

The present invention relates to a process for the preparation of a phenol or of substituted phenols. It relates, more particularly, to a catalytic process for the hydroxylation of aromatic substrates.

5 The hydroxylation of phenol, substituted phenols or phenol ethers using hydrogen peroxide, in order to prepare diphenols, substituted diphenols or alkoxyphenols, is a known reaction.

10 French Patent FR 2 071 464 has described a process in which the reaction is catalysed by a strong acid, such as, for example, perchloric acid or sulphuric acid.

15 German Patent No. 2 410 742 has described a similar process to the above, in which hydrogen peroxide is used in the form of a virtually anhydrous organic solution.

These two processes are very advantageous and the first process is used industrially.

20 However, in the last few years, a means of catalysing the hydroxylation reaction with the aid of solids which are not dissolved in the medium has been sought, in order to simplify their separation from the reaction medium and their possible recycling, and to avoid saline by-products which usually form during
25 elimination of the dissolved acid catalysts.

Thus, French Patent FR 2 489 816 recommends the use of titanium silicalite as a heterogeneous catalyst for the hydroxylation of aromatic compounds using hydrogen peroxide.

30 In fact, this catalysis presents great difficulties regarding reproducibility. In addition, the fine size of the catalyst particles used makes their separation from the reaction medium very difficult and their recycling problematic whereas, in an industrial process,
35 it is essential to be able to recycle an expensive catalyst.

In order to overcome this problem of separation of the catalyst, it has been proposed, in the European patent application published under No. 200 260, to use

agglomerates of these fine titanium silicalite particles.

As a result of the difficulty of controlling the safety of plants during the use of hydrogen peroxide and the relative mediocrity of the yields obtained in the prior art, chemists have for several years sought to introduce a hydroxyl group directly onto the aromatic ring in the absence of any peroxide derivative. This chemistry has for a long time met with no success.

In addition, direct hydroxylation of an aromatic compound not containing a substituent or containing a deactivating substituent, such as benzene or halobenzenes, has hardly been described hitherto.

One publication describing the direct introduction of a hydroxyl group onto a benzene ring consists of a paper by IWAMOTO published in the Journal of Physical Chemistry, 87, 6, 1983.

This reaction for the hydroxylation of benzene is performed using nitrous oxide (N_2O) in the presence of a catalyst based on a metal oxide from groups V or VI of the Periodic Classification.

Vanadium oxide is the preferred oxide from among the metal oxides of groups V and VI of the Classification. It is preferable to use this oxide in the presence of a silica-based support in an amount by weight of between 1 and 10 % relative to the support. The support preferably consists of silica, alumina resulting mostly in the formation of a mixture of carbon oxides.

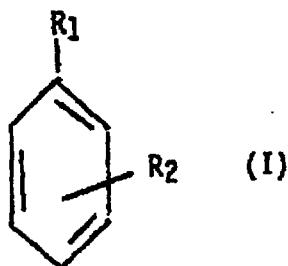
The IWAMOTO process was advantageous, but the use of these catalysts reduced its industrial appeal.

The publication by E. SUZUKI (Chemistry Letters pp. 953-956, 1988) is also known and this describes the production of phenol by reacting nitrous oxide with benzene, in the presence of an H-ZSM5 type zeolite. However, the yield and the efficiency of production of phenol remain poor.

Industry is thus still seeking a hydroxylation process which is capable of being applied to various aromatic substrates without the use of hydrogen peroxide.

The present invention represents a solution to

this problem. The subject of the invention is a process for the hydroxylation of an aromatic compound of formula (I):



in which

5 - R₁ represents:

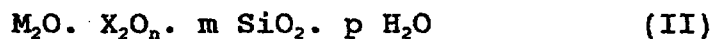
- an OH group,
- a bromine atom,
- a chlorine atom,
- a fluorine atom,
- 10 • a hydrogen atom,
- a linear or branched alkyl radical having 1 to 6 carbon atoms,
- a linear or branched alkoxy radical having 1 to 6 carbon atoms,

15 - R₂ represents:

- a hydrogen atom,
- a linear or branched alkyl radical having 1 to 6 carbon atoms,
- a linear or branched alkoxy radical having 1 to 6 carbon atoms,

20 characterized in that the aromatic compound of formula (I) is brought together at a temperature from 250°C to 500°C with nitrous oxide and a modified zeolite chosen from:

25 a) modified zeolites of ZSM-5 type of general formula (II), expressed in terms of oxide ratios:



in which

30 - M represents a cation chosen from hydrogen and the alkali metals, M being, at least in

part, a hydrogen atom;

- X represents Ga, Fe, B, In, Cr, Sc, Co, Ni, Be, Zn, Cu, Sb, As, V;

- n represents 2, 3 or 5 depending on the valence of the element X;

- m represents a number greater than or equal to 20;

- p represents a number from 0 to 40;

b) modified zeolites of ZSM-11 type of general formula (III), expressed in terms of oxide ratios:



in which:

- M represents a cation chosen from hydrogen and the alkali metals, M being, at least in part, a hydrogen atom;

- Z represents Al, Ga, Fe, B, In, Cr, Sc, Co, Ni, Be, Zn, Cu, Sb, As, V;

- n represents 2, 3 or 5 depending on the valence of the element Z;

- m represents a number greater than or equal to 20;

- x represents a number from 6 to 12.

The modified zeolites of formula (II) or (III) may simultaneously contain several of the elements represented by X or Z.

The modified zeolites of ZSM-5 and ZSM-11 type of formula (II) or (III), which are most generally used in the process of the invention, are those for which the symbol X or the symbol Z more particularly represents the trivalent elements (Al, Ga, Fe, B, In, Cr and Sc) or the divalent elements (Co, Ni, Be, Zn and Cu) indicated above, alone or combined with themselves or combined with the pentavalent elements (Sb, As and V) indicated above respectively.

The mode of preparation of ZSM-5 type zeolites may be that which is described in American Patent No. 3 702 888, replacing the aluminium compound by an inorganic gallium, iron, boron, indium, chromium, scandium,

cobalt, nickel, beryllium, zinc, copper, antimony, arsenic or vanadium compound or by a mixture of inorganic compounds of several of these elements.

5 The procedure can be carried out by preparing a solution containing a tetraalkylammonium hydroxide, for example such as tetrapropylammonium hydroxide; sodium oxide; a gallium, iron, boron, indium, chromium, scandium, cobalt, nickel, beryllium, zinc, copper, antimony, arsenic or vanadium oxide or a mixture of
10 several of these oxides; silica and water; the solution contains oxide ratios within the following ranges;

- OH^-/SiO_2 : from 0.07 to 10.0;
- tetraalkylammonium/tetraalkylammonium + Na^+ : from 0.2 to 0.95;
- 15 - $\text{H}_2\text{O}/\text{OH}^-$: from 10 to 300;
- $\text{SiO}_2/\text{X}_2\text{O}_n$ greater than or equal to 20.

An excess of tetraalkylammonium hydroxide may be used, which increases the OH^-/SiO_2 ratio indicated above. However, the excess hydroxide does not participate in the
20 reaction.

The treatment for the production of zeolite crystals generally consists in heating the above mixture to a temperature of 100°C to 200°C for a period of a few hours to 60 days under autogenous pressure.

25 The reaction conditions are preferably from about 10 hours to about 10 days at a temperature of 150°C to 200°C . The crystals are subsequently separated off from the liquid by cooling to room temperature, filtering and washing with water.

30 The zeolite is subsequently dried at about 100°C , generally for a period of several hours. The preparation of ZSM-5 type zeolites may be carried out by using compounds which generate the required oxides.

35 Gallium, iron, boron, indium, chromium, scandium, cobalt, nickel, beryllium, zinc, copper, antimony, arsenic or vanadium nitrates, sodium silicate, silica gels, silicic acid, sodium hydroxide or mixtures of these various compounds may thus be used with the corresponding oxides.

The mode of preparation of ZSM-11 type zeolites may be that which is described in American Patent No. 3 709 979, replacing the aluminium compound where appropriate by an inorganic gallium, iron, boron, indium, chromium, scandium, cobalt, nickel, beryllium, zinc, copper, antimony, arsenic or vanadium compound or by a mixture of inorganic compounds of several of these elements.

The procedure can thus be carried out as described above for ZSM-5 type zeolites by preparing a solution containing tetraalkylammonium oxide; sodium oxide; an aluminium, gallium, iron, boron, indium, chromium, scandium, cobalt, nickel, beryllium, zinc, copper, antimony, arsenic or vanadium oxide or a mixture of several of these oxides; silica and water, the oxide ratios of this solution being within the following ranges:

- $\text{Na}_2\text{O}/\text{SiO}_2$: from 0.05 to 0.7;
- tetraalkylammonium oxide/ SiO_2 : from 0.02 to 0.20;
- $\text{H}_2\text{O}/\text{Na}_2\text{O}$: from 50 to 800;
- $\text{SiO}_2/\text{Z}_2\text{O}_n$ greater than or equal to 20.

Modified zeolite is understood in the present text to refer to ZSM-5 and ZSM-11 type zeolites as defined above, at least a part of the cations M of which consists of hydrogen and preferably the majority or all of the cations M of which consist of hydrogen.

This may be obtained by treating the ZSM-5 and ZSM-11 type zeolites, in the formula for which the cations M are exclusively alkali metals or tetraalkylammonium ions, with an organic or inorganic acid.

By way of examples of inorganic acids which may be used to acidify zeolites, hydrochloric acid, sulphuric acid, nitric acid, perchloric acid and phosphoric acid may be mentioned.

By way of examples of organic acids which may be used to acidify zeolites, halosulphonic acids such as chlorosulphonic acid and fluorosulphonic acid, halomethanesulphonic acids such as trifluoromethanesulphonic

acid and halocarboxylic acids such as trichloroacetic acid, dichloroacetic acid, monochloroacetic acid, trifluoroacetic acid, difluoroacetic acid, monofluoroacetic acid and monobromoacetic acid may be mentioned.

5 According to one embodiment which is generally used, acidification of ZSM-5 or ZSM-11 type zeolite is carried out by passing through it from 10 cm³ to 100 cm³ of acid, having a normality of 0.1 N to 2 N, per gram of zeolite.

10 Such a throughput may take place in a single step or, preferably, in several successive steps.

In the formulae (II) and (III) for the modified ZSM-5 and ZSM-11 type zeolites the cation M, which optionally does not represent hydrogen, is preferably
15 sodium.

Modified ZSM-5 and ZSM-11 type zeolites may also be obtained by exchange of all or part of the alkali metals with the aid of an inorganic ammonium salt solution such as, for example, ammonium nitrate, ammonium
20 fluoride or ammonium chloride, followed by a thermal treatment for a few hours at temperatures from 300°C to 800°C in a stream of dry air, in order to create H⁺ sites.

Within the context of the present invention,
25 ZSM-5 type zeolites of formula (II) and ZSM-11 type zeolites of formula (III) in which formulae the SiO₂/X₂O_n or SiO₂/Z₂O_n ratio is between 20 and 500 are preferred.

This SiO₂/X₂O_n or SiO₂/Z₂O_n ratio in formulae (II) and (III) is more preferably between 40 and 300.

30 It is favourable to proceed with a treatment of the modified ZSM-5 and ZSM-11 type zeolites in order to improve their catalytic activity in the reaction for the hydroxylation of aromatic compounds of formula (I), using nitrous oxide.

35 This activation may especially consist in thermally treating the modified zeolites at a temperature of 300°C to 800°C, and preferably of 400°C to 700°C, for a few hours in a stream of a dry inert gas such as nitrogen. The thermal treatment may optionally be continued

for a few hours at a temperature within the regions indicated above, but under a stream of dry air.

5 The modified zeolites of ZSM-5 and ZSM-11 type may be used in various forms in the process of the invention: powder, especially in the laboratory tests, or products formulated as granules (for example cylinders or beads), beads, pellets or monoliths (lumps in the form of a beehive) which are obtained by extrusion, moulding, compacting or any other known type of process.

10 In practice, from an industrial viewpoint, the granule, bead and monolith forms show the greatest number of advantages, both from an efficiency viewpoint and from the viewpoint of ease of handling.

15 As the process of the invention is carried out in the vapour phase, the problem of recovering the catalyst is thus solved.

20 Among the compounds of formula (I) fluorobenzene, phenol, benzene, toluene, ortho-cresol, meta-cresol, para-cresol, anisole, bromobenzene, chlorobenzene, ortho-chlorophenol and para-chlorophenol may more particularly be mentioned.

Phenol, fluorobenzene and benzene are compounds of formula (I) which are very particularly advantageous to hydroxylate according to the process of the invention.

25 Nitrous oxide is used pure or mixed with an inert gas containing no oxygen, such as nitrogen.

30 The compound of formula (I) is preferably introduced as a mixture with nitrous oxide in a molar ratio of nitrous oxide relative to the compound of formula (I) of between 1 and 10.

35 According to a preferred implementation process, compound (I) is vaporized and mixed with nitrous oxide in the proportions defined beforehand, and the mixture is circulated over the zeolites of formula (II) or (III). The reaction preferably proceeds at a temperature between 300 and 500°C.

The gases obtained after the reaction, containing, where appropriate, a mixture of isomers, are condensed and separated by any technique known to a person

skilled in the art.

When it is applied to phenol, the process of the invention is particularly advantageous because it makes it possible to obtain the 3 dihydroxybenzenes, pyrocatechol, hydroquinone and resorcinol, whereas known processes for the hydroxylation of phenol lead almost exclusively to a pyrocatechol/hydroquinone mixture. The process thus provides an original route of access to resorcinol.

10 The examples which follow illustrate the invention.

EXAMPLE 1 - Preparation of a gallium ZSM-5 type zeolite

 A solution of 5.94 g of gallium nitrate hydrate ($\text{Ga}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$) and 16 g of H_2SO_4 (of 96 % concentration) in 200 g of water is run into a solution of 200 g of sodium silicate ($\text{SiO}_2/\text{Na}_2\text{O} = 3.22$) in 200 g of water.

 24 g of tetrapropylammonium bromide in 100 g of water are added to the gel thus obtained.

 The mixture is stirred for several minutes and is then placed in a 1-litre autoclave incorporating agitation, under autogenous pressure, for 4 days at 170°C.

 The white solid thus obtained is filtered off, washed with water and dried at 100°C.

 X-ray diffraction analysis of the powder confirms the formation of gallium ZSM-5 type zeolite.

 The $\text{SiO}_2/\text{Ga}_2\text{O}_3$ molar ratio is 80.

 Activation of this gallium zeolite consists in treating it thermally at 550°C, for 18 hours under a stream of dry nitrogen, and then for 5 hours at 550°C under a stream of dry air.

 After cooling, the zeolite is exchanged with a 1 M NH_4NO_3 solution and treated thermally for 10 hours in a stream of dry air, in order to form the H^+ sites.

EXAMPLE 2 - Preparation of an iron ZSM-5 type zeolite

 A solution of 50 g of sodium metasilicate ($\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$) in 50 g of water is run into a solution containing 2.1 g of ferric nitrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) in 50 g of water.

 The pH is adjusted to a strongly acidic value

using 5.0 g of 96 % H_2SO_4 .

6.5 g of tetrapropylammonium bromide in 10 g of water are added to the pale yellow gel obtained.

After vigorous stirring, the mixture is placed in
5 a stainless-steel autoclave which is closed and heated under autogenous pressure and with stirring for 5 days at 170°C .

The white solid obtained is filtered off, washed with water and dried at 100°C .

10 X-ray diffraction analysis of the powder confirms the formation of iron ZSM-5 type zeolite.

The $\text{SiO}_2/\text{Fe}_2\text{O}_3$ molar ratio is 90.

According to the same experimental procedure, iron zeolites are prepared having $\text{SiO}_2/\text{Fe}_2\text{O}_3$ molar ratios
15 of 120 and 150 respectively, by modifying the sodium metasilicate/ferric nitrate proportion and by adjusting the hydroxide content.

These iron zeolites are activated in the manner described in Example 1 in order to form the H^+ sites.

20 EXAMPLE 3 - Preparation of a cobalt ZSM-5 type zeolite

In a first step, a solution A of 0.27 g of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 0.6 g of H_2SO_4 in 15 g of water is prepared.

A solution B of 10 g of silica (marketed under
25 the trade name Q-Brand) in 10 g of water is subsequently prepared, followed by a solution C of 1 g of tetrapropylammonium bromide in 10 g of water.

Solutions A and B are first of all rapidly mixed and are stirred until a gel is obtained.

30 Solution C is then added to this gel with gentle stirring.

The gel thus obtained is placed in an autoclave incorporating agitation for 3 days at 170°C .

35 The white solid obtained is filtered off, washed with water and dried at 100°C .

X-ray diffraction analysis of the powder confirms the formation of cobalt ZSM-5 type zeolite.

The $\text{SiO}_2/2\text{CoO}$ molar ratio is 80.

This cobalt zeolite is activated in the manner

described in Example 1 in order to form the H^+ sites.

EXAMPLE 4 - Preparation of an aluminium ZSM-11 type zeolite

5 A solution A is prepared containing 55 g of sodium silicate ($SiO_2/Na_2O = 3.22$) and 1.3 g of 1,8-diaminooctane in 38 g of water.

A solution B is prepared containing 0.7 g of sodium aluminate in 40 g of water.

Solutions A and B are mixed.

10 The pH of the gel thus obtained is adjusted to 11 using sulphuric acid.

The gel is placed in an autoclave and stirred for 3 days at $220^\circ C$.

15 The white solid obtained is filtered off, washed with water and dried at $100^\circ C$.

X-ray diffraction analysis of the powder confirms the formation of aluminium ZSM-11 type zeolite.

The SiO_2/Al_2O_3 molar ratio is 40.

20 This aluminium zeolite is activated in the manner described in Example 1 in order to form the H^+ sites.

EXAMPLES 5 to 10 - Hydroxylation of fluorobenzene to fluorophenols

Experimental conditions

- vapour phase: continuous
 - 25 - catalyst: modified ZSM-5 or ZSM-11 type zeolite prepared in Examples 1, 2, 3 and 4 (see table below)
 - temperature: $400^\circ C$
 - hourly liquid space velocity (weight of substrate/weight of catalyst/hour: $1.5 h^{-1}$
 - 30 - fluorobenzene/ N_2 / N_2O molar ratios: 2/5/8
- 2 cm^3 of modified zeolite powder (approximately 1 g), dispersed in 4 cm^3 of quartz grains (less than 0.8 mm), are introduced into a tubular quartz reactor of length
- 35 16 cm and internal diameter 1.8 cm.

A bed of 10 cm^3 of glass beads, which makes it possible to homogenize the gaseous mixture, is subsequently added to the reactor.

The reactor thus charged is conditioned for

2 hours at 400°C under nitrogen in a tubular oven.

The reaction is carried out continuously by introducing 1.5 cm³/h of fluorobenzene, 1.44 l/h of N₂O and 0.9 l/h of nitrogen.

5

The results are indicated in the table below.

Examples	Modified zeolite	SiO ₂ /X ₂ O _n or SiO ₂ /Z ₂ O _n molar ratio	DC % of fluoro- benzene	CY % of fluoro- phenols
Example 5	Ga ZSM-5	80	4.1	75
Example 6	Co ZSM-5	80	1.3	89
Example 7	Fe ZSM-5	90	16.0	90
Example 8	Fe ZSM-5	120	9.8	93
Example 9	Fe ZSM-5	150	8.4	94
Example 10	Al ZSM-11	40	1.2	69

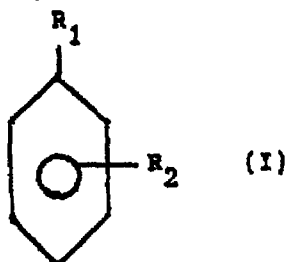
DC % = degree of conversion

CY % = yield relative to the fluorobenzene converted



CLAIMS

1. Process for the hydroxylation of an aromatic compound of formula (I):



in which:

5 - R_1 represents:

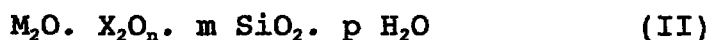
- an OH group,
- a bromine atom,
- a chlorine atom,
- a fluorine atom,
- 10 • a hydrogen atom,
- a linear or branched alkyl radical having 1 to 6 carbon atoms,
- a linear or branched alkoxy radical having 1 to 6 carbon atoms,

15 - R_2 represents:

- a hydrogen atom,
- a linear or branched alkyl radical having 1 to 6 carbon atoms,
- a linear or branched alkoxy radical having 1 to 20 6 carbon atoms,

characterized in that the aromatic compound of formula (I) is brought together at a temperature from 250°C to 500°C with nitrous oxide and a modified zeolite chosen from:

25 a) modified zeolites of ZSM-5 type of general formula (II), expressed in terms of oxide ratios:



in which:

- 30 - M represents a cation chosen from hydrogen and the alkali metals, M being, at least in part, a hydrogen atom;
- X represents Ga, Fe, B, In, Cr, Sc, Co, Ni, Be,



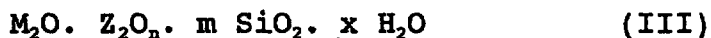
Zn, Cu, Sb, As, V;

- n represents 2, 3 or 5 depending on the valence of the element X;

- m represents a number greater than or equal to 20;

- p represents a number from 0 to 40;

b) modified zeolites of ZSM-11 type of general formula (III), expressed in terms of oxide ratios:



in which:

- M represents a cation chosen from hydrogen and the alkali metals, M being, at least in part, a hydrogen atom;

- Z represents Al, Ga, Fe, B, In, Cr, Sc, Co, Ni, Be, Zn, Cu, Sb, As, V;

- n represents 2, 3 or 5 depending on the valence of the element Z;

- m represents a number greater than or equal to 20;

- x represents a number from 6 to 12.

2. Process according to claim 1, characterized in that the modified zeolites of ZSM-5 and ZSM-11 type which are used in the process of the invention are those for which the symbol X or the symbol Z represents the trivalent elements (Al, Ga, Fe, B, In, Cr and Sc) or the bivalent elements (Co, Ni, Be, Zn and Cu) indicated in claim 1, alone or combined with themselves or combined with the elements Sb, As and/or V respectively.

3. Process according to either of claims 1 and 2, characterized in that the modified zeolites of ZSM-5 and ZSM-11 type are obtained by treating the zeolites of ZSM-5 and ZSM-11 type, in the formula for which the cations M are exclusively alkali metals or tetraalkylammonium ions, with an organic or inorganic acid.

4. Process according to claim 3, characterized in that an inorganic acid chosen from hydrochloric acid, sulphuric acid, nitric acid, perchloric acid and phosphoric acid is used.

5. Process according to claim 3, characterized in



that an organic acid chosen from halosulphonic acids such as chlorosulphonic acid and fluorosulphonic acid, halo-methanesulphonic acids such as trifluoromethanesulphonic acid and halocarboxylic acids such as trichloroacetic acid, dichloroacetic acid, monochloroacetic acid, tri-fluoroacetic acid, difluoroacetic acid, monofluoroacetic acid and monobromoacetic acid is used.

5
6. Process according to one of claims 3 to 5, characterized in that acidification of ZSM-5 or ZSM-11
10 type zeolite is carried out by passing through it from 10 cm³ to 100 cm³ of acid, having a normality from 0.1 N to 2 N, per gram of zeolite.

7. Process according to claim 6, characterized in that acidification of ZSM-5 or ZSM-11 type zeolite is
15 carried out in a single step or, preferably, in several successive steps.

8. Process according to either of claims 1 and 2, characterized in that in the formulae (II) and (III) for the modified ZSM-5 and ZSM-11 type zeolites the cation M,
20 which optionally does not represent hydrogen, is preferably sodium in which the alkyl radicals have 2 to 5 carbon atoms.

9. Process according to either of claims 1 and 2, characterized in that the modified zeolites of ZSM-5 and
25 ZSM-11 type are obtained by exchange of all or part of the alkali metals with the aid of a solution of an inorganic ammonium salt, followed by a thermal treatment for a few hours at temperatures from 300°C to 800°C in a stream of dry air, in order to create H⁺ sites.

30 10. Process according to one of claims 1 to 9, characterized in that ZSM-5 type zeolites of formula (II) and ZSM-11 type zeolites of formula (III) in which formulae the SiO₂/X₂O_n or SiO₂/Z₂O_n ratio is between 20 and 500 are used.

35 11. Process according to one of claims 1 to 10, characterized in that ZSM-5 type zeolites of formula (II) and ZSM-11 type zeolites of formula (III) in which formulae the SiO₂/X₂O_n or SiO₂/Z₂O_n ratio is between 40 and 300 are used.



12. Process according to one of claims 1 to 11, characterized in that the modified ZSM-5 and ZSM-11 type zeolites are treated at a temperature from 300°C to 800°C, and preferably from 400°C to 700°C, for a few hours in a stream of a dry inert gas such as nitrogen.
13. Process according to claim 12, characterized in that the thermal treatment is continued for a few hours at a temperature within the regions indicated above, but under a stream of dry air.
14. Process according to one of claims 1 to 13, characterized in that the compound of formula (I) is chosen from fluorobenzene, phenol, benzene, toluene, ortho-cresol, meta-cresol, para-cresol, anisole, bromobenzene, chlorobenzene, ortho-chlorophenol and para-chlorophenol.
15. Process according to one of claims 1 to 13, characterized in that the compound of formula (I) is chosen from fluorobenzene, phenol and benzene.
16. Process according to one of claims 1 to 15, characterized in that the compound of formula (I) is introduced as a mixture with nitrous oxide according to a molar ratio of nitrous oxide relative to the compound of formula (I) of between 1 and 10.