at 120°C in molten form are metered directly into the vaporisation zone, mixed with the pre-heated vapor and vaporised. By electrically heating the reactor, the vapor mixture is heated to the reaction temperature of 550°C. After the reaction in the approximately 35 mm high catalyst layer, the product vapors are cooled to room temperature in a vessel, so that the mixture of iminodibenzyl and iminostilbene is obtained in the form of powder in condensed water. The condensates taken at different times during the experiment are homogenised with dioxane and analysed by capillary gas chromatography (without water). During the 860-hour duration of the experiment, the following product composition (without water) is obtained:

| time [h] | IDB % by wt. | IS % by wt. | sec. comp. % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|
| 2 | 25.0 | 51.4 | 20.9 | 75 | 72 |
| 25 | 28.9 | 49.6 | 21.5 | 71 | 70 |
| 92 | 27.4 | 48.4 | 24.2 | 73 | 67 |
| 554 | 30.9 | 47.5 | 21.6 | 68 | 69 |
| 630 | 26.7 | 49.9 | 21.6 | 73 | 69 |
| 860 | 29.9 | 47.2 | 22.9 | 70 | 67 |

In addition to iminodibenzyl (IDB) and iminostilbene (IS), mainly 9-methylacridine and acridine are detected as secondary compounds. The experiment is carried out continuously for about 100 hours each time with interruptions at weekends. After an interruption the catalyst is scavenged for one hour with water vapor at the reaction temperature and then cooled to room temperature under nitrogen. When the experiment is started up again, that procedure is followed in reverse order.

Example 1 demonstrates the long service life and the continuous operation of the catalyst.

Example 2: 80 g of a catalyst having the following composition is introduced into a quartz glass tube reactor according to Example 1:

Composition:
12.3 % by weight of potassium oxide, calculated as $K_2O$
1.9 % by weight of chromium oxide, calculated as $Cr_2O_3$
1.6 % by weight of tungsten oxide, calculated as $WO_3$
2.4 % by weight of cerium oxide, calculated as $Ce_2O_3$
1.5 % by weight of vanadium oxide, calculated as $V_2O_5$
0.3 % by weight of cobalt oxide, calculated as $Co_2O_3$
remainder iron oxide

particle size: 1-2 mm

A stream of 40 g/h of iminodibenzyl and 480 g/h of water vapor at 590° is passed over the

catalyst for 1145 hours with the interruptions at weekends described in Example 1. The product, which is separated from water by filtration, has the following composition (as a function of the reaction time):

| time [h] | IDB % by wt. | IS % by wt. | byproducts % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|
| 39 | 13.5 | 76.2 | 10.3 | 87 | 88 |
| 148 | 13.5 | 74.7 | 11.8 | 87 | 87 |
| 250 | 12.6 | 76.9 | 10.5 | 86 | 88 |
| 367 | 13.1 | 76.8 | 10.1 | 87 | 88 |
| 460 | 13.7 | 76.1 | 10.2 | 87 | 88 |
| 550 | 14.6 | 75.3 | 10.1 | 86 | 88 |
| 685 | 12.7 | 76.2 | 11.1 | 87 | 87 |
| 914 | 11.6 | 77.4 | 11.0 | 89 | 88 |
| 1028 | 13.0 | 75.8 | 11.2 | 87 | 87 |
| 1145 | 14.6 | 74.4 | 11.0 | 86 | 87 |

Example 2 demonstrates the good selectivity, the long service life of more than 1100 hours and the continuous operation of a catalyst comprising tungsten oxide, cerium oxide, vanadium oxide and cobalt oxide as promotors.

Examples 3 to 6: Using the same experimental procedure and the same conditions as in Example 1, the amount of water in relation to the metered amount of IDB is varied within a wide range:

AUSTRALIAN PATENT OFFICE

| Example | water [g/h] | IDB % by wt. | IS % by wt. | sec. comp. % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|---|
| 3 | 11.7 | 38.6 | 32.0 | 29.4 | 61 | 52 |
| 4 | 27.7 | 26.7 | 49.9 | 23.4 | 73 | 68 |
| 5 | 55.4 | 20.6 | 60.2 | 19.2 | 79 | 76 |
| 6 | 110.8 | 1.7 | 60.1 | 38.2 | 98 | 61 |

Examples 3 to 6 demonstrate the large amount of influence exerted by the ratio of water to IDB on the conversion and on the selectivity of the reaction.

Examples 7 to 11: Using the same experimental procedure and 30 g of the same catalyst as in Example 1, the dehydrogenation is carried out at 570°C. The metered amounts of IDB and water are altered in such a manner that the ratio of water to IDB remains constant.

| Example | water [g/h] | IDB [g/h] | IDB % by wt. | IS % by wt. | sec. comp. % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|---|---|
| 7 | 276.9 | 30 | 34.0 | 55.4 | 10.6 | 66 | 84 |
| 8 | 138.5 | 15 | 16.4 | 67.3 | 16.3 | 84 | 81 |
| 9 | 92.3 | 10 | 9.6 | 70.9 | 19.5 | 90 | 78 |
| 10 | 55.4 | 6 | 8.1 | 64.7 | 27.2 | 92 | 70 |
| 11 | 27.7 | 3 | 5.5 | 57.5 | 37.0 | 95 | 61 |

Examples 7 to 11 demonstrate the influence exerted by the contact time (contact time factor = 1-10 h per hour) on the conversion or the selectivity of the reaction,

$$\left(\text{contact time factor} = \frac{g_{(\text{catalyst})} \times h}{g_{(\text{iminodibenzyl})}}\right)$$

with a constant ratio of water to iminodibenzyl (IDB).

Examples 12 to 15: Using the same experimental procedure and 30 g of the same catalyst

AUSTRALIAN PATENT OFFICE

and the same metered amounts of water and IDB as in Example 1, the dehydrogenation reaction is carried out at various temperatures. The Table below shows the product composition.

| Example | temperature [°C] | IDB % by wt. | IS % by wt. | sec. comp. % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|---|
| 12 | 510 | 55.1 | 32.7 | 12.2 | 45 | 73 |
| 13 | 530 | 39.5 | 43.4 | 17.1 | 61 | 72 |
| 14 | 550 | 26.7 | 49.9 | 23.4 | 73 | 68 |
| 15 | 570 | 25.5 | 45.0 | 29.5 | 75 | 60 |

Examples 12 to 15 demonstrate the strong influence exerted by the temperature on the rate of reaction and the selectivity of the reaction.

Example 16: 30 g of a catalyst having the following properties are introduced into the reactor of Example 1:

Composition:
9.0 % by weight of a potassium compound, calculated as $K_2O$
0.04 % by weight of chromium oxide, calculated as $Cr_2O_3$
2.7 % by weight of a calcium compound, calculated as CaO
4.8 % by weight of cerium oxide, calculated as $Ce_2O_3$
2.5 % by weight of magnesium oxide, calculated as MgO
2.0 % by weight of molybdenum oxide, calculated as $MoO_3$
remainder iron oxide

particle size: 1.5 mm

At 0.4 bar (absolute) and 570°C, a vaporous stream of 6.0 g/h of IDB and 55.4 g/h of water is passed over the catalyst. The product comprises 2.9 % by weight IDB, 74.0 % by weight IS and 23.1 % by weight secondary compounds (mainly acridine and 9-methyl-acridine) without taking account of the water. The same experiment at normal pressure results in a product composition of 6.2 % by weight IDB, 66.9 % by weight IS and 26.9 % by weight secondary compounds.

AUSTRALIAN PATENT OFFICE

Example 16 demonstrates the influence exerted by reduced pressure (vacuum) on the conversion and the selectivity of the reaction.

Examples 17 to 20: In the following Examples the influence of the composition of the catalyst on the conversion and on the selectivity of the reaction under the same test conditions is demonstrated. In the test apparatus of Example 1. 30 g of each of the catalysts given below are tested (particle size 1-2 mm). At 550°C, 15 g/h of IDB and 180 g/h of water in vapor form are passed over the catalysts.

| Example | IDB % by wt. | IS % by wt. | sec. comp. % by wt. | conversion % | selectivity % |
|---|---|---|---|---|---|
| 17 | 28.2 | 59.0 | 12.8 | 72 | 82 |
| 18 | 18.0 | 67.7 | 14.3 | 82 | 83 |
| 19 | 20.9 | 63.2 | 15.9 | 79 | 80 |
| 20 | 27.2 | 60.0 | 12.8 | 73 | 82 |

The composition of the catalysts used in Examples 17 to 20 is given in the following Table:

| Example | $Fe_2O_3$ [% by wt.] | $K_2O$ [% by wt.] | $Cr_2O_3$ [% by wt.] | CaO [% by wt.] | $Ce_2O_3$ [% by wt.] | MgO [% by wt.] | $MoO_3$ [% by wt.] |
|---|---|---|---|---|---|---|---|
| 17 | 61 | 22 | 1.1 | --- | --- | --- | --- |
| 18 | 85 | 9 | 1.9 | --- | --- | --- | --- |
| 19 | 77 | 9 | 0.04 | 2.7 | 4.8 | 2.5 | 2.0 |
| 20 | 44 | 31 | 2.5 | --- | 0.5 | --- | 0.1 |

Examples 17 to 20 demonstrate that the reaction can be carried out using various catalysts of very different composition.

Example 21: Using the experimental procedure of Example 1, 30 g of a catalyst having the following properties are introduced:

Composition: 1.3 % by weight of chromium oxide, calculated as $Cr_2O_3$
13.1 % by weight of a potassium compound, calculated as $K_2O$
remainder iron oxide

particle size: 1-2 mm

At 550°C and 1 bar absolute, a vaporous stream of 6 g/h of iminodibenzyl and 72 g/h of water is passed over the catalyst. By filtering and drying the condensed products it is possible to recover a mass of 99 %, based on the amount of substance used (iminodibenzyl). The loss in mass of 1 % is attributable to the removal of hydrogen and methane from the reactants. The isolated product has the following composition: 76.2 % by weight iminostilbene, 3.8 % by weight iminodibenzyl and 20 % by weight secondary compounds. The total yield of iminostilbene, taking account of the losses in mass, is therefore 75.4 %.

That Example demonstrates the high yields that can be attained and the low losses in mass when the reaction is carried out continuously.

Comparison Example 1: (Comparison with the cyclic process)

Using the experimental procedure of Example 1, 50 g of the catalyst described in Example 1 of US-3,449,324 are introduced. In accordance with the data given in the above-mentioned Patent, the reaction is carried out at 430°C, 1 bar absolute and with cyclic operation. The cycle times are as follows: 5 minutes pre-scavenging with water vapor, 18 minutes reaction, 5 minutes post-scavenging with water vapor and 30 minutes regeneration (oxidation of the catalyst with air). The metered amounts are: prescavenging with water vapor: 38.3 g/h, reaction: 8.3 g/h of IDB and 38.3 g/h of water vapor, post-scavenging: 38.3 g/h of water vapor, and regeneration: 100 ml/minute of air and 38.3 g/h of water vapor. The product collected during 20 cycles is filtered off and dried. The amount of substance recovered was 88 % by weight of the amount of IDB used. Since a portion of the product remains on the catalyst after the reaction phase, that adsorbed portion is oxidised with air during the regeneration phase to form unusable products. The product composition is on average: 65 % by weight iminostilbene, 28 % by weight iminodibenzyl and 7 % by weight secondary compounds. The total yield, taking account of the losses in mass, is accordingly 57 %.

That Comparison Example demonstrates the significantly poorer yield obtained using the

AUSTRALIAN PATENT OFFICE

cyclic process in comparison with the continuous process.

Comparison Example 2: (Comparison with the cyclic process)

Using the same experimental procedure, the same cycle times, the same catalyst and the same conditions as in Comparison Example 1, 5 g/h of IDB and 23.3 g/h of water vapor are passed over the catalyst. After filtering and drying the reaction mass it is possible to isolate a further 78 % of the amount of substance used. The product composition is on average: 72 % by weight iminostilbene, 16 % by weight iminodibenzyl and 12 % by weight secondary compounds. The total yield, taking account of the losses in mass, is accordingly 56 %.

That Comparison Example shows that although it is possible to increase the conversion by reducing the ratio of the metered amount of IDB to the amount of catalyst, the total yield is not increased, owing to the greater losses in mass occurring on regeneration of the catalyst.

AUSTRALIAN PATENT OFFICE

The claims defining the invention are as follows:

1. A process for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapour phase, wherein there is used an iron oxide/potassium salt contact catalyst comprising from 44 to 85 % by weight of an iron compound, calculated as $Fe_2O_3$, and from 7 to 35 % by weight of a potassium compound, calculated as $K_2O$, in addition to from 0.0 to 3.5 % by weight of a chromium compound, calculated as $Cr_2O_3$, and from 0.0 to 3.0 % of a calcium compound, calculated as CaO, and optionally customary promotors, and wherein the reaction is carried out at a temperature of 500° to 600° C.

2. A process according to claim 1, wherein there is used as promotor a magnesium, cerium, molybdenum, cobalt, vanadium or tungsten compound.

3. A process according to claim 1, wherein there is used as promotor a tungsten oxide, cerium oxide, vanadium oxide and cobalt oxide.

4. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 9 to 31 % by weight of a potassium compound, calculated as $K_2O$, is used.

5. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.02 to 2.5 % by weight of a chromium compound, calculated as $Cr_2O_3$, is used.

6. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.02 to 2.0 % by weight of a tungsten compound, calculated as $WO_3$, is used.

7. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 1.0 to 3.0 % by weight of a calcium compound, calculated as CaO, is used.

8. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.5 to 5.0 % by weight of a cerium compound, calculated as $Ce_2O_3$, is used.

AUSTRALIAN PATENT OFFICE

9. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.02 to 2.5 % by weight of a vanadium compound, calculated as $V_2O_5$, is used.

10. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 2.0 to 3.0 % by weight of a magnesium compound, calculated as MgO, is used.

11. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.05 to 2.5 % by weight of a molybdenum compound, calculated as $MoO_3$, is used.

12. A process according to claim 1, wherein an iron oxide/potassium salt contact catalyst comprising from 0.02 to 1.0 % by weight of a cobalt compound, calculated as $Co_2O_3$, is used.

13. A process according to claim 1, wherein the contact catalyst is brought into contact with iminodibenzyl vapours in an adiabatic fixed bed reactor or in a tube bundle reactor, fluid bed reactor or fluidised bed reactor.

14. A process according to claim 1, wherein the contact catalyst is used in the form of geometric moulded bodies approximately 1 to 6 mm in length and approximately 0.5 to 5 mm in diameter or in the form of granules having an average particle size of about from 0.5 to 5 mm.

15. A process according to claim 1, wherein the iminodibenzyl vapours are diluted with an inert carrier gas.

16. A process according to claim 1, wherein the iminodibenzyl vapours are diluted with a 50- to 200-fold molar amount of water vapour.

17. A process according to claim 1, wherein the operation is carried out at from 0.2 to 1.1 bar bar (absolute).

AUSTRALIAN PATENT OFFICE

18. A process according to claim 1 substantially as herein described with reference to any one of the foregoing examples thereof.

DATED this 27th day of June, 1995.

**CIBA-GEIGY AG**
By Its Patent Attorneys,
DAVIES COLLISON CAVE

4-19088/A

# Novel high-temperature dehydrogenation process

The invention relates to a novel process for the preparation of iminostilbene by high-temperature dehydrogenation of iminodibenzyl on an iron oxide/potassium salt contact catalyst in the vapour phase, wherein there is used an iron oxide/potassium salt contact catalyst comprising from 35 to 90 % by weight of an iron compound, calculated as $Fe_2O_3$, and from 7 to 35 % by weight of a potassium compound, calculated as $K_2O$, in addition to from 0.0 to 3.5 % by weight of a chromium compound, calculated as $Cr_2O_3$, and optionally customary promotors.