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(54) Title: ALKYLATION OF AROMATIC COMPOUNDS USING ZEOLITE ITQ-33

(57) Abstract: The present invention relates to a method of alkylation of aromatic compounds, characterized in that it comprises at least: a. feed of the zeolitic material ITQ-33 into a reactor, b. supplying the reactor with at least one alkylatable aromatic compound and at least one alkylating agent, c. leaving the zeolitic material ITQ-33 and the mixture of at least one alkylatable aromatic compound and at least one alkylating agent in contact for the time required for the reaction to take place.

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ALKYLATION OF AROMATIC COMPOUNDS USING
ZEOLITE ITQ-33

TECHNICAL FIELD OF THE INVENTION

5

[0001] Heterogeneous catalysis.

DESCRIPTION OF THE PRIOR ART

10 **[0002]** Ethylbenzene is a product of commercial interest for the production of styrene. Furthermore, cumene is a product of commercial interest that is used as starting material for the production of phenol and acetone. Numerous works have studied the alkylation of benzene with ethylene and propylene using acid catalysts. General information on the catalysts and processes used can be found in:

15 *"Encyclopedia of Chemical Processing and Design", J.J. McKezta and W.A. Cunningham Editors, V.14, pp. 33-55 (1982)".* The process of alkylation of benzene with propylene aims to achieve a high degree of conversion of propylene to alkylation products with long catalyst life. Moreover, the amount of n-propyl benzene (NPB) formed needs to be minimized. This is because NPB interferes

20 with the process of oxidation of cumene to produce phenol and acetone, and consequently a cumene stream with minimum possible NPB impurities is required. As it is difficult to separate cumene and NPB by conventional methods, such as distillation, it will be understood that the NPB yield must be the minimum possible, and in any case very low, during the alkylation of benzene with

25 propylene.

[0003] From the standpoint of the catalysts used in this process, conventionally acids such as H_3PO_4 , $AlCl_3$ and HCl have been used, although they present problems due to corrosion and loss of selectivity through formation of

30 polyalkylated products. Zeolites have also been used as catalysts of alkylation of aromatics and, for example, zeolite ZSM-5 is described in patent US-429457 as catalyst for alkylation of benzene with propylene. However, probably on account

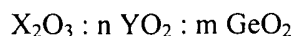
of the reduced diameter of its channels, this zeolite proves to have poor selectivity for the desired process, since it produces excessive amounts of n-propylbenzene. There are also many patents that describe the use of faujasite and modified faujasites as catalysts for the production of cumene by alkylation of benzene with propylene. More specifically, zeolite Y displays good activity at temperatures between 130 and 180°C with good selectivity for the desired products. However, this selectivity drops sharply when benzene conversion increases and it is therefore essential to work with high benzene/propylene ratios in the feed. This leads to high costs for benzene recycling. Zeolite BETA has also been claimed as catalyst for alkylation of benzene with propylene in various patents, for example: US-4891458, US-5030786, EP-432814, EP-439632, EP-629599. This zeolite is used commercially in this process and produces good results with respect to activity and selectivity, but its behaviour could be improved both with respect to selectivity for NPB, and with regard to catalyst stability. Other zeolites used in commercial processes for production of ethylbenzene and/or cumene are ZMS-5, MCM-22, mordenite, and zeolite Y. Further details can be found in two recent reviews "*Applied Catalysis A: General* 221 (2001) 283-294, and *Catalysis Today* 73 (2002) 3-22".

[0004] In the present invention, a method is described based on the use of a catalyst containing the zeolitic material ITQ-33 in the alkylation of aromatic products, in which said catalyst is not only active, but, in the particular case of cumene, produces a very low yield of n-propylbenzene and of products of oligomerization of propylene, greatly increasing the life of the catalyst relative to catalysts used at present.

DESCRIPTION OF THE INVENTION

[0005] The present invention relates to a method of alkylation of aromatic compounds which uses a zeolitic material designated ITQ-33.

The zeolitic material used as catalyst in the method of the present invention has a composition in its calcined, anhydrous form that is given by the following formula:



5

in which (n + m) is at least 5, X is a trivalent element, Y corresponds to one or more tetravalent elements other than Ge, the ratio Y/Ge is greater than 1, and it has an X-ray diffraction pattern whose principal lines for its synthesized, uncalcined form are as given in Table 1:

10 Table 1: Zeolite ITQ-33, uncalcined

(d ± 0.2) (Å)	Relative intensity
16.59	vs
11.63	vw
9.57	vw
8.31	vw
6.76	vw
4.81	vw
4.62	vw
4.27	w
3.64	w
3.15	w

where "vs" denotes relative intensity 80-100, "w" denotes relative intensity 20-40 and "vw" denotes relative intensity 0-20, calculated as a percentage relative to the peak with greatest intensity.

15

[0006] Once calcined, the crystal structure of zeolite ITQ-33 is characterized by an X-ray diffraction pattern whose most characteristic lines are shown in Table 2:

20

Table 2: Zeolite ITQ-33, calcined.

$(d \pm 0.2) (\text{\AA})$	I_{rel}
16.81	vs
11.50	vw
9.71	vw
8.41	vw
6.79	vw
4.84	vw
4.65	vw
4.24	vw
3.66	vw
3.17	vw

where "vs" denotes relative intensity 80-100, "w" denotes relative intensity 20-40 and "vw" denotes relative intensity 0-20, calculated as a percentage relative to the peak with greatest intensity.

[0007] Zeolite ITQ-33 can be synthesized in a fluoride medium or in an OH⁻ medium, using an organic structure-controlling agent, for example the hexamethonium cation, and forming a gel whose composition comprises a source of at least one T^{IV} element, among which Si and Ge are preferred, and of one or more T^{III} elements, among which Al, B, Fe and Ga are preferred, and preferably Al, B or a mixture of the two, with the ratio T^{IV}/T^{III} being at least 5. The synthesized zeolite is submitted to a process for removing the organic matter trapped inside the material, which is accomplished by extraction, thermal treatment at temperatures above 250°C for a period of time between 2 minutes and 25 hours, or a combination of both, so that, directly or by means of conventional ion exchanges, the acid form is obtained, which is the form preferred in the catalytic composition. The structure of zeolite ITQ-33 has a system of three-directional channels, with straight channels of ultra-large pore size parallel to the *c* axis, defined by circular openings formed by 18 atoms in tetrahedral coordination. These channels are interconnected by a system of channels of 10 in plane *ab*.

The zeolitic catalyst used in the present invention is pelletized by methods that are well known in the literature, using a diluent such as SiO₂ or Al₂O₃ or a clay, zirconia, magnesium oxide or mixture thereof, at zeolite/diluent ratios between 20 and 95 wt.% and preferably between 40 and 90 wt.%.

- 5 The resultant catalyst, once calcined at a temperature between 450 and 700°C, is used as the catalyst in the process of alkylation of aromatic compounds according to the present invention.

[0008] The method of alkylation of aromatic compounds that is described in
10 the present invention is characterized in that it comprises at least:

- a. feed of the zeolitic material ITQ-33 into a reactor,
- b. supplying the reactor with at least one alkylatable aromatic
15 compound and at least one alkylating agent,
- c. leaving the zeolitic material ITQ-33 and the mixture of at least one alkylatable aromatic compound and at least one alkylating agent in contact for the time necessary for the reaction to take
20 place.

Preferably, the alkylatable aromatic compound is selected from benzene, naphthalene, anthracene, phenanthrene, their substituted derivatives and combinations thereof. More preferably, said alkylatable aromatic compound is
25 benzene.

[0009] Also preferably, the alkylating agent is selected from olefins, alcohols, polyalkylated aromatic compounds and combinations thereof. According to a particular embodiment, the alkylating agent used in the method of
30 the present invention is an olefin that can be selected from ethylene, propylene and combinations thereof. According to this particular embodiment, said olefin and the alkylatable aromatic compound are, at least partially, in the liquid phase.

Said olefin can have between 2 and 20 carbon atoms. Moreover, preferably, the molar ratio of alkylatable aromatic compound to olefin is between 2 and 20. Said alkylatable aromatic compound can be selected from benzene, alkylbenzene, hydroxybenzene, alkoxybenzene, naphthalene, anthracene, phenanthrene and combinations thereof. Preferably, the alkylatable aromatic compound is benzene. According to a preferred embodiment, the alkylatable aromatic compound is benzene and the alkylating agent is ethylene. According to this embodiment, the compound that is obtained is preferably ethylbenzene. The molar ratio of the benzene/ethylene mixture can be between 2 and 20, and preferably between 2 and 15. Moreover, according to this preferred embodiment, the temperature at which the reaction is carried out can be between 60 and 350°C, and preferably between 80 and 300°C, and the pressure can be between 1.4 and 7.0 MPa, preferably between 1.4 and 4.1 MPa. The space velocity (WHSV) of the reagents can be between 0.1 and 150 h⁻¹, and preferably between 0.2 and 30 h⁻¹.

15

[0010] According to another preferred embodiment, the alkylatable aromatic compound is benzene and the alkylating agent is propylene. According to this embodiment, the compound that is obtained is preferably cumene. The molar ratio of the benzene/propylene mixture can be between 2 and 20, and preferably between 2 and 15. Moreover, according to this preferred embodiment, the temperature at which the reaction is carried out can be between 60 and 350°C, and preferably between 80 and 300°C, and the pressure can be between 1.4 and 7.0 MPa, preferably between 1.4 and 4.1 MPa. The space velocity (WHSV) of the reagents can be between 0.1 and 150 h⁻¹, and preferably between 0.2 and 30 h⁻¹.

25

[0011] In this method we can use a batch reactor, heating the catalyst, the benzene and the propylene in an autoclave with stirring at a temperature and a pressure (stated previously) sufficient to maintain, at least partially, a liquid phase. However, when using this catalyst, a system is preferred that operates continuously, using a fixed-bed reactor that operates in "up" or "downflow" conditions, or using a moving-bed reactor in which the catalyst and hydrocarbons function in co- or in counter-current. In a particular embodiment for the

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production of cumene, the benzene/propylene molar ratio in the feed can vary between 2 and 20, and preferably between 2 and 15, as stated previously. The heat of reaction can be controlled by supplying unreactive paraffins at different points of the catalyst bed, and the space velocity (WHSV) of the reagents is between 0.2 and 150 h⁻¹ and preferably between 0.5 and 15 h⁻¹.

[0012] Another particular embodiment of the present invention is that in which the alkylating compound is a polyalkylated aromatic compound. According to this embodiment, the alkylatable aromatic compound is an unalkylated aromatic compound. In this way, the alkylating compound can transfer at least one alkyl group to the alkylatable aromatic compound. According to this embodiment, the alkylating compound preferably has between 2 and 20 carbon atoms, and preferably between 6 and 20. The alkylatable aromatic compound according to this embodiment is selected from benzene, naphthalene, anthracene, phenanthrene, the corresponding substituted substances, the substituents being other than alkyl groups, and combinations thereof. Preferably, the polyalkylated aromatic compound (alkylating compound) is polyisopropylbenzene and the unalkylated aromatic compound (alkylatable compound) is benzene.

[0013] According to the method of the present invention, it was found that when the catalysts prepared with the crystal structure described above, zeolite ITQ-33, in their acid form, are employed in the alkylation of aromatic compounds with olefins, alcohols, aromatic compounds and combinations thereof, among others, and preferably when used as catalyst in the alkylation of benzene with ethylene or propylene, it proves to be a very active catalyst, and in the case of alkylation with propylene, with a low selectivity for the production of NPB, and a higher selectivity for alkylbenzenes and a lower selectivity for products of oligomerization of the olefin than were obtained with a commercial zeolite BETA used as reference.

30

BRIEF DESCRIPTION OF THE DRAWINGS

[0014] Fig. 1: X-ray powder diffraction pattern of zeolite ITQ-33, uncalcined;

[0015] Fig. 2: Conversion of propylene obtained with zeolites ITQ-33 (•)
5 and

[0016] BETA (o). $T = 125^{\circ}\text{C}$, $P = 3.5 \text{ MPa}$, $\text{WHSV} = 12 \text{ h}^{-1}$,
benzene/propylene = $3.5 \text{ (mol.mol}^{-1}\text{)}$.

10 [0017] A number of examples are given below, illustrating the preparation of the catalyst and its use in the alkylation of aromatic compounds with olefins, alcohols, aromatic compounds and combinations thereof, among others, illustrated with the alkylation of benzene with propylene.

15 EXAMPLES

Example 1: Synthesis of a sample of the zeolitic component ITQ-33.

[0018] Dissolve 3.484 g of GeO_2 in 14.543 g of hexamethonium hydroxide
20 solution (24.5 wt.%) and 8.035 g of hexamethonium bromide solution (50 wt.%). Add 1.043 g of aluminium isopropoxide and 14.205 g of tetraethyl-orthosilicate (TEOS). Finally, once the alkoxides have been hydrolysed, add 1.254 g of hydrofluoric acid solution (48 wt.%) and, while stirring, leave the mixture to evaporate until the reaction mixture reaches a final composition:

25
 $0.67 \text{ SiO}_2 : 0.33 \text{ GeO}_2 : 0.025 \text{ Al}_2\text{O}_3 : 0.15 \text{ R(OH)}_2 : 0.10 \text{ R(Br)}_2 : 0.30 \text{ HF} : 1.5 \text{ H}_2\text{O}$

where R is hexamethonium.

30

[0019] Heat the gel at 175°C in static conditions for 20 hours in Teflon-lined steel autoclaves. The solid obtained after filtration, washing with distilled water and drying at 100°C is ITQ-33.

- 5 **[0020]** The X-ray powder diffraction pattern of the solid obtained after filtration, washing and drying at 100°C is shown in Figure 1 and in Table 3.

Table 3: Zeolite ITQ-33, uncalcined

(d ± 0.2) (Å)	Relative intensity
16.59	vs
11.63	vw
9.57	vw
8.31	vw
6.76	vw
4.81	vw
4.62	vw
4.27	w
4.02	vw
3.86	vw
3.82	vw
3.64	w
3.33	vw
3.15	w
2.78	vw
2.75	vw
2.54	vw
2.38	vw
2.27	vw

[0021] The relative intensity of the lines is calculated as a percentage relative to the peak of highest intensity, and is regarded as very strong (vs) = 80-100, strong (s) = 60-80, medium (m) = 40-60, weak (w) = 20-40, and very weak (vw) = 0-20.

Example 2: Activation by calcination of the zeolitic component ITQ-33

[0022] The zeolite obtained in Example 1 was calcined in an air stream at 540°C for 3 hours. The X-ray diffraction pattern of the calcined material is shown in Table 4.

Table 4: Zeolite ITQ-33, calcined

(d ± 0.2) (Å)	Relative intensity
16.81	vs
11.50	vw
9.71	vw
8.41	vw
6.79	vw
4.84	vw
4.65	vw
4.24	vw
4.01	vw
3.84	w
3.81	vw
3.66	vw
3.36	w
3.17	vw
2.81	vw
2.56	vw
2.37	vw
2.27	vw

10 [0023] The relative intensity of the lines is calculated as a percentage relative to the peak of highest intensity, and is regarded as very strong (vs) = 80-

100, strong (s) = 60-80, medium (m) = 40-60, weak (w) = 20-40, and very weak (vw) = 0-20.

Example 3: Use of the zeolitic component ITQ-33 in the alkylation of benzene with propylene

5

[0024] The present example illustrates the use of a material prepared according to Example 1 as catalyst in the alkylation of benzene with propylene.

[0025] A sample with ratio $T^{IV}/T^{III} = 12$, prepared according Example 1,
10 was pelletized, selecting a particle size between 0.25 and 0.42 mm, for carrying out the reaction. The zeolite (0.75 g) was diluted with silicon carbide (0.59-0.84 mm) to give a final SiC/zeolite ratio of 2.8 by weight. The diluted catalyst was placed in a tubular steel reactor 1 cm in diameter, and it was calcined in an air stream following the method described in Example 2. Next, the temperature was
15 lowered to the reaction temperature of 125°C in a stream of N₂, the flow of N₂ was stopped and benzene was supplied until the pressure reached 3.5 MPa. At this point the reactor was switched off for feed of a mixture of benzene (400 µl) and propylene (90 µl), at a benzene/propylene molar ratio of 3.5, through a parallel line until a constant composition was achieved, and then the feed was again
20 passed through the reactor, this being regarded as the start of the reaction. The results for propylene conversion are presented in Figure 2, and are compared with those obtained in the same conditions with a commercial zeolite BETA (Zeolyst CP811) with ratio Si/Al = 13. The distribution of products obtained with ITQ-33 and with BETA at different reaction times is compared in Tables 5 and 6,
25 respectively.

Table 5: Selectivity in the alkylation of benzene with propylene at 125°C, B/P = 3.5 mol.mol⁻¹, WHSV prop = 12 h⁻¹, P = 3.5 MPa obtained with zeolite ITQ-33

Reaction time (min)	Conversion (%)	Selectivity relative to propylene (%)			
		Cumene	Di- + Trialkyls	NPB	Others
30	98.2	77.9	18.8	<0.01	1.7
120	97.9	76.1	22.1	<0.01	2.8
240	97.4	73.7	23.9	<0.01	3.3
480	96.5	68.9	25.6	<0.01	3.9

Table 6: Selectivity in the alkylation of benzene with propylene at 125°C, B/P = 3.5 mol.mol⁻¹, WHSV prop = 12 h⁻¹, P = 3.5 MPa obtained with zeolite BETA.

5

Reaction time (min)	Conversion (%)	Selectivity relative to propylene (%)			
		Cumene	Di- + Trialkyls	NPB	Others
30	98.4	89.9	8.8	<0.01	2.2
120	89.7	90.0	6.1	<0.01	4.0
240	78.1	90.2	4.7	<0.01	4.9
480	55.0	90.5	3.3	<0.01	5.8

[0026] The results in Figure 2 clearly show that zeolite ITQ-33, as claimed in this patent, is more active than the commercial zeolite BETA, maintaining a degree of conversion of propylene greater than 95% at 8 hours of reaction, whereas the conversion obtained with zeolite BETA at 8 hours of reaction is of the order of 55%. Comparing Tables 5 and 6, it can be seen that the selectivity for the unwanted product NPB obtained with ITQ-33 is less than 0.01%, and the selectivity for products other than alkylation products (others) is less than that obtained with zeolite BETA. The di- and trialkylated products, obtained in higher proportion with zeolite ITQ-33 could be converted to cumene in the associated transalkylation units.

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Example 4: Use of the zeolitic component ITQ-33 in the alkylation of benzene with propylene; effect of space velocity

[0027] This example shows the influence of the space velocity (WHSV) (24 h⁻¹) on the conversion and selectivity for the alkylation of benzene with propylene using the same catalysts as in Example 3, with the other reaction conditions the same as in Example 3.

Table 7: Selectivity in the alkylation of benzene with propylene at 125°C, B/P = 3.5 mol.mol⁻¹, WHSV prop = 24 h⁻¹, P = 3.5 MPa obtained with zeolite ITQ-33

Reaction time (min)	Conversion (%)	Selectivity relative to propylene (%)			
		Cumene	Di- + Trialkyls	NPB	Others
30	98.7	61.7	32.9	<0.01	5.4
120	94.9	63.8	30.7	<0.01	5.4
240	89.8	66.7	27.9	<0.01	5.4

Table 8: Selectivity in the alkylation of benzene with propylene at 125°C, B/P = 3.5 mol.mol⁻¹, WHSV prop = 24 h⁻¹, P = 3.5 MPa obtained with zeolite BETA.

Reaction time (min)	Conversion (%)	Selectivity relative to propylene (%)			
		Cumene	DIPB	NPB	Others
30	94.5	89.0	7.1	<0.01	3.9
120	69.1	90.4	4.8	<0.01	4.9
240	35.2	92.2	1.6	<0.01	6.2

The results presented in Tables 7 and 8 show that at this higher space velocity, the differences in activity between zeolite ITQ-33 and zeolite BETA are even greater than at a space velocity of 12 h⁻¹, the zeolite according to the present patent being much more active, while maintaining, for this zeolite, low selectivities for NPB

and for products different from those obtained by alkylation of benzene with propylene (others).

CLAIMS

1. Method of alkylation of aromatic compounds, characterized in that it comprises at least:

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- a. feed of the zeolitic material ITQ-33 into a reactor,
- b. supplying the reactor with at least one alkylatable aromatic compound and at least one alkylating agent,
- c. leaving the zeolitic material ITQ-33 and the mixture of at least one alkylatable aromatic compound and at least one alkylating agent in contact for the time necessary for the reaction to take place.

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2. Method of alkylation according to Claim 1, characterized in that the alkylatable aromatic compound is selected from benzene, naphthalene, anthracene, phenanthrene, their substituted derivatives and combinations thereof.

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3. Method of alkylation according to Claim 2, characterized in that the alkylatable aromatic compound is benzene.

4. Method of alkylation according to Claim 1, characterized in that the alkylating agent is selected from olefins, alcohols, polyalkylated aromatic compounds and combinations thereof.

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5. Method of alkylation according to Claim 4, characterized in that the alkylating agent is an olefin.

30

6. Method of alkylation according to Claim 5, characterized in that the olefin is selected from ethylene, propylene and combinations thereof.

7. Method of alkylation according to one of Claims 5 and 6, characterized in that at least one olefin and at least one alkylatable aromatic compound are at least partially in the liquid phase.

8. Method of alkylation according to one of Claims 5 to 7,
5 characterized in that the molar ratio of alkylatable aromatic compound to olefin is between 2 and 20.

9. Method of alkylation according to one of Claims 5 to 8, characterized in that the olefin has between 2 and 20 carbon atoms.

10

10. Method of alkylation according to one of Claims 5 to 9, characterized in that the alkylatable aromatic compound is selected from benzene, alkylbenzene, hydroxybenzene, alkoxybenzene, naphthalene, anthracene, phenanthrene and combinations thereof.

15

11. Method of alkylation according to Claim 10, characterized in that the alkylatable aromatic compound is benzene.

12. Method of alkylation according to one of the preceding claims,
20 characterized in that the alkylatable aromatic compound is benzene and the alkylating agent is ethylene.

13. Method of alkylation according to Claim 12, characterized in that the alkylated compound that is obtained is ethylbenzene.

25

14. Method of alkylation according to one of Claims 12 and 13, characterized in that the molar ratio of the benzene/ethylene mixture is between 2 and 20.

15. Method of alkylation according to Claim 14, characterized in that the molar ratio of the benzene/ethylene mixture is between 2 and 15.

30

16. Method of alkylation according to one of Claims 12 to 15, characterized in that it is carried out at a reaction temperature between 60 and 350°C.

5 17. Method of alkylation according to Claim 16, characterized in that it is carried out at a reaction temperature between 80 and 300°C.

18. Method of alkylation according to one of Claims 12 to 17, characterized in that it is carried out at a pressure between 1.4 and 7.0 MPa.

10

19. Method of alkylation according to Claim 18, characterized in that it is carried out at a pressure between 1.4 and 4.1 MPa.

20. Method of alkylation according to one of Claims 12 to 19, characterized in that the space velocity (WHSV) of the reagents is between 0.1 and 150 h⁻¹.

15

21. Method of alkylation according to Claim 20, characterized in that the space velocity (WHSV) of the reagents is between 0.2 and 30 h⁻¹.

20

22. Method of alkylation according to one of Claims 1 to 11, characterized in that the aromatic compound is benzene and the alkylating agent is propylene.

25 23. Method of alkylation according to Claim 22, characterized in that the alkylated aromatic compound that is obtained is cumene.

24. Method of alkylation according to one of Claims 22 and 23, characterized in that the benzene/propylene molar ratio is between 2 and 20.

30

25. Method of alkylation according to Claim 24, characterized in that the benzene/propylene molar ratio is between 2 and 15.

26. Method of alkylation according to one of Claims 22 to 25,
characterized in that it is carried out at a reaction temperature between 60 and
350°C.
- 5
27. Method of alkylation according to Claim 26, characterized in
that it is carried out at a reaction temperature between 80 and 300°C.
28. Method of alkylation according to one of Claims 22 to 27,
10 characterized in that it is carried out at a pressure between 1.4 and 7.0 MPa.
29. Method of alkylation according to Claim 28, characterized in
that it is carried out at a pressure between 1.4 and 4.1 MPa.
- 15
30. Method of alkylation according to one of Claims 22 to 29,
characterized in that the space velocity (WHSV) of the reagents is between 0.1
and 150 h⁻¹.
31. Method of alkylation according to Claim 30, characterized in
20 that the space velocity (WHSV) of the reagents is between 0.2 and 30 h⁻¹.
32. Method of alkylation according to one of Claims 1 to 4,
characterized in that the alkylating agent is a polyalkylated aromatic compound.
- 25
33. Method of alkylation according to Claim 32, characterized in
that the alkylatable aromatic compound is an unalkylated aromatic compound.
34. Method of alkylation according to one of Claims 32 and 33,
characterized in that the polyalkylated aromatic compound transfers at least one
30 alkyl group to the unalkylated aromatic compound.

35. Method of alkylation according to one of Claims 32 to 34, characterized in that the alkyl group of the polyalkylated aromatic compound has between 2 and 20 carbon atoms.

5 36. Method of alkylation according to Claim 35, characterized in that the alkyl group of the polyalkylated aromatic compound has between 6 and 20 carbon atoms.

10 37. Method of alkylation according to one of Claims 32 to 36, characterized in that the alkylatable aromatic compound is selected from benzene, naphthalene, anthracene, phenanthrene, the corresponding substituted substances with substituents other than alkyl groups, and combinations thereof.

15 38. Method of alkylation according to one of Claims 32 to 37, characterized in that the polyalkylated aromatic compound is a polyisopropylbenzene and the unalkylated aromatic compound is benzene.

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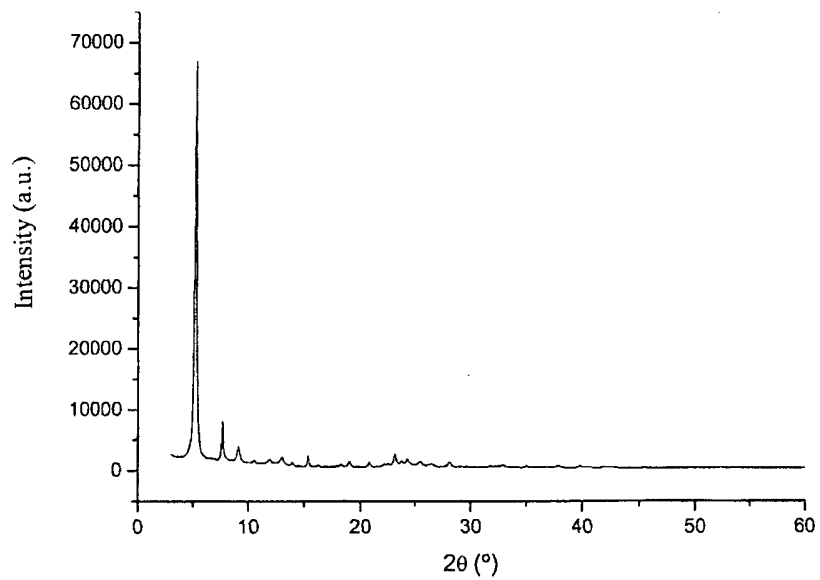


Figure 1

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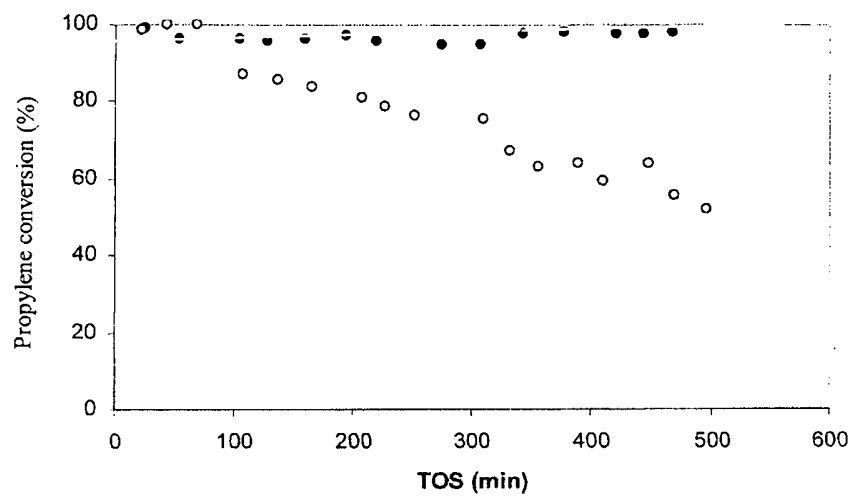


Figure 2

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2007/006550

A. CLASSIFICATION OF SUBJECT MATTER

INV. C07C2/66 C07C15/085 C07C15/073 C01B39/48 C01B39/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C07C C01B

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

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

EPO-Internal, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2006/075041 A (CONSEJO SUPERIOR INVESTIGACION [ES]; UNIV VALENCIA POLITECNICA [ES]; C) 20 July 2006 (2006-07-20) page 10, lines 18-20; claims, e.g. claim 26	1-38
Y	EP 1 252 927 A (CONSEJO SUPERIOR INVESTIGACION [ES]; UNIV VALENCIA POLITECNICA [ES]) 30 October 2002 (2002-10-30) [0009]-[0011]; Tables; claims;	1-38
Y	US 2004/087822 A1 (BUCHANAN JOHN SCOTT [US] ET AL) 6 May 2004 (2004-05-06) [0026]-[0030]; [0045]-[0055]; [0090]-[0091]; examples, e.g. Example 5; claims	1-38



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

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- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- * & * document member of the same patent family

Date of the actual completion of the international search

16 November 2007

Date of mailing of the international search report

27/11/2007

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2007/006550

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