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Catalyst, process for producing the catalyst and
process for preparing vinyl acetate using the catalyst

5 The present invention relates to a catalyst comprising
palladium and/or its compounds, gold and/or its
compounds and also at least one alkali metal compound,
a process for producing it and its use for preparing
vinyl acetate in the gas phase from acetic acid,
10 ethylene and oxygen or oxygen-containing gases.

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It is known from the prior art that vinyl acetate can
be prepared in the gas phase from ethylene, oxygen and
acetic acid in the presence of catalysts which comprise
palladium, gold and alkali metal compounds on a porous
15 support material (such as silicon dioxide).

The distribution of the noble metals on the support
material is of particular importance for the activity
and selectivity of these catalysts. Since the reactants
20 in the reaction to be catalyzed cannot readily diffuse
into the intermediate or inner regions of the porous
support material, the reaction essentially takes place
only on the outermost or surface regions of the
catalyst. Thus, the metal components present in the
25 interior or the intermediate regions of the catalyst do
not contribute significantly to the reaction mechanism,
which leads to a reduction in the productivity of the
catalyst based on the weight of the noble metals.

30 In the development of more effective catalysts for
vinyl acetate production, efforts are therefore
directed at producing catalysts in which the catalyti-
cally active metals are present in a shell on the
support particles while the core of the support
35 particles is largely free of noble metals. Such shell
catalysts can be produced in principle by impregnating
the support material with soluble noble metal
compounds, subsequently precipitating insoluble noble
metal compounds on the support by means of alkaline

compounds and finally reducing them to the noble metals.

5 US-A-4,048,096 describes a process for producing a palladium-, gold- and potassium-containing catalyst for vinyl acetate production. The catalyst support is first impregnated with a solution comprising a mixture of the dissolved palladium and gold salts. It is essential to that invention that this solution has the same volume
10 as the pores of the support material in the dry state. During this impregnation step, the support particles are kept in motion in a rotating vessel. Without prior drying of the impregnated support, the noble metal salts on the support particles are subsequently
15 converted into insoluble compounds by addition of alkalis and thus fixed to the support particles. Final treatment with a reducing agent converts the palladium and gold compounds to the corresponding metals. Application of an alkali metal compound in a further
20 impregnation step gives a catalyst which has the desired shell structure comprising palladium and gold in a thickness of 0.5 mm on the surface of the support material.

25 US-A-3,775,342 also describes the production of a palladium-, gold- and potassium-containing catalyst for vinyl acetate production. In this process, the support material is treated in any order with two solutions of which one comprises the dissolved palladium and gold
30 salts and the other comprises an alkaline substance. After treatment with the first solution, the support is dried in an intermediate step before being brought into contact with the second solution. The volume of both solutions in each case corresponds to the pore volume
35 of the support material.

Furthermore, US-A-5,332,710 discloses the production of a catalyst for producing vinyl acetate in which the

insoluble noble metal salts are likewise precipitated on the support particles by addition of alkali. For this purpose, the support particles are immersed in the alkaline solution and, from the commencement of precipitation, are kept in rotary motion in a drum for at least half an hour. This process is known as "rotation-immersion".

In the preparation of vinyl acetate, the catalysts produced by the abovementioned processes frequently lead to undesirably high formation of degradation products and by-products, e.g. carbon dioxide, which has an adverse effect on the activity and selectivity of the overall reaction.

In view of the fact that vinyl acetate is produced in large volume on an industrial scale, it is an object of the present invention to provide a catalyst which displays further improved selectivity in the preparation of vinyl acetate in the gas phase.

The invention provides a process for producing a catalyst for the preparation of vinyl acetate in the gas phase from ethylene, acetic acid and oxygen or oxygen-containing gases, which catalyst comprises palladium and/or its compounds, gold and/or its compounds and also alkali metal compounds on a particulate, porous support and is produced by

- a) impregnating the support with soluble palladium and gold compounds,
- b) converting the soluble palladium and gold compounds into insoluble palladium and gold compounds by addition of an alkaline solution to the support,
- c) reducing the insoluble palladium and gold compounds on the support by means of a

reducing agent in the liquid or gaseous phase,

- d) impregnating the support with at least one soluble alkali metal compound and
- 5 e) finally drying the support at a maximum of 150°C.

In this process, the catalyst is irradiated with micro-waves before, during or after one of the process steps
10 a) to e).

The invention also provides a catalyst for preparing vinyl acetate in the gas phase from ethylene, acetic acid and oxygen or oxygen-containing gases which
15 comprises palladium and/or its compounds, gold and/or its compounds and also alkali metal compounds on a particulate, porous support and is obtainable by the above-described process.

20 The invention further provides a process for preparing vinyl acetate in the gas phase from ethylene, acetic acid and oxygen and/or oxygen-containing gases in the presence of a catalyst which is obtainable by the above-described process.

25 In the preparation of vinyl acetate, the catalysts of the invention surprisingly lead not only to improved selectivity of the reaction, but also make possible a higher space-time yield.

30 The support particles of the catalyst of the invention can have any geometric shape, for example spheres, pellets, cylinders, rings or stars with a regular or irregular configuration. The dimensions of the support
35 particles, i.e. the diameter or the length and thickness are generally from 1 to 10 mm, in particular from 3 to 9 mm. Preference is given to using spherical support particles having a diameter of from 4 to 8 mm.

Supports which can be used are the known inert support materials such as silica, aluminum oxide, aluminosilicates, silicates, titanium oxide, zirconium oxide, titanates, silicon carbide and carbon. Other suitable support materials are the pyrogenic silicas obtained by flame hydrolysis of silicon tetrachloride or the pyrogenic $\text{SiO}_2\text{-M}_x\text{O}_y$ mixtures obtained by flame hydrolysis of silicon tetrachloride and another metal chloride such as aluminum chloride (US-A-3,939,199 and EP-A-0 723 810). Preference is given to using silica (SiO_2), baddeleyite (ZrO_2) and $\text{SiO}_2\text{-Al}_2\text{O}_3$ mixtures as support material. In the case of the pyrogenic support materials, the pressed bodies described in DE-A-38 03 895 and DE-A-39 12 504 are particularly suitable.

For a support material to be suitable, it is particularly important that it retains its mechanical strength under the reaction conditions of the catalytic process for preparing vinyl acetate, in particular in the presence of acetic acid.

Particularly suitable supports of the abovementioned type are those having a specific surface area of from 50 to $400 \text{ m}^2/\text{g}$ (measured by the BET method) and a mean pore radius of from 50 to 2000 Å (measured by means of mercury porosimetry).

In step a) of the process of the invention, the impregnation step, the support particles are impregnated with the soluble palladium and gold compounds. Suitable palladium and gold compounds are all salts and complexes which are soluble in the solvents described below, can also be precipitated as hydroxide or oxide and in the finished catalyst, possibly after a washing step, leave no substances which impair the performance of the catalyst.

Examples of suitable palladium compounds are palladium(II) chloride, sodium or potassium chloropalladate(II), palladium(II) nitrate, nitrite, sulfate, oxalate, acetylacetonate or acetoacetate or hydrated
5 palladium(II) oxide. It is also possible to use palladium salts of aliphatic monocarboxylic acids having from 2 to 5 carbon atoms, preferably palladium(II) acetate. Gold(III) chloride, gold(III) acetate, tetrachloroauric(III) acid and its alkali
10 metal salts can be used as soluble gold compounds. These compounds are generally used in such amounts that the finished catalyst comprises from 2 to 14 g/l, preferably from 4 to 8 g/l, of palladium and from 1 to 8 g/l, preferably from 2 to 5 g/l, of gold.

15 Suitable solvents for the palladium and gold compounds and also for the alkali metal compounds to be applied in step d) are all compounds in which the chosen salts are soluble and which can easily be removed again in an
20 optional drying step after impregnation. Particularly suitable solvents are water or unsubstituted carboxylic acids having from 2 to 10 carbon atoms, e.g. acetic acid, propionic acid, n- and iso-butyric acid and n- and iso-valeric acids. Owing to its favorable physical
25 properties and also for economic reasons, the preferred carboxylic acid is acetic acid. It is advantageous to use an additional solvent when the carboxylic acid used is one in which the palladium and gold compounds are not sufficiently soluble. Thus, for example,
30 palladium(II) chloride dissolves significantly better in aqueous acetic acid than in glacial acetic acid. Suitable additional solvents are those which are inert and at the same time miscible with the carboxylic acid, e.g. water, ethers such as tetrahydrofuran or dioxane
35 and hydrocarbons such as benzene.

In the impregnation of the support material, each of the metals to be applied can be in the form of a

plurality of salts of the respective metal, but preference is given to using only one salt per metal.

5 The impregnation of the support material with the soluble palladium and gold compounds in step a) can be carried out using a solution which simultaneously contains all soluble palladium and gold compounds. The support material can be impregnated once or a plurality of times with this solution. Since the amount of
10 palladium and gold compounds applied should be identical for single and multiple impregnation, the total volume of the solution should be appropriately divided in the case of multiple impregnation. Preference is given to a single impregnation with the
15 total volume of the solution.

In an alternative embodiment, the impregnation of the support material can also be carried out using two separate solutions of which one contains the palladium
20 compounds and the other contains the gold compounds. In this case, the two solutions can be brought into contact with the support material either simultaneously or in any order. In the latter case, the support has to be dried after impregnation with the first solution.

25 For effective impregnation, the total volume of the noble metal salt solution or the two noble metal salt solutions should be about 90-100%, preferably 95-100% and in particular 98-99%, of the pore volume of the
30 support material in the dry state. In practice, it is also possible to cover the support particles with an excess of the noble metal salt solution and then to pour away or filter off the excess solution. However, preference is given to adding only the above-indicated
35 amount of solution corresponding approximately to the pore volume of the catalyst support.

It has been found to be advantageous to keep the support particles in motion during impregnation in order to achieve intimate mixing. This can be done by means of a rotating or shaken flask or a mixing drum.

5 The rotational speed or, in general terms, the intensity of the motion should be sufficient to achieve complete wetting of the support particles with the impregnation solution but must not be so great that appreciable abrasion of the support material occurs.

10

If desired, the impregnated support material is dried at temperatures of at most 150°C, preferably 80-150°C and in particular 100-150°C. This drying can be carried out, for example, in a stream of hot air in a fan-
15 forced dryer or else in a drying oven in a stream of inert gas, in particular a stream of nitrogen or carbon dioxide. Drying may be carried out at atmospheric pressure or under reduced pressure, preferably 0.01-0.08 MPa.

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In step b), the fixing step, the soluble palladium and gold compounds located on the support particles are converted into insoluble compounds by addition of an alkaline solution and thus fixed to the support. It is
25 assumed that the insoluble compounds are the hydroxides and/or oxides of the noble metals.

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Suitable alkaline solutions are all those which are able to convert the soluble palladium and gold
30 compounds into insoluble compounds. Alkaline reagents which can be used are alkali metal hydroxides, alkali metal silicates and alkali metal carbonates. Preference is given to an aqueous solution of the alkali metal hydroxides, in particular potassium or sodium
35 hydroxide. It is also possible to use aqueous solutions containing boron compounds as alkaline solutions. These are, in particular, aqueous solutions of sodium tetraborate decahydrate (borax), potassium tetraborate or

mixtures of alkali metal hydroxides and boric acid. The alkaline solution can have buffer properties.

5 The amount of alkaline compound present in the aqueous solution is advantageously selected such that it is at least sufficient for the stoichiometric reaction with the soluble palladium and gold compounds applied. However, it is also possible to use an excess of the alkaline compound, usually 1-10 times the stoichio-
10 metrically required amount.

Two methods I and II which are suitable for carrying out the fixing step b) and can be employed for producing the catalyst of the invention are described
15 below.

In method I, the support material which has been impregnated in step a) is placed for a sufficient time in an alkaline solution whose concentration is such
20 that the desired, insoluble noble metal compounds are precipitated. The volume of the alkaline solution is selected such that it is sufficient to completely cover and immerse the impregnated support particles. Furthermore, the impregnated support particles which are
25 immersed in the alkaline solution are placed in rotary motion commencing with the precipitation of the insoluble palladium and gold compounds; the rotation should be carried out for at least half an hour, preferably one hour and at most up to 4 hours. This
30 fixing method is known as "rotation-immersion" and is described in detail in US-A-5,332,710, which is hereby incorporated by reference.

If method II described below is employed for fixing the
35 palladium and gold compounds to the support particles, the support impregnated in step a) should be dried before the fixing step b).

In method II, the fixing step b) comprises at least two separate stages of treatment with the alkaline fixing solution. In the first fixing stage, the impregnated and then dried support is brought into contact with the alkaline fixing solution. The volume of this first fixing solution corresponds to the pore volume and thus the absorptive capacity of the support material in the dry state. The amount of alkaline compound present therein should be such that the molar ratio of alkali metal from the alkaline compound to anions from the soluble metal salt is in the range from 0.7:1 to 2:1. For absorption by the support particles, the alkaline fixing solution is poured onto the support particles and they are then allowed to stand for up to 24 hours, preferably 2-8 hours.

In this method II, the second fixing stage can be carried out in two variants A) and B).

In both variants, the molar ratio of alkali metal from the alkaline compound to the anion from the metal salt in the fixing solution is from about 0.2:1 to 2:1.

In variant A) of method II, the undried support particles are brought into contact with a second fixing solution which should have a volume at least sufficient to just cover the supports. For absorption by the support particles, the alkaline fixing solution is poured onto the support particles and they are then allowed to stand for up to 16 hours, but at least 2 hours and preferably at least 4 hours.

In variant B), the supports after contact with the first fixing solution are treated in a second stage by the rotation-immersion process of US-A-5,332,710. Here, the supports are immersed in the alkaline fixing solution of the second stage and at the same time subjected to rotary motion. The rotation should take

place for at least half an hour, preferably one hour and at most up to 4 hours.

- Regardless of whether variant A) or B) is employed, the treatment in the second fixing stage can be equivalent to the treatment in the first stage in that a fixing solution of the same concentration is used and the volume of the second fixing solution likewise corresponds to the pore volume and thus the absorptive capacity of the support material in the dry state. The total molar ratio of alkali metal to anion from the metal salt for both fixing stages together is preferably in the range from 1.1:1 to 3.3:1.
- Subsequent to the fixing step of method I or the last fixing step of method II, the supports can be washed with water, preferably with distilled water, to remove any anions, e.g. chlorides, which originate from the impregnation step, have been set free by the precipitation of the noble metals and are still present on the support material. Furthermore, this washing also removes any excess of the alkaline compound which may still be present.
- If desired, the impregnated material is dried at temperatures of at most 150°C, preferably 80-150°C and in particular 100-150°C, after the fixing step. This drying can be carried out, for example, in a stream of hot air in a fan-forced dryer or else in a drying oven in a stream of inert gas, in particular in a stream of nitrogen or carbon dioxide. Drying may be carried out at atmospheric pressure or under reduced pressure, preferably 0.01-0.08 MPa. Such drying is particularly advantageous at this point when the reduction step c) described below is carried out in the gas phase. On the other hand, if the reduction is carried out in the liquid phase, prior drying is not necessary.

In step c), the support and the insoluble palladium and gold compounds deposited thereon are treated with a reducing agent in order to convert the precipitated palladium and gold compounds into the metallic form.

5 This reduction can be carried in the liquid phase at a temperature of 0-90°C, preferably 15-25°C. Examples of reducing agents used are hydrazine, formic acid or an alkali metal borohydride, preferably sodium borohydride. As an alternative, a reduction in the gas
10 phase using hydrogen, ethylene, propylene, isobutylene, butylene or other olefins as reducing agent is also possible. In this case, it is advantageous to carry out the reaction at an increased temperature of 40-260°C, preferably 70-200°C. It is also advantageous to dilute
15 the reducing agent with an inert gas. The inert gas used can be, for example, nitrogen, carbon dioxide or a noble gas. Usually, such a reducing agent/inert gas mixture contains 0.01-50% by volume, preferably 0.5-20% by volume, of reducing agent. Regardless of whether the
20 reduction is carried out in the liquid or gas phase, the reducing agent should be added in an excess based on the catalyst to be reduced so as to ensure that all the insoluble noble metal compounds are converted into the metallic form.

25 Subsequent to the reduction, the support particles can again be washed once or a plurality of times, preferably with distilled water, to remove interfering anions, e.g. chlorides, and residues of the alkaline
30 compound used. The washing procedure can also serve to remove residues of the reducing agent from step c).

The catalyst can subsequently be dried again. Here, the drying conditions selected should be similar to those
35 described for drying after the fixing step b)

Finally, the addition of at least one alkali metal compound is necessary. The catalyst is therefore, in

step d), preferably impregnated with an aqueous solution of an alkali metal compound. Alkali metal compounds which can be used are sodium, potassium, rubidium or cesium compounds; preference is given to potassium compounds. Suitable anions in these alkali metal compounds are, in particular, carboxylates, especially acetates or propionates. Particular preference is given to using potassium acetate. However, it is also possible to use compounds which form alkali metal acetates under the reaction conditions, i.e. when acetic acid is used as solvent these are the alkali metal hydroxides, oxides or carbonates. In principle, this impregnation is carried out in the same way as the impregnation of the support material in step a). The solvents which can be used are subject to the same conditions and definitions as in the case of the solutions in impregnation step a). The alkali metal compound is used in such an amount that the catalyst after the drying step described below contains 0.1-10% by weight of alkali metal, preferably 1-4% by weight of alkali metal, in particular potassium, based on the total mass of the catalyst.

The catalyst is finally dried in step e) at temperatures of at most 150°C, preferably 80-150°C and in particular 100-150°C. This drying can be carried out, for example, in a stream of hot air in a fan-forced drier or else in a drying oven in a stream of inert gas, in particular in a stream of nitrogen or carbon dioxide. Drying may be carried out at atmospheric pressure or under reduced pressure, preferably 0.01-0.08 MPa.

In the above-described process comprising the steps a) to e), the catalyst is irradiated with microwaves before, during or after one of the steps a) to e).

One embodiment of the process of the invention comprises exposing the untreated catalyst support to microwave irradiation before the impregnation step a).

- 5 As an alternative thereto, the catalyst can be irradiated with the microwaves after the impregnation step a) and before the fixing step b).

10 It is also possible to use the microwaves during the reduction step c), particularly when the reduction is carried out in the gas phase.

Furthermore, the catalyst can also be irradiated with microwaves and heated in this way only after step e).

15

The catalyst is usually irradiated at a radiation frequency of from 300 MHz to 30 GHz; irradiation is preferably carried at 2.45 GHz. A microwave power of 10-2000 W, preferably 180-900 W, in particular 20 300-600 W, and an irradiation time of up to 10 hours, preferably 1-60 min, in particular 5-15 min, have been found to be useful.

25 The catalyst which can be produced by steps a) to e) of the process of the invention and the microwave irradiation essential to the invention comprises, based on the total mass of the catalyst, 0.2-2.5% by weight, preferably 0.6-1.5% by weight, of palladium, 0.2-2.5% by weight, preferably 0.3-1.0% by weight, of gold and 30 0.1-10% by weight of alkali metal, preferably 1.0-4.0% by weight of alkali metal, in particular potassium.

35 The preparation of vinyl acetate is carried out by passing acetic acid, ethylene and oxygen or oxygen-containing gases at temperatures of from 100 to 220°C, preferably from 120 to 200°C, and pressures of from 0.1 to 2.5 MPa, preferably from 0.1 to 2 MPa, over the catalyst of the invention. Here, unreacted components

can be circulated. In some cases it is also advantageous to dilute the reaction system with inert gases such as nitrogen or carbon dioxide. Carbon dioxide is particularly suitable for dilution in a circulation mode of operation since it is formed in any case during the reaction.

It has been found to be useful to carry out the preparation of the vinyl acetate in a stirred reactor, namely a Berty reactor, in circulation mode in the gas phase at a constant oxygen conversion of about 45%. The reactor is first charged with the catalyst. Subsequently, a measured amount of acetic acid together with ethylene and oxygen diluted with nitrogen is introduced and the temperature is raised to the desired value by means of a heating mantle. The reaction is usually stopped after about 18 hours, provided that it has been possible to set a temperature at which the oxygen conversion is constant at 45%. The composition of the product mixture is determined by means of gas chromatography.

The higher selectivity achievable using the catalysts of the invention and the greater space-time yield can in practice be utilized in two ways: firstly in that a greater amount of vinyl acetate can be produced per unit volume and unit time in existing plants while retaining all other reaction conditions. Owing to the higher selectivity, the product mixture taken from the reaction also has a higher proportion of vinyl acetate and contains less by-products, in particular carbon dioxide. This makes the work-up, i.e. the isolation of the vinyl acetate, easier because, for example, the amount of carbon dioxide to be separated off is lower and accordingly the loss of entrained ethylene associated with the removal of carbon dioxide also drops. This makes it possible to save starting material. The principle of the work-up of the product

mixture subsequent to the preparation of vinyl acetate is described, for example, in EP-A-0 423 658.

5 The second possible way of utilizing the improved properties of the catalysts of the invention is to lower the reaction temperature in the preparation of vinyl acetate while maintaining the same space-time yield. A lower reaction temperature in turn has a positive effect on the total operational life of the catalyst.

Examples 1 and 2:

15 The catalyst support used is a spherical silicon dioxide from Südchemie AG having a diameter of 7 mm and a pore volume of 324 ml of water/g. 250 ml of the support are impregnated with an aqueous solution comprising sodium tetrachloropalladate and sodium tetrachloroaurate. The supports are subsequently dried in hot air at a temperature which does not exceed 20 100°C. The supports treated in this way are impregnated with an aqueous sodium hydroxide solution. The volume of the sodium hydroxide solution is equal to the dry absorptive capacity of the supports. After the first stage, the support which has been treated with base is 25 allowed to stand for 4 hours and is subsequently poured into a second sodium hydroxide solution. The volume of this second sodium hydroxide solution is likewise equal to the dry absorptive capacity of the support material. After the second treatment, the material which has been 30 treated with base is allowed to stand for an additional time of about 16 hours. After the fixing procedure, the material which has been treated with base is washed thoroughly with distilled water. The catalyst is dried in a constant stream of nitrogen at a temperature of 35 not more than 150°C. The dried catalysts are subsequently reduced with ethylene at a temperature of 150°C. The reducing gas comprises 5% of ethylene in nitrogen and is passed over the catalysts for 5 hours

at atmospheric pressure. The reduced catalyst is impregnated with an aqueous solution comprising 10 g of potassium acetate and having a volume which corresponds to the absorptive capacity of the support. The catalysts are dried at a temperature of not more than 150°C. Subsequently, the catalyst is irradiated at room temperature with microwaves having a frequency of 2.45 GHz at a microwave power of 600 W for 10 minutes.

10 **Comparative Example:**

The catalyst is produced as described in Example 1 but the irradiation with microwaves is omitted after the last drying step.

15 **Preparation of vinyl acetate:**

The catalysts produced in Examples 1 and 2 and in the Comparative Example are used to prepare vinyl acetate in the gas phase from ethylene, acetic acid and oxygen-containing gases in a Berty reactor.

20 The experimental results are summarized in Table 1 below.

Table 1:

Preparation of vinyl acetate using the catalysts obtained in Examples 1 and 2 and in the Comparative Example

Example	Space-time yield (g V A M / L _{catalyst} ·h)	CO ₂ selectivity (% based on the amount of ethylene reacted)
1	746	8.8
2	742	8.9
comparative	715	9.5

Claims

1. A process for producing a catalyst for the preparation of vinyl acetate in the gas phase from ethylene, acetic acid and oxygen or oxygen-containing gases, which catalyst comprises palladium and/or its compounds, gold and/or its compounds and also alkali metal compounds on a particulate, porous support and is produced by
- impregnating the support with soluble palladium and gold compounds,
 - converting the soluble palladium and gold compounds into insoluble palladium and gold compounds by addition of an alkaline solution to the support,
 - reducing the insoluble palladium and gold compounds on the support by means of a reducing agent in the liquid or gaseous phase,
 - impregnating the support with at least one soluble alkali metal compound and
 - finally drying the support at a maximum of 150°C,
- wherein the catalyst is irradiated with microwaves before, during or after one of the process steps a) to e).
2. The process as claimed in claim 1, wherein the irradiation of the catalyst is carried out at a radiation frequency of from 300 MHz to 30 GHz, preferably at 2.45 GHz, and a microwave power of from 10 to 2000 W, preferably 180-900 W, in particular 300-600 W and an irradiation time of up to 10 hours, preferably 1-60 min, in particular 5-15 min.



3. The process as claimed in claim 1 or 2, wherein the catalyst support is subjected to the microwave irradiation prior to the impregnation step a).
- 5 4. The process as claimed in claim 1 or 2, wherein the catalyst is irradiated with the microwaves after the impregnation step a) and before the fixing step b).
- 10 5. The process as claimed in claim 1 or 2, wherein microwave irradiation is carried out during the reduction step c), preferably when the reduction is carried out in the gas phase.
- 15 6. The process as claimed in claim 1 or 2, wherein, the catalyst is irradiated with microwaves after step e).
- 20 7. The process as claimed in claim 1, wherein, in step c), the insoluble palladium and gold compounds on the support are either treated in the liquid phase at a temperature of 0-90°C, preferably 15-25°C, with hydrazine, formic acid or an alkali metal borohydride, preferably sodium
25 borohydride, as reducing agent or else in the gas phase at a temperature of 40-260°C, preferably 70-200°C, with hydrogen, ethylene, propylene, isobutylene or butylene as reducing agent.
- 30 8. The process as claimed in claim 1, wherein, in step d), the support is impregnated with an aqueous solution of an alkali metal compound, preferably an alkali metal carboxylate, in particular potassium acetate.
- 35 9. A catalyst for preparing vinyl acetate in the gas phase from ethylene, acetic acid and oxygen or oxygen-containing gases, which comprises palladium



and/or its compounds, gold and/or its compounds and also alkali metal compounds on a particulate, porous support and is obtainable by the process as claimed in one or more of claims 1-8.

- 5
10. A catalyst as claimed in claim 9 which comprises, based on the total mass of the catalyst, 0.2-2.5% by weight, preferably 0.6-1.5% by weight of palladium, 0.2-2.5% by weight, preferably 0.3-1.0% by weight, of gold and 0.1-10% by weight of alkali metal, preferably 1.0-4.0% by weight of alkali metal, in particular potassium.
- 10
11. A process for preparing vinyl acetate in the gas phase from ethylene, acetic acid and oxygen and/or oxygen-containing gases in the presence of the catalyst as claimed in claim 9 or 10.
- 15
12. The process as claimed in claim 11 carried out at temperatures of from 100 to 220°C, preferably from 120 to 200°C, and pressures of from 0.1 to 2.5 MPa, preferably from 0.1 to 2 MPa.
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