

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
13 November 2003 (13.11.2003)

PCT

(10) International Publication Number
WO 03/093230 A2

(51) International Patent Classification⁷: **C07D**

(21) International Application Number: PCT/US03/10724

(22) International Filing Date: 8 April 2003 (08.04.2003)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
10/138,061 30 April 2002 (30.04.2002) US

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(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NI, NO, NZ, OM, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, RO, SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— without international search report and to be republished upon receipt of that report

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: PROCESS FOR AROMATICS ALKYLATION EMPLOYING ZEOLITE BETA PREPARED BY THE IN-EXTRU-
DATE METHOD

(57) Abstract: A process for the alkylation of an aromatic hydrocarbon which comprises contacting the aromatic hydrocarbon with an C₂-C₄ olefin alkylating agent under at least partial liquid phase conditions, in the presence of a catalyst comprising zeolite beta prepared by an in-extrudate method. The alkylation products comprise cumene or ethylbenzene.



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**PROCESS FOR AROMATICS ALKYLATION EMPLOYING
ZEOLITE BETA PREPARED BY THE IN-EXTRUDATE METHOD**

FIELD OF THE INVENTION

This invention relates to a process for alkylation of aromatics employing zeolite beta prepared by the in-extrudate method, and more particularly to a process for the synthesis of cumene employing this alkylation method.

BACKGROUND OF THE INVENTION

Aromatic alkylation, particularly to produce cumene and ethylbenzene, may occur using a variety of different methods. At one time, benzene was alkylated with C₂ to C₄ olefins using the Friedel-Crafts method. Suitable Friedel-Crafts catalysts include aluminum chloride, boron trifluoride, hydrofluoric acid, liquid and solid phosphoric acid, sulfuric acid, etc. These materials are highly corrosive to process equipment, create operational problems, and are often difficult to dispose of in an environmentally acceptable manner.

Non-corrosive, solid catalysts, such as zeolites, have been used for aromatic alkylation as well. Catalysts which comprise zeolite beta, ZSM-5, X or Y zeolites have all been used in cumene manufacture. There have been difficulties involving cumene selectivity, catalyst life and ease of regeneration with many of these zeolites, however.

Zeolite beta, disclosed in U.S. Pat. No. 3,308,069, is a porous crystalline synthetic material having the following composition:



wherein x is smaller than 1, y is comprised within the range of from 5 to 100, w is comprised within the range of from 0 to 4, M is a metal belonging to the Groups IA, IIA, IIIA or is a transition metal, and TEA is tetraethyl-ammonium.

1 U.S. Pat. No. 4,891,458 discloses a process for the alkylation of an aromatic
2 hydrocarbon by contacting a stoichiometric excess of the aromatic
3 hydrocarbon with a C₂ to C₄ olefin under at least partial liquid phase
4 conditions and in the presence of a catalyst comprising zeolite beta. U.S. Pat.
5 No. 5,081,323 is a continuation of U.S. Pat. 4,891,458, and discloses the
6 reaction occurring in two catalyst beds or reactors in series, with at least a
7 portion of the aromatic hydrocarbon being added between the catalyst beds or
8 reactors.

9

10 SUMMARY OF THE INVENTION

11

12 The present invention relates to an aromatic alkylation process which employs
13 a catalyst comprising a new particulate beta zeolite which is useful for acid
14 catalyzed hydrocarbon conversion reactions. The catalyst possesses
15 properties of: a SiO₂/Al₂O₃ mole ratio of greater than 15; a water adsorption
16 capacity of greater than 12 wt. %; a microporosity of greater than 0.13 cc/gm;
17 and a total acidity greater than 0.7 mmole/gm of catalyst. This catalyst is
18 prepared by the in-extrudate method, which is disclosed in U.S. Pat.
19 No. 5,558,851.

20

21 This catalyst has excellent regeneration capacity for the production of cumene
22 or ethylbenzene from the alkylation of benzene with propylene or ethylene. It
23 also possesses higher activity and longer catalyst life than zeolite beta
24 catalyst prepared by previous techniques. Furthermore, it has a high
25 selectivity for cumene.

26

27 The alkylation process is described generally below:

28

29 A process for the alkylation of an aromatic hydrocarbon to produce at least
30 one product, the process comprising contacting a stoichiometric excess of the
31 aromatic hydrocarbon with a C₂ to C₄ olefin under at least partial liquid phase
32 conditions with a catalyst comprising zeolite beta which has been prepared by
33 a process comprising the following steps:

34

- 1 (a) preparing a reaction mixture comprising at least one active source of
2 silica, optionally at least one active source of silica, optionally at least
3 one active source of alumina, an organic templating agent capable of
4 forming said crystalline zeolite, and sufficient water to shape said
5 mixture; and
6
7 (b) heating said reaction mixture at crystallization conditions and in the
8 absence of an external liquid phase for sufficient time to form a
9 crystallized material containing crystals of said zeolite, wherein said
10 zeolite crystals have a silica/alumina molar ratio greater than 15.
11

12 DETAILED DESCRIPTION OF THE INVENTION

13 Catalyst

14 Zeolite beta is a known synthetic crystalline aluminosilicate originally
15 described in U.S. Pat. Nos. 3,308,069 and Re 28,341, to which reference is
16 made for further details of this zeolite, its preparation and properties. Zeolite
17 beta is a large pore zeolite having a constraint index of less than 1. In the
18 instant invention, zeolite beta is prepared using the "in-extrudate" method,
19 which is described in U.S. Pat. No. 5,558,851, which is incorporated by
20 reference. The catalysts of the instant invention possess a higher water
21 absorptive capacity and acidity.
22
23

24
25 The catalyst comprising zeolite beta is prepared from a reaction mixture
26 having the following composition ranges:
27

28 $YO_2/W_2O_3 = >15$; preferably 15-30

29 $M+/YO_2 = 0.03-0.5$, preferably 0.05-0.3

30 $R/YO_2 = 0.07-0.30$, preferably 0.10-0.20

31 $OH-/YO_2 = 0.10-0.30$, preferably 0.12-0.25

32 $H_2O/YO_2 = 1.5-4.0$, preferably 1.7-3.2
33

1 Where Y is silicon, germanium or both; W is aluminum, boron, gallium, iron or
2 a mixture thereof; M⁺ is an alkali metal ion; and R is a templating agent.

3

4 Zeolites may be crystallized within the reaction mixture or within the shaped
5 particles made from the reaction mixture. Crystallization of the zeolite takes
6 place in the absence of an external liquid phase, i.e., in the absence of a
7 liquid phase separate from the reaction mixture. A more detailed description
8 of the method of preparation of in-extrudate zeolites, including crystallization
9 steps, is found in U.S. Pat. No. 5,558,851.

10

11 Suitable templating agents are organic cations which are derived in aqueous
12 solution from tetraethylammonium bromide or hydroxide, dibenzyl-1,4-
13 diazabicyclo[2.2.2]octane chloride, dimethyldibenzyl ammonium chloride,
14 1,4-di(1-azonium bicyclo[2.2.2]octane)butane dibromide or dihydroxide, and
15 the like. These organic cations are known in the art and are described, for
16 example, in European Patent Applications Nos. 159,846 and 159,847, and
17 U.S. Pat. No. 4,508,837. The preferred organic cation is the
18 tetraethylammonium ion. Typically, the templating agent will be an organic
19 compound which contains nitrogen or phosphorus. The sources of organic
20 nitrogen-containing cations may be primary, secondary, or tertiary amines or
21 quaternary ammonium amines. Templating agents are discussed in more
22 detail in U.S. Pat. No. 5,558,851.

23

24 M is typically a sodium ion from the original synthesis but may also be a metal
25 ion added by ion exchange techniques. Suitable metal ions include those
26 from Groups IA, IIA or IIIA of the Periodic Table or a transition metal.
27 Examples of such ions include ions of lithium, potassium, calcium,
28 magnesium, barium, lanthanum, cerium, nickel, platinum, palladium, and the
29 like.

30

31 For high catalytic activity, the zeolite beta should be predominantly in its
32 hydrogen ion form. Generally, the zeolite is converted to its hydrogen form by
33 ammonium exchange followed by calcination. If the zeolite is synthesized
34 with a high enough ratio of organonitrogen cation to sodium ion, calcination

1 alone may be sufficient. It is preferred that, after calcination, hydrogen ions
2 and/or rare earth ions occupy a major portion of the cation sites. It is
3 especially preferred that at least hydrogen ions and/or rare earth ions occupy
4 80% of the cation sites.

5
6 The zeolite, as synthesized in shaped form, may be used as a catalyst. The
7 shaped form within which the zeolite is crystallized may contain inorganic
8 oxide binders, by adding one or more of those binders to the zeolite reaction
9 mixture. It is believed that the addition of these binder materials improves
10 intra-particle diffusion. The final catalyst may contain from 70 to 100 wt. %
11 zeolite beta. Usually, the zeolite beta content will range from 80 to 100 wt. %, and more typically from 90 to 100 wt. %. The preferred inorganic binder is
12 alumina. The mixture, prior to crystallization, may be formed into tablets or
13 extrudates having the desired shape by methods well known in the art. The
14 extrudates or tablets will usually be cylindrical in shape. With cross-sectional
15 diameters ranging from 1/2 to 1/64 inch, other shapes with enhanced
16 surface-to-volume ratios, such as fluted or polylobed cylinders, can be
17 employed to enhance mass transfer rates and, thus, catalytic activity.

18
19

20 Feeds

21

22 Examples of suitable aromatic hydrocarbon feedstocks which may be
23 alkylated or transalkylated by the process of the invention include aromatic
24 compounds such as benzene, toluene and xylene. The preferred aromatic
25 hydrocarbon is benzene. Mixtures of aromatic hydrocarbons may also be
26 employed.

27

28 Suitable olefins for the alkylation of the aromatic hydrocarbon are those
29 containing 2 to 4 carbon atoms, such as ethylene, propylene, butene-1,
30 trans-butene-2 and cis-butene-2, or mixtures thereof. Preferred olefins are
31 ethylene and propylene. An especially preferred olefin is propylene. These
32 olefins may be present in admixture with the corresponding C₂ to C₄ paraffins,
33 but it is usually preferable to remove dienes, acetylenes, water, sulfur
34 compounds or nitrogen compounds which may be present in the olefin

1 feedstock stream, to prevent rapid catalyst deactivation. In some cases,
2 however, it may be desirable to add, in a controlled fashion, small amounts of
3 water or nitrogen compounds to optimize catalytic properties.

4

5 When transalkylation is desired, the transalkylating agent is a polyalkyl
6 aromatic hydrocarbon containing two or more alkyl groups that each may
7 have from 2 to about 4 carbon atoms. For example, suitable polyalkyl
8 aromatic hydrocarbons include di-, tri- and tetra-alkyl aromatic hydrocarbons,
9 such as diethylbenzene, triethylbenzene, diethylmethylbenzene
10 (diethyltoluene), diisopropylbenzene, triisopropylbenzene, diisopropyltoluene,
11 dibutylbenzene, and the like. Preferred polyalkyl aromatic hydrocarbons are
12 the dialkyl benzenes. A particularly preferred polyalkyl aromatic hydrocarbon
13 is diisopropylbenzene.

14

15 Reaction products which may be obtained from the process of the invention
16 include ethylbenzene from the reaction of benzene with either ethylene or
17 polyethylbenzenes, cumene from the reaction of benzene with propylene or
18 polyisopropylbenzenes, ethyltoluene from the reaction of toluene with
19 ethylene or polyethyltoluenes, cymenes from the reaction of toluene with
20 propylene or polyisopropyltoluenes, and sec-butylbenzene from the reaction
21 of benzene and n-butenes or polybutylbenzenes. The production of cumene
22 from the alkylation of benzene with propylene or the transalkylation of
23 benzene with di-isopropylbenzene is especially preferred.

24

25 Conditions

26

27 When alkylation is the process conducted according to this invention, reaction
28 conditions are as follows. The aromatic hydrocarbon feed should be present
29 in stoichiometric excess. It is preferred that the molar ratio of aromatics to
30 olefins be at least about four to one (4:1) to prevent rapid catalyst fouling.
31 The reaction temperature may range from 100°F to 600°F, preferably 250°F to
32 450°F. In the case of cumene production, a temperature range of 250°F to
33 375°F is most preferred to reduce product impurities. The reaction pressure

1 should be sufficient to maintain at least a partial liquid phase in order to retard
2 catalyst fouling. This is typically 50 to 1000 psig depending on the feedstock
3 and reaction temperature. Contact time may range from 10 seconds to
4 10 hours, but is usually from 5 minutes to an hour. The weight hourly space
5 velocity (WHSV), in terms of grams (pounds) of aromatic hydrocarbon and
6 olefin per gram (pound) of catalyst per hour, is generally within the range of
7 about 0.5 to 50.

8
9 When transalkylation is the process conducted according to the invention, the
10 molar ratio of aromatic hydrocarbon to polyalkyl aromatic hydrocarbon will
11 generally range from about 1:1 to about 50:1, and preferably from about 2:1 to
12 about 20:1. The reaction temperature may range from about 100°F to 600°F,
13 but it is preferably about 250°F to 450°F. The reaction pressure should be
14 sufficient to maintain at least a partial liquid phase, typically in the range of
15 about 50 psig to 1000 psig, preferably 300 psig to 600 psig. The weight hour
16 space velocity will range from about 0.1 to 10.

17
18 Various types of reactors can be used in the process of this invention. For
19 example, the process can be carried out in batchwise fashion by adding the
20 catalyst and aromatic feedstock to a stirred autoclave, heating to reaction
21 temperature, and then slowly adding the olefinic or polyalkylaromatic
22 feedstock. A heat transfer fluid can be circulated through the jacket of the
23 autoclave, or a condenser can be provided, to remove the heat of reaction
24 and maintain a constant temperature. Large scale industrial processes may
25 employ a fixed bed reactor operating in an upflow or downflow mode or a
26 moving bed reactor operating with concurrent or countercurrent catalyst and
27 hydrocarbon flows. These reactors may contain a single catalyst bed or
28 multiple beds and may be equipped for the interstage addition of olefins and
29 interstage cooling. Interstage olefin addition and more nearly isothermal
30 operation enhance product quality and catalyst life. A moving bed reactor
31 makes possible the continuous removal of spent catalyst for regeneration and
32 replacement by fresh or regenerated catalysts.

33

1 In a preferred embodiment of the present invention, the alkylation process is
2 carried out with addition of olefin in at least two stages. Preferably, there will
3 be two or more catalyst beds or reactors in series, wherein at least a portion
4 of the olefin is added between the catalyst beds or reactors. Interstage
5 cooling can be accomplished by the use of a cooling coil or heat exchanger.
6 Alternatively, interstage cooling can be affected by staged addition of the
7 aromatic feedstock, that is, by addition of the aromatic feedstock in at least
8 two stages. In this instance, at least a portion of the aromatic feedstock is
9 added between the catalyst beds or reactors, in similar fashion to the staged
10 addition of olefin described above. The staged addition of aromatic feedstock
11 provides additional cooling to compensate for the heat of reaction.

12

13 In a fixed bed reactor or moving bed reactor, alkylation is completed in a
14 relatively short reaction zone following the introduction of olefin. Ten to thirty
15 percent of the reacting aromatic molecules may be alkylated more than once.

16

17 The alkylation reactor effluent contains the excess aromatic feed,
18 monoalkylated product, polyalkylated products, and various impurities. The
19 aromatic feed is recovered by distillation and recycled to the alkylation
20 reactor. Usually, a small bleed is taken from the recycle stream to eliminate
21 unreactive impurities from the loop. The bottoms from the benzene distillation
22 are further distilled to separate monoalkylated product from polyalkylated
23 products and other heavies. In most cases, the recovered monoalkylated
24 product must be very pure. For example, current specifications call for 99.9%
25 cumene purity with less than 500 ppm each of ethylbenzene and
26 butylbenzene. Since only a small fraction of by-product ethylbenzene and
27 n-propylbenzene can be economically removed by distillation, it is important to
28 have a feedstock containing very little ethylene and a catalyst which makes
29 very small amounts of these impurities.

30

31 The following examples are provided to illustrate the invention in accordance
32 with the principles of the invention but are not to be construed as limiting the
33 invention in any way except as indicated by the appended claims.

34

EXAMPLESExample 1

To 120 grams of silica (Hi-Sil 233, a hydrated silica manufactured by PPG) were added 18 grams of NaAlO_2 in a Baker-Perkins mixer and mixed for 10 minutes. To this was added 150 grams of a 40 wt. % aqueous solution of tetraethylammonium hydroxide (TEAOH) and 8 grams of a 50 wt. % aqueous solution of NaOH. This mixture was mixed for three hours. Then 25 grams of water was added with mixing to bring the mixture to a paste. Another 30 grams of silica was added along with 7 grams of kaolin clay. Steam heat was applied to the jacket around the walls of the mixer to bring the mixture to an extrudable consistency at a volatiles level of 44% (by LOI). Molar ratios in the synthesis mix were as follows:

$$\text{TEAOH}/\text{SiO}_2 = 0.16$$

$$\text{OH}/\text{SiO}_2 = 0.20$$

$$\text{Na}/\text{SiO}_2 = 0.14$$

$$\text{SiO}_2/\text{Al}_2\text{O}_3 = 17$$

$$\text{H}_2\text{O}/\text{SiO}_2 = 1.9$$

The mix was then extruded through a 1/16-inch die. The extrudate was placed in a Teflon bottle in a stainless steel pressure vessel and heated to 150°C at autogenous pressure for four days. The extrudate was washed with a 10% aqueous solution of ammonium nitrate containing 6 cc HNO_3 per 1000 grams of solution, dried overnight in a vacuum oven at 110°C, and calcined in air at 538°C for six hours. X-ray diffraction analysis showed the extrudate to be 96% beta. The micropore volume (by N_2 adsorption) was 0.20 cc/g and the surface area (by BET) was 568 m^2/g .

Example 2

The catalyst of Example 1 was tested in a down-flow micro-unit for cumene production. The catalyst was crushed to 24-42 mesh and calcined in air at 650°F before installing the reactor in the micro-unit. The reactor was then pressured up to 600 psig using nitrogen and then lined out at a temperature of 300°F prior to introducing feed. The feed was an 8/1 volumetric mixture of benzene/propylene, run at a WHSV of 5.7. A 4A mole sieve drier was installed in the feed line to remove any residual water. Samples were taken every three hours using an on-line gas chromatograph. Results at 40 hours on-stream are shown in Table I, along with results at the same conditions for a commercial prototype Beta catalyst made using conventional synthesis and containing 80% Beta zeolite bound with alumina. These results show the catalyst of Example 1 to give close to 95% selectivity to cumene at 100% propylene conversion.

Table I**Cumene Synthesis Test****8/1 Benzene/Propylene, 5.7 WHSV, 300°F, 600 psig (40 Hr On-stream)**

Catalyst	Commercial	Example 1
SiO ₂ /Al ₂ O ₃	25	17
Propylene Conv, %	100	100
Cumene, wt. %	91.0	94.9
1,3 DIPB	4.9	2.3
1,4 DIPB	3.7	2.1
Other	0.4	0.7

Example 3

To 1800 grams of silica (Hi-Sil 233, a hydrated silica manufactured by PPG) were added 78 grams of NaAlO₂ in a Baker-Perkins mixer. To this was added 1344 grams of a 35 wt. % aqueous solution of tetraethylammonium hydroxide (TEAOH), 144 grams of a 50 wt. % aqueous solution of NaOH, 114 grams

1 alumina (Versal 250, manufactured by UOP, 74% Al_2O_3), 84 grams of kaolin
2 clay, 145 grams of Beta zeolite powder as seed, then 1035 grams of water.
3 This mixture was mixed for 20 minutes. Steam heat (160°F) was applied to
4 the jacket around the walls of the mixer to bring the mixture to an extrudable
5 consistency at a volatiles level of 53% (by LOI). The mix was then extruded
6 through a 1/16-inch die, and air dried on trays to 43% LOI. Molar ratios in the
7 synthesis mix were as follows:

8
9 $\text{TEAOH}/\text{SiO}_2 = 0.12$
10 $\text{OH}/\text{SiO}_2 = 0.18$
11 $\text{Na}/\text{SiO}_2 = 0.10$
12 $\text{SiO}_2/\text{Al}_2\text{O}_3 = 17$
13 $\text{H}_2\text{O}/\text{SiO}_2 = 2.3$

14
15 The extrudate was placed in a jacketed rotating stainless steel pressure
16 vessel and heated for 36 hours at autogenous pressure to 150°C by
17 circulating hot oil through the jacket. The extrudate was washed with a 10%
18 aqueous solution of ammonium nitrate containing 6 cc HNO_3 per 1000 grams
19 of solution, dried overnight in a vacuum oven at 110°C, and calcined in air at
20 538°C for six hours. X-ray diffraction analysis showed the extrudate to be
21 97% beta.

22

23 Example 4

24

25 The catalyst of Example 3 was tested for cumene production using the same
26 method as given in Example 2, except the WHSV was 41.6 to accelerate
27 aging in order to determine catalyst cycle life. The run was considered over
28 when the propylene conversion dropped below 95%. At 40 hours on-stream,
29 cumene selectivity was 92%, and was 94% at 122 hours. The cycle life with
30 this catalyst was 131 hours.

31

1 Comparative Example

2

3 The run of Example 4 was repeated, this time using a commercial Beta zeolite
4 extrudate containing 80 wt. % Beta zeolite and 20% wt. % alumina binder.

5 The cycle life with this catalyst was 43 hours.

6

7 Example 5

8

9 The catalyst of Example 4 was mildly crushed and pre-hydrated for seven
10 days, then analyzed by NMR and found to have a water-free tetrahedral Al
11 content of 3.1 wt. %, corresponding to a framework $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio of
12 27. This catalyst was then compared against a commercial Beta zeolite of the
13 same framework $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio for water adsorption using the
14 H Corr-Purcell-Meiboom-Gill (CPMG) method. This method is described in
15 T.C Farrar and E.D. Becker, "Pulse and Fourier Transform NMR," 1971
16 Edition, Academic Press, New York. Results are shown in Table II.

17

<u>Table II</u>		
Percent Water Adsorption by H CPMG Method		
	H ₂ O, Wt. %	H ₂ O, Wt. % (repeat analysis)
Example 3 catalyst	19.6	20.1
Commercial Beta zeolite	15.2	17.0

18

19

20 These results show the catalyst of Example 3 has greater capacity for water.
21 The commercial beta zeolite in this example was not bound with alumina.
22 Had the catalyst of Example 3 been compared against a conventional Beta
23 catalyst, in which the zeolite is bound with alumina, the difference in H₂O
24 capacity would have been expected to be even greater.

25

Example 6

The acidity measurement by temperature programmed desorption of isopropylamine is applicable to determining the amount of Bronsted acid sites due to their strong interaction.

The chemically adsorbed isopropylamine molecules decompose to propylene at temperatures above 280°C. The amount of acid sites on a zeolite can be determined by using thermogravimetric technique (TGA) to measure the amount of propylene decomposed. In a thermogravimetric experiment, the sample is activated at a temperature of 500°C. Isopropylamine is adsorbed onto the zeolite. The sample is allowed to release any physisorbed isopropylamine, then heated to release the remaining chemisorbed isopropylamine.

Since each IPA molecule stoichiometrically interacts with one proton on the zeolite, the acid density or acidity can be calculated based on the amount of isopropylamine desorbed between 280°C and 500°C. The following procedure is carried out:

1. Load 25-35 milligrams of sample onto (tarred) platinum pan.
2. Insert balance arm into furnace portion of TGA.
3. Run TGA program (under constant He purge):
 - Ramp to 500°C (@10°C/min.) and hold for 15 minutes to activate sample.
 - Cool to 100°C (freefall) to allow adsorption of IPAm(isopropylamine).
 - Introduce IPAm vapor until weight gain reaches a plateau, or for 15 minutes (adsorption of IPAm).
 - Ramp to 280°C (@10°C/min.), hold for 30 minutes (to allow for desorption of physisorbed IPAm).
 - Ramp to 500°C to allow for desorption of chemisorbed IPAm.

1 The residual weight of the sample is measured at 100°C, before introduction
2 of IPAm (anhydrous weight) and at point right before ramp to 500°C (after
3 desorption of physisorbed IPAm).

4

5 The total acid site density is obtained in mmol/g as follows:

6

7 Acid site density (mmol/g) =

8

9
$$\frac{\text{Wt. of zeolite with chemisorbed IPAm} - \text{Anhydrous wt. of Zeolite}}{\text{Anhydrous wt. of Zeolite} \times \text{MW (IPAm)}}$$

10

11

12 The acidity of the catalyst of Example 3, as determined by the isopropylamine
13 TPD test, was 0.97 meq/g. The same analysis done on a commercial Beta
14 catalyst similar to that of Example 5 showed an acidity of 0.5 mmole/g.

1 WHAT IS CLAIMED IS:

2

3 1. A process for the alkylation of an aromatic hydrocarbon to produce at
4 least one product, the process comprising contacting a stoichiometric
5 excess of the aromatic hydrocarbon with a C₂ to C₄ olefin under at least
6 partial liquid phase conditions with a catalyst comprising zeolite beta
7 which has been prepared by a process comprising the following steps:

8

- 9 (a) preparing a reaction mixture comprising at least one active source
10 of a first oxide selected from the group consisting of silica and
11 germanium or mixtures thereof, optionally at least one active
12 source of a second oxide selected from the group consisting of
13 alumina, boron oxide, gallium oxide, and iron oxide or mixtures
14 thereof, an organic templating agent capable of forming said
15 crystalline zeolite, and sufficient water to shape said mixture; and
16
17 (b) heating said reaction mixture at crystallization conditions and in the
18 absence of an external liquid phase for sufficient time to form a
19 crystallized material containing crystals of said zeolite, wherein
20 said zeolite crystals have a first oxide/second oxide molar ratio
21 greater than 30.

22

- 23 2. The process of claim 1, in which the reaction mixture of step (a)
24 comprises a water to silica mole ratio of from 2 to 5.

25

- 26 3. The process of claim 1, wherein said reaction mixture possesses the
27 following composition ranges:

28

29 $YO_2/W_2O_3 = >15$

30 $M+/YO_2 = 0.03-0.5$

31 $R/YO_2 = 0.07-0.30$

32 $OH-/YO_2 = 0.10-0.30$

33 $H_2/YO_2 = 1.5-4$

34

1 Where Y is selected from silicon, germanium or a mixture of both; W is
2 selected from aluminum, boron, gallium, iron or a mixture thereof; M+ is
3 an alkali metal ion, and R is a templating agent.

4

5 4. The process of claim 3, wherein the catalyst comprising zeolite beta has
6 the following composition ranges:

7

8 $\text{SiO}_2/\text{Al}_2\text{O}_3 = >15$

9 $\text{Na}^+/\text{SiO}_2 = 0.03\text{-}0.5$

10 $\text{R}/\text{SiO}_2 = 0.07\text{-}0.30$

11 $\text{OH}/\text{SiO}_2 = 0.10\text{-}0.30$

12 $\text{H}_2\text{O}/\text{SiO}_2 = 1.5\text{-}4$

13

14 where R is a templating agent.

15

16 5. The process of claim 3, wherein the molar ratio of templating agent to
17 silica is in the range from 0.1 to 0.2.

18

19 6. The process of claim 3, wherein the silica/alumina ratio is in the range
20 from 15 to 30.

21

22 7. The process of claim 1, in which the reaction mixture of step (a) is
23 formed into a desired shape prior to the crystallization step.

24

25 8. The process of claim 1, in which selectivity for cumene is in the range
26 from 93-97%.

27

28 9. The process of claim 1, wherein the molar ratio of aromatic hydrocarbon
29 to olefin is at least 2:1.

30

31 10. The process of claim 3, wherein the templating agent is selected from
32 primary, secondary, tertiary amines or quaternary ammonium
33 compounds.

34

- 1 12. The process of claim 3, where the templating agent is
2 tetraethylammonium hydroxide (TEAOH).
3
- 4 13. The process of claim 1, wherein the molar ratio of aromatic hydrocarbon
5 to olefin is at least 4:1.
6
- 7 14. A process for the alkylation of an aromatic hydrocarbon to produce at
8 least one product, the process comprising contacting a stoichiometric
9 excess of the aromatic hydrocarbon with a C₂ to C₄ olefin under at least
10 partial liquid phase conditions with a catalyst comprising zeolite beta,
11 said catalyst having a water adsorption capacity of greater than 18 wt. %
12 and an acidity greater than 0.8 mmole/g.
13
- 14 15. The process of claim 14, wherein the zeolite beta of which the catalyst is
15 comprised is synthesized from a reaction mixture comprising a water to
16 silica molar ratio of from 1.5 to 4.0.
17
- 18 16. The process of claim 1, wherein the aromatic hydrocarbon is selected
19 from the group consisting of benzene, toluene, and xylene, or mixtures
20 thereof.
21
- 22 17. The process of claim 14, wherein the aromatic hydrocarbon is benzene.
23
- 24 18. The process of claim 1, wherein the olefin is a member selected from the
25 group consisting of ethylene, propylene, butene-1, trans-butene-2, and
26 cis-butene-2 or mixtures thereof.
27
- 28 19. The process of claim 1, wherein alkylation is carried out at a temperature
29 in the range of about 250°F to 450°F, a pressure in the range of about
30 300 psig to 600 psig, and a weight hourly space velocity of about 0.1 to
31 10.
32

1 20. The process of claim 1, wherein the product of the alkylation is cumene
2 or ethylbenzene.
3

4 21. The process of claim 1, wherein the catalyst comprising zeolite beta is
5 shaped into a sphere or cylinder and has a cross-sectional diameter
6 between 1/64 inch and 1/2 inch.
7

8 22. A process for the transalkylation of an aromatic hydrocarbon, wherein a
9 stoichiometric excess of an aromatic hydrocarbon is contacted with a
10 polyalkyl aromatic hydrocarbon, under at least partial liquid phase
11 conditions with a catalyst comprising zeolite beta which has been
12 prepared by a process comprising the following steps:
13

- 14 (a) preparing a reaction mixture comprising at least one active source
15 of first oxide selected from the group consisting of silica and
16 germanium or mixtures thereof, optionally at least one active
17 source of a second oxide selected from the group consisting of
18 alumina, boron oxide, gallium oxide, and iron oxide or mixtures
19 thereof, an organic templating agent capable of forming said
20 crystalline zeolite, and sufficient water to shape said mixture; and
21
22 (b) heating said reaction mixture at crystallization conditions and in the
23 absence of an external liquid phase for sufficient time to form a
24 crystallized material containing crystals of said zeolite, wherein
25 said zeolite crystals have a first oxide/second oxide molar ratio
26 greater than 30.