PROCESS FOR THE PREPARATION OF PHENOL AND CYCLOHEXANONE

Process for the preparation of phenol and cyclohexanone which comprises: 

a) the synthesis of cyclohexylbenzene by the hydro-alkylation of benzene by contact with hydrogen or the alkylation of benzene with cyclohexene using Y zeolites; b) the selective aerobic oxidation of cyclohexylbenzene to the corresponding hydroperoxide catalyzed by N-hydroxy-derivatives in the presence of polar solvents; and c) the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone by homogeneous or heterogeneous acid catalysts; characterized in that the synthesis of cyclohexylbenzene takes place in the presence of a catalytic system comprising a Y zeolite and an inorganic ligand wherein the Y zeolite has a crystalline structure with openings consisting of (12) tetrahedra and the inorganic ligand is γ-alumina, and wherein said catalytic composition is characterized by a pore volume, obtained by adding the mesoporosity and macroporosity fractions, greater than or equal to 0.7 cm$^3$/g, wherein at least 30% of said volume consists of pores with a diameter greater than 100 nanometers.
PROCESS FOR THE PREPARATION OF PHENOL AND CYCLOHEXANONE

FIELD OF THE INVENTION

The present invention relates to a process for the preparation of phenol and cyclohexanone.

More specifically, the present invention relates to a process for the preparation of cyclohexylbenzene and its transformation to phenol and cyclohexanone.

Cyclohexylbenzene is prepared by contact between benzene and hydrogen under hydro-alkylation conditions or by the alkylation of benzene with cyclohexene. Both processes are mediated by new catalysts described and claimed herein for this purpose. The cyclohexylbenzene is converted to the corresponding hydroperoxide by selective aerobic oxidation under mild conditions in the presence of a specific catalytic system. The hydroperoxide is then transformed to phenol and cyclohexanone in a process catalyzed by acids.

ART PRIOR TO THE INVENTION

The industrial production of phenol is based on the Hock process, which involves the autoxidation of cumene to the corresponding hydroperoxide and its subsequent acid-catalyzed decomposition to phenol and acetone. (Ullman's Encyclopedia of Industrial Organic Chemicals, Vol. A9, 1958, 225, Wiley-VCH).

The most complex phase which most widely influences the whole process is the autoxidation, in which the
hydroperoxide formed acts as initiator of the radical chain by decomposition to cumyloxyl radical. The latter can form cumyl alcohol by the hydrogen abstraction from cumene or undergo β-scission giving acetophenone and methyl radical. These aspects create various drawbacks which influence the whole process. The selectivity in the formation of the hydroperoxide decreases to the same extent to which the hydroperoxide itself acts as initiator. On the other hand, the decomposition of the hydroperoxide increases with the conversion and temperature. A high conversion causes a higher concentration of hydroperoxide and therefore a greater decomposition and lower selectivity. Furthermore, the methyl radical, formed in the β-scission of the cumyloxyl radical, is oxidized under the reaction conditions to formic acid. The latter catalyzes the decomposition of the hydroperoxide to phenol, which inhibits the oxidation process. In industrial processes, the presence of formic acid therefore creates the necessity of operating in the presence of a base, to neutralize the carboxylic acid. This problem also arises in the oxidation of other alkyl aromatic derivatives, for example in the oxidation of sec-butylbenzene in which significant quantities of acetic acid are formed (PCT/US2008/079150).

In order to eliminate or reduce these
disadvantages, various expedients have been taken into consideration, such as the use of suitable metallic complexes as catalysts or co-catalysts, which increase the conversion rate and allow to work at lower temperatures at which the hydroperoxide is more stable (Ishii, Y. at al. J. MoI. Catalysis A, 1987, 117, 123). The higher thermal stability of the hydroperoxides at low temperatures, however, is negatively balanced by the redox decomposition caused by the metal salts.

These catalytic systems have consequently proved to be inadequate for the preparation of hydroperoxides whereas they are of great industrial interest for the preparation of other oxygenated products (alcohols, aldehydes, ketones, carboxylic acids).

In the presence of N-hydroxyphthalimide (NHPI), aldehydes have proved to have a considerable activity for the production of hydroperoxide of alkyl aromatic compounds under mild aerobic conditions, with a high conversion and selectivity. (Minisci et al. PCT/EP07/008341; Minisci et al. MI2008A000460).

The use of N-hydroxy-derivatives has definite advantages with respect to non-catalyzed autoxidations, but also various disadvantages deriving from the decomposition of the initiators.

The Applicants have recently found that N-hydroxyphthalimide can catalyze the peroxidation of alkyl aromatic compounds under mild conditions if the aerobic oxidation is carried out in the presence of a moderate quantity of polar solvents (ketones, nitriles, esters, dialkyl carbonates and tertiary alcohols) which are completely stable under the operative conditions (Minisci et al. MI2008A000461). The polar solvent has a key function in favouring the dissolution of the organic catalyst, otherwise essentially insoluble under the operative conditions necessary for guaranteeing a high selectivity to hydroperoxide (temperature preferably lower than 110°C). Under the same operating
conditions, in the absence of N-hydroxy-derivative, there is no significant reaction.

The selectivity of this process to hydroperoxide is extremely high. The catalyst remains unaltered and can be easily recovered at the end of the reaction by means of crystallization and/or extraction with water or adsorption on various substrates.

In the peroxidation of cumene, acetone is the most advantageous among polar solvents as it is obtained as co-product during the acid decomposition of the hydroperoxide to phenol.

The demand for phenol, however, is constantly growing with respect to that for acetone. There is consequently a growing interest in processes for the production of phenol which avoid the formation of acetone.

The peroxidation of sec-butylbenzene is interesting as the cost of propylene, with respect to that of butenes, is continuously increasing and the offer is lower than the market request.

Cyclohexylbenzene is even more interesting. It has also been possible to carry out the peroxidation process on this derivative with the method object of the present invention. The corresponding cyclohexanone, obtained together with phenol, is of great industrial interest for the production of caprolactone (precursor...
of nylon 6) and adipic acid. Furthermore, the possibility of converting phenol to cyclohexanone by hydrogenation, and cyclohexanone to phenol by dehydrogenation (Sheldon et al. Tetrahedron 2002, page 9055) allows the production to be programmed on the basis of the variation in the market requests for the two products.

It is known that cyclohexylbenzene can be produced by reacting benzene with hydrogen in the presence of a catalyst based on ruthenium and nickel supported on beta zeolite (US patent N° 5,053,571); or in the presence of carbon monoxide and X or Y zeolites containing palladium (US patent N° 5,146,024); or in the presence of a bifunctional catalyst which includes a molecular sieve of the MCM-22 family and a metallic catalyst selected from palladium, ruthenium, nickel, cobalt or a mixture of these (US patent N° 6,037,513).

More recently, it has been found that the efficiency of the bifunctional catalyst described above is increased by supporting the metal on an organic oxide, used in the form of a composite with molecular sieves. This catalyst leads to a greater selectivity to mono- and di-cyclohexylbenzene, reducing the formation of cyclohexane (PCT/EP2008/006072).

An alternative method for the preparation of cyclohexylbenzene is the Friedel-Crafts reaction
between benzene and cyclohexene. These types of processes are being more and more frequently carried out by means of heterogeneous acid catalysis, preferably based on zeolites.

The Applicants have recently developed an innovative Y zeolite, belonging to the group of large-porosity zeolites, which includes γ-alumina as inorganic ligand (Bencini et al. Patent application U.S.A. 2006/0258893 Al).

This catalyst has proved to be particularly effective in the transalkylation of polyalkylated aromatic hydrocarbons, under such operative conditions that the reaction takes place at least partially in liquid phase.

The present invention relates to the application of this catalyst system to the alkylation reaction of benzene with cyclohexene and, in the presence of a hydrogenating metal, to the hydro-alkylation reaction of benzene by contact with hydrogen.

DESCRIPTION OF THE INVENTION

The object of the present invention, better described in the enclosed claims, therefore relates to:

1) the preparation of cyclohexylbenzene by the hydro-alkylation of benzene by contact with hydrogen or alkylation of benzene with cyclohexene using innovative Y zeolites as catalysts; 2) the selective aerobic
oxidation of cyclohexylbenzene to the corresponding hydroperoxide catalyzed by N-hydroxy-derivatives in the presence of polar solvents; 3) the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone by means of homogeneous or heterogeneous acid catalysts;

**Preparation of cyclohexylbenzene**

The cyclohexylbenzene is prepared according to two different methods. The first method comprises the alkylation reaction of benzene with cyclohexene in the presence of a catalyst based on Y zeolite prepared as described in US patent 2006/0258893.

The feeding mixture consisting of benzene and cyclohexene, before being introduced into the reactor, is treated with adequate dehydrating agents, for example alumina, to reduce the quantity of water contained therein to below 100 ppm, for example below 50 ppm.

The reaction temperature does not exceed 220°C and preferably ranges from 120 to 180°C, for example 150°C. The operating pressure ranges from 25 to 50 bar, for example 37 bar. The space velocity, expressed as LHSV (Liquid Hourly Space Velocity), ranges from 0.5 to 4 hours⁻¹, for example 2 hours⁻¹. In order to maximize the yield to mono-alkylated product, an excess of benzene is used, with benzene/cyclohexene molar ratios ranging
from 5 to 30, for example equal to 10.

The reaction conditions guarantee an almost total conversion of cyclohexene, with a high selectivity to the mono-alkylation product of benzene. Any possible polyalkylation products can be easily recycled by transalkylation in the presence of the same catalyst.

Alternatively, the cyclohexylbenzene is prepared by contact between benzene and hydrogen in the presence of a hydro-alkylating catalyst based on Y zeolite, prepared as described in US patent 2006/0258893, and containing a percentage of supported metal according to impregnation techniques known in literature (for example those indicated in US patent 5,146,024). The percentage of metal ranges from 0.05 to 1.5% by weight, preferably from 0.2 to 0.5%, and is equal for example to 0.3% by weight. The metal can be selected from palladium, platinum, nickel, ruthenium, palladium being preferred.

The reaction temperature does not exceed 300°C, and preferably ranges from 120 to 200°C, for example 130°C. The operating pressure ranges from 10 to 30 bar, for example 15 bar. The space velocity, expressed as WHSV (Weight Hourly Space Velocity), ranges from 1 to 4 hours⁻¹, for example 2 hours⁻¹. The hydrogen/benzene molar ratio ranges from 0.1:1 to 10:1, preferably from 0.5:1 to 5:1, for example 1:1.
The reaction conditions guarantee a good conversion of benzene, with a high selectivity to the hydro-mono-alkylation product.

Oxidation of cyclohexylbenzene

The cyclohexylbenzene, prepared according to one of the two procedures described above, is converted to the corresponding hydroperoxide by aerobic oxidation in the presence of a catalytic system, which includes N-hydroxyimides or N-hydroxsulphamides, preferably N-hydroxyphthalimides, for example N-hydroxyphthalimide associated with a polar solvent. The temperature does not exceed 130°C, and ranges for example from 50 to 110°C, preferably from 80 to 100°C. The polar solvent can be a C₃-C₁₀ acyclic or cyclic ketone (for example acetone, methylethylketone, 2-pentanone, 3-pentanone, methyl-t-butylketone, cyclopentanone), also the same cyclohexanone coming from the acid decomposition of the hydroperoxide, or other solvents such as nitriles, esters, tertiary alcohols, dialkylcarbonates, also stable under the reaction conditions.

The quantity of N-hydroxy-derivative catalyst ranges from 0.1 to 10% in moles, for example from 0.5 to 5%, preferably from 1 to 2% in moles.

The ratio between the volume of polar solvent with respect to the volume of alkylbenzene preferably varies within the range of 5:1 to 1:20.
The reaction is carried out with oxygen or air or mixtures of \( \text{N}_2/\text{O}_2 \) having a ratio between \( \text{N}_2 \) and \( \text{O}_2 \) ranging from 10:1 to 1:10, operating at pressures ranging from 1 to 20 bar.

At the end of the reaction, the N-hydroxy-derivative, in particular the more convenient N-hydroxyphthalimide, is for the most part recovered by crystallization from the reaction mixture from which the polar solvent has been removed by distillation. The small quantity of residual catalyst can be recovered by extraction with water or adsorption on various substrates from the reaction mixture.

Under the same operating conditions, in the absence of N-hydroxyphthalimide, the reaction does not take place to a significant degree.

**Scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone**

The hydroperoxide of cyclohexylbenzene, obtained in the oxidation phase of cyclohexylbenzene according to the procedure described above, is finally transformed to phenol and cyclohexanone by contact with an acid catalyst in homogeneous and heterogeneous phase.

After removing the polar solvent used in the oxidation reaction, and after recovering the catalyst, the reaction mixture is introduced into the scission reactor, preferably at a concentration of hydroperoxide.
obtained in the oxidation process, ranging for example from 20 to 30%. Alternatively, the oxidation reaction mixture can be concentrated to up to 80% of hydroperoxide before being introduced into the scission reactor, by removal of the cyclohexylbenzene at reduced pressure. Alternatively, the oxidation reaction mixture can be diluted with inert solvent which favours the removal of the heat developed.

The scission reaction can be carried out in a distillation unit. The process is carried out at a temperature ranging from 0 to 150°C, preferably from 20 to 90°C. The pressure preferably ranges from 1 to 20 bar.

Protic acids can be used as homogeneous catalysts (for example sulphuric acid, phosphoric acid, chloride acid, p-toluenesulphonic acid) or Lewis acids (for example ferric chloride, zinc chloride, boron trifluoride). Acid zeolites such as, for example, beta zeolites, zeolites Y, X, ZSM-5, ZSM-12 or mordenite, can be used as heterogeneous catalysts.

The mixture deriving from the scission is subjected to distillation to recover cyclohexanone, phenol and non-reacted cyclohexylbenzene.

The phenol can be converted to cyclohexanone by hydrogenation and the cyclohexanone to phenol by dehydrogenation.
The following examples are provided for illustrative but non-limiting purposes for the process of the present invention.

**Example 1**

For the alkylation of benzene with cyclohexene, the catalyst based on Y zeolite prepared as described in Example 1 of Patent Application USA 2006/0258893, is used as alkylation catalyst. The reactor used for the catalytic test is of the Berty type, consisting of a reaction chamber having a capacity of 250 cm³ inside which there is a 50 cm³ drum into which the above catalyst is charged. The head of the reactor is situated in the upper part of the reaction chamber, and supports a rotor which rotates by means of a magnetic joint. The reactor is equipped with a temperature and pressure regulation system. Before being introduced into the reactor, the feeding mixture is passed through an alumina column to reduce the quantity of water contained therein to below 50 ppm and is then fed in continuous to the reactor. The conditions under which the test is carried out are the following: reaction temperature equal to 150°C, reaction pressure equal to 37 bar, space velocity expressed as LHSV equal to 2 hours⁻¹, benzene/cyclohexene molar ratio equal to 10.

The effluent from the reactor is collected in a tank and analyzed by means of gas chromatography using an HP
5890 Series 2 instrument equipped with a capillary column with a Carbovax 2OM stationary phase and a detector of the flame ionization type (FID).

Under the above reaction conditions, a conversion of cyclohexene of 99.6% is obtained, with a selectivity to cyclohexylbenzene of 87.7% and a selectivity to useful aromatic compounds (intended as the sum of the desired product cyclohexylbenzene and dicyclohexylbenzenes which can be recovered by transalkylation) of 98.3%.

Example 2

For the hydro-alkylation of benzene, the catalyst based on Y zeolite prepared as described in Example 1 of Patent Application USA 2006/0258893 and containing 0.3% of palladium supported according to impregnation techniques known in literature, is used as catalyst. The experimental device used is the same as that described in Example 1. The conditions under which the test is carried out, after activation of the metal by pretreatment with hydrogen, are the following: reaction temperature equal to 120°C, reaction pressure equal to 15 bar, space velocity expressed as WHSV equal to 2 hours⁻¹, hydrogen/benzene molar ratio 1:1. The effluent from the reactor is collected in a tank and analyzed by means of gaschromatography using an HP 5890 Series 2 instrument equipped with a capillary column with a
Carbovax 20M stationary phase and a detector of the flame ionization type (FID).

Under the above reaction conditions, a conversion of benzene equal to 52.5% is obtained, with a selectivity to cyclohexylbenzene of 41.6%.

Example 3

A solution consisting of 5 mL of cyclohexylbenzene (29.4 mmoles) prepared as described in Examples 1 or 2, 1.90 mL of acetonitrile and 0.29 mmole of N-hydroxyphthalimide is stirred at 70°C for 24 hours in an oxygen atmosphere at a pressure of 1 bar. ¹H-NMR analysis of the reaction mixture indicated a conversion of cyclohexylbenzene equal to 28% with a selectivity to cyclohexylbenzene hydroperoxide of 99% (result confirmed by iodometric titration, GC-MS analysis in the presence of an internal standard after reduction of the hydroperoxide to the corresponding alcohol with PPh₃ and HPLC analysis of the reaction mixture without any treatment). There was no decomposition of the N-hydroxyphthalimide. The acetonitrile is removed by distillation and 0.52 mmoles of N-hydroxyphthalimide are recovered.

Example 4

The same procedure is adopted as described in Example 3, using 2-pentanone at 100°C instead of acetonitrile, leaving the mixture to react for 6 hours.
`H-NMR analysis of the reaction mixture indicated a conversion of cyclohexylbenzene equal to 35% with a selectivity to cumyl hydroperoxide of 99% (result confirmed by iodometric titration and HPLC analysis with an internal standard). 0.51 mmoles of N-hydroxyphthalimide are recovered.

**Example 5**

The same procedure is adopted as described in Example 3, using propionitrile at 100°C instead of acetonitrile, leaving the mixture to react for 6 hours. `H-NMR analysis of the reaction mixture indicated a conversion of cyclohexylbenzene equal to 36% with a selectivity to cumyl hydroperoxide of 99% (result confirmed by iodometric titration and HPLC analysis with an internal standard). 0.51 mmoles of N-hydroxyphthalimide are recovered.

**Example 6 - Comparative**

The same procedure is adopted as described in Example 4, without N-hydroxyphthalimide. There is no significant conversion of cyclohexylbenzene after 6 hours. After 24 hours, the conversion is high, but the selectivity to hydroperoxide decreases drastically, due to the decomposition of the hydroperoxide.

**Example 7 - Comparative**

The same procedure is adopted as described in Example 3, without acetonitrile. The conversion of
cyclohexylbenzene is < 1%.

**Example 8 - Comparative**

The same procedure is adopted as described in Example 4, without 2-pentanone. The conversion of cyclohexylbenzene after 6 hours is < 10%.
CLAIMS

1. A process for the preparation of phenol and cyclohexanone which comprises:
   a. the synthesis of cyclohexylbenzene by the hydro-alkylation of benzene by contact with hydrogen or the alkylation of benzene with cyclohexene using Y zeolites;
   b. the selective aerobic oxidation of cyclohexylbenzene to the corresponding hydroperoxide catalyzed by N-hydroxy-derivatives in the presence of polar solvents; and
   c. the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone by homogeneous or heterogeneous acid catalysts;

characterized in that the synthesis of cyclohexylbenzene takes place in the presence of a catalytic system comprising a Y zeolite and an inorganic ligand wherein the Y zeolite has a crystalline structure with openings consisting of 12 tetrahedra and the inorganic ligand is γ-alumina, and wherein said catalytic composition is characterized by a pore volume, obtained by summing the mesoporosity and macroporosity fractions, greater than or equal to 0.7 cm³/g, wherein at least 30% of said volume consists of pores with a diameter greater than 100 nanometers.

2. The process according to claim 1, wherein the
catalytic composition comprising Y zeolite and the inorganic ligand has an apparent density lower than 0.5 cm\(^3\)/g.

3. The process according to claim 1 or 2, wherein the catalytic composition comprising Y zeolite and the inorganic ligand is in the form of particles having a diameter larger than 1.8 mm.

4. The process according to any of the previous claims, wherein the feeding mixture of step (a) is pretreated with dehydrating agents to reduce the quantity of water to below 100 ppm.

5. The process according to any of the previous claims, wherein the synthesis of cyclohexylbenzene is effected by the alkylation of benzene with cyclohexene at a temperature ranging from 120 to 180\(^\circ\)C.

6. The process according to claim 5, wherein the synthesis of cyclohexylbenzene is effected at a pressure ranging from 25 to 50 bar.

7. The process according to claim 5 or 6, wherein the synthesis of cyclohexylbenzene is effected at a space velocity, expressed as LHSV, ranging from 0.5 to 4 hours\(^{-1}\).

8. The process according to any of the previous claims from 5 to 7, wherein the synthesis of cyclohexylbenzene is carried out with a benzene/cyclohexene ratio ranging from 5 to 30.
9. The process according to any of the previous claims from 1 to 4, wherein the cyclohexylbenzene is produced by contact between benzene and hydrogen in hydrogen/benzene molar ratios ranging from 0.1:1 to 10:1.

10. The process according to claim 9, wherein the catalyst for the synthesis of cyclohexylbenzene contains a supported metal in a percentage ranging from 0.05 to 1.5% by weight.

11. The process according to claim 9 or 10, wherein the catalyst is pretreated with hydrogen.

12. The process according to claim 9, 10 or 11, wherein the metal is selected from palladium, platinum, nickel, ruthenium, palladium being preferred.

13. The process according to any of the previous claims from 9 to 12, wherein the synthesis of cyclohexylbenzene is carried out at a temperature ranging from 120 to 200°C.

14. The process according to any of the previous claims from 9 to 13, wherein the synthesis of cyclohexylbenzene is carried out at a pressure ranging from 10 to 30 bar.

15. The process according to any of the previous claims from 9 to 14, wherein the synthesis of cyclohexylbenzene is carried out at a space velocity, expressed as WHSV, ranging from 1 to 4 hours⁻¹.
16. The process according to any of the previous claims, wherein the feeding mixture of step (a) also contains dicyclohexylbenzene which is put in contact with benzene under transalkylation conditions to produce further cyclohexylbenzene.

17. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene of step (b) is carried out in the presence of oxygen, a catalytic system, which includes a hydroxyderivative selected from an N-hydroxyimide or an N-hydroxysulfamide, and a polar solvent.

18. The process according to claim 17, wherein the catalyst is selected from N-hydroxyphthalimide and N-hydroxysaccharin.

19. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene is carried out at a temperature lower than 130°C.

20. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene is carried out with oxygen, air or mixtures of N₂/O₂ having a ratio between N₂ and O₂ ranging from 10:1 to 1:10, at a pressure ranging from 1 to 20 bar.

21. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene is carried out in the presence of a polar solvent selected from ketones, nitriles, esters, tertiary alcohols,
dialkyl carbonates.

22. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene is carried out with a quantity of N-hydroxyderivative catalyst ranging from 0.1 to 10% in moles with respect to the cyclohexylbenzene.

23. The process according to any of the previous claims, wherein the oxidation of cyclohexylbenzene is carried out with a ratio between the volume of polar solvent with respect to the volume of cyclohexylbenzene ranging from 5:1 to 1:20.

24. The process according to any of the previous claims, wherein the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone of step (c) is carried out in the presence of homogeneous or heterogeneous acid catalysts selected from protic acids and Lewis acids.

25. The process according to claim 24, wherein the protic acids are selected from sulfuric acid, phosphoric acid, chloride acid, p-toluenesulfonic acid, Amberlyst and the Lewis acids are selected from ferric chloride, zinc chloride, boron trifluoride.

26. The process according to claim 24, wherein the heterogeneous acids are selected from beta zeolite, Y zeolite, X zeolite, ZSM-5 zeolite, ZSM-12 zeolite or mordenite.
27. The process according to any of the previous claims, wherein the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone is carried out at a temperature ranging from 0 to 150°C.

28. The process according to any of the previous claims, wherein the scission of the hydroperoxide of cyclohexylbenzene to phenol and cyclohexanone is carried out at a pressure ranging from 1 to 20 bar.
### A. CLASSIFICATION OF SUBJECT MATTER

INV. C07C2/66  C07C2/74  C07C37/08  C07C39/04  C07C13/28  
C07C49/403  C07C45/53  C07C409/16

### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

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**X** Further documents are listed in the continuation of Box C

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- Special categories of cited documents
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