Spruson & Ferguson

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CONVENTION APPLICATION FOR STANDARD PATENT OR A STANDARD PATENT OF ADDITION

41811/85

Full name(s) of Applicant(s)

I/We SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

Address(es) of Applicant(s)

of Carel van Bylandtlaan 30, 2596 HR, The Hague the Netherlands

hereby apply for the grant of a standard patent ~~patent of addition~~

for an invention entitled

Title of Invention

"PROCESS FOR THE PREPARATION OF AN AROMATIC HYDROCARBON MIXTURE"

which is described in the accompanying complete specification.

DETAILS OF BASIC APPLICATION(~~S~~)

Number(~~s~~) of Basic Application(~~s~~)

8401384

Name(~~s~~) of Convention Country(~~ies~~) in which Basic Application(~~s~~) was/~~were~~ filed

the Netherlands

(~~respectively~~)

Date(~~s~~) of Basic Application(~~s~~)

2 May, 1984

(~~respectively~~)

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AUTH. OFFICER

LODGED AT SUB-OFFICE 30 APR 1985 Sydney

APPLICATION ACCEPTED AND AMENDMENTS ALLOWED 22-1-90

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Dated this TWENTY-SIXTH day of APRIL 1985.

SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

By: .....

Registered Patent Attorney

To: The Commissioner of Patents

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12/83

COMMONWEALTH OF AUSTRALIA

Patents Act 1952-55

DECLARATION IN SUPPORT OF A CONVENTION APPLICATION FOR A PATENT

41811/85

In support of the Convention Application made by
SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.
for a patent for an invention entitled:

"Process for the preparation of an aromatic hydrocarbon mixture"

I, Onno Aalbers, of Carel van Bylandtlaan 30, The Hague, the Netherlands, do solemnly and sincerely declare as follows:

1. I am authorized by SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V., the applicant for the patent, to make this declaration on its behalf.

2. The basic application(s) as defined by Section 141 of the Act was/~~were~~ made in the Netherlands
on the 2nd day of May, 1984
by SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

3. Eduard Philip KIEFFER, a Netherlands national of Badhuisweg 3, 1031 CM Amsterdam, the Netherlands

is/~~are~~ the actual inventor(s) of the invention and the facts upon which the Applicant Company is entitled to make application are as follows: as Assignees of the inventor(s).

4. The basic application(s) referred to in paragraph 2 of this Declaration was/~~were~~ the first application(s) made in a Convention Country in respect of the invention the subject of the application.

Declared at
The Hague

Dated this 2nd day of April 1985.

Signature of Declarant

To.: The Commissioner of Patents,
Commonwealth of Australia

JB2H04

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(54) Title
CATALYTIC PREPARATION OF AROMATICS FROM OLEFINS

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(56) Prior Art Documents
AU 76264/81 542145 C07C 2/42
EP 0021475

(57) The invention relates to a process for the preparation of an aromatic hydrocarbon mixture from mono-olefins with two to four carbon atoms per molecule or from aliphatic hydrocarbon mixtures, at least 50 wt.% of which consists of the said olefins.

CLAIM

1. Process for the preparation of an aromatic hydrocarbon mixture, characterized in that a feed of one or more olefins with two to four carbon atoms per molecular or aliphatic hydrocarbons, at least 50 %wt of which consisting of the said olefins, are contacted at a temperature of between 325 and 700°C with a catalyst containing a crystalline gallium silicate, which silicate has the following distinguishing features after calcination for one hour in air at 500°C:

(a) an X-ray powder diffraction pattern that contains as strongest lines the four lines listed in Table A

TABLE A

d(Å)
11.1 ± 0.1
10.0 ± 0.1
3.84 ± 0.07
3.72 ± 0.06

12

and

(b) in the formula which gives the composition of the silicate expressed in moles of the oxides, the SiO_2/Ga_2O_3 mol. ratio is between 25 and 250,

and that, if the silicate has a SiO_2/Ga_2O_3 mol. ratio of 100-250, the catalyst is subjected one or more times to a two-stage treatment comprising a first stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with a hydrogen-containing reducing gas, followed by a second stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with an oxygen-containing oxidizing gas.

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Form 10

COMPLETE SPECIFICATION

(ORIGINAL)

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Related Art :

Name of Applicant(s): SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

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Complete Specification for the invention entitled :

"PROCESS FOR THE PREPARATION OF AN AROMATIC HYDROCARBON MIXTURE"

The following statement is a full description of this invention, including the best method of performing it known to ~~me~~/us :

K 5729

PROCESS FOR THE PREPARATION OF AN AROMATIC HYDROCARBON MIXTURE

The invention relates to a process for the preparation of an aromatic hydrocarbon mixture from mono-olefins with two to four carbon atoms per molecule or from aliphatic hydrocarbon mixtures, at least 50 wt.% of which consists of the said olefins.

Mono-olefins with two to four carbon atoms per molecule can be converted at a temperature of 325-700°C into aromatic hydrocarbon mixtures by contacting the olefins with a catalyst containing a crystalline aluminium silicate with a special structure. Such crystalline aluminium silicates are characterized by the fact that after calcination in air for one hour at 500 °C they possess the following distinguishing features:

a) an X-ray powder diffraction pattern containing as strongest lines the four lines listed in Table A,

TABLE A

d(Å)
11.1 ± 0.1
10.0 ± 0.1
3.84 ± 0.07
3.72 ± 0.06

and,

b) in the formula giving the composition of the silicate expressed in moles of the oxides, the SiO_2/Al_2O_3 mol. ratio is 25-400.

The above-mentioned process results in a product in which, besides the desired C_5^+ hydrocarbons, C_4^- hydrocarbons formed as byproducts also occur. The C_5^+ fraction present in the product

is more valuable according as it has a higher aromatic content.

Experimental research into the above-mentioned process has shown that the aromatic content of the C_5^+ fraction obtained depends to a large extent on the chosen reaction temperature and SiO_2/Al_2O_3 mol. ratio of the crystalline silicate. According as a higher reaction temperature and/or a crystalline silicate with a lower SiO_2/Al_2O_3 mol. ratio is used in the process, a product is obtained whose C_5^+ fraction has a higher aromatic content. For both these measures, however, the preparation of a product whose C_5^+ fraction has a higher aromatic content is accompanied by a reduction in the yield of the C_5^+ fraction. The research showed that a particular relationship exists between the increase in the aromatic content of the C_5^+ fraction and the reduction in the yield of the C_5^+ fraction, so that, in principle, for every aromatic content of the C_5^+ fraction being prepared, there is a particular potential yield of that C_5^+ fraction. This relationship is unfavourable to the extent that only low yields of C_5^+ fractions with a relatively high aromatic content can be obtained. Since a possible application of the process on a commercial scale depends not only on the aromatic content of the C_5^+ fraction, but also on the potential yield of that C_5^+ fraction, there is a practical limit to the maximum aromatic content of the C_5^+ fraction being prepared, due to the fact that the corresponding yield of the C_5^+ fraction must still be acceptable.

Further research into this subject has now shown that considerably better results can be obtained in the above-mentioned process if, as crystalline silicate with the previously mentioned special structure, a silicate containing gallium instead of aluminium is used, and if, moreover, the following two requirements are met:

a) in the formula giving the composition of the gallium silicate expressed in moles of the oxides, the SiO_2/Ga_2O_3 mol. ratio should be 25-250, and

b) if the gallium silicate has a SiO_2/Ga_2O_3 mol. ratio of 100-250, the catalyst should be subjected one or more times to a two-stage treatment comprising a first stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700 °C with a hydrogen-containing reducing gas, followed by a second stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700 °C with an oxygen-containing oxidizing gas.

The research showed that in the case of the present crystalline gallium silicates corresponding with the previously mentioned crystalline aluminium silicates, the higher the reaction temperature and/or the lower the SiO_2/Ga_2O_3 mol. ratio of the silicate employed, the higher the aromatic content of the C_5^+ fraction of the product obtained, and also that for both measures the preparation of a product whose C_5^+ fraction has a higher aromatic content is accompanied by a reduction in the yield of the C_5^+ fraction. For the crystalline gallium silicates there is also a relationship between the increase in the aromatic content of the C_5^+ fraction and the reduction in the yield of the C_5^+ fraction, whereby, in principle, for every aromatic content of the C_5^+ fraction being prepared there is a particular potential yield of that C_5^+ fraction. The important difference between the crystalline aluminium silicates and the present crystalline gallium silicates is that the previously mentioned relationship for the gallium silicates is considerably more favourable, so that considerably higher yields of C_5^+ fractions with a relatively high aromatic content can be obtained than is the case for the aluminium silicates. Comparison of the results obtained from a systematic investigation into processes for preparing aromatic hydrocarbons from a given feed, but employing a variety of aluminium silicates and gallium silicates at various temperatures showed that if the processes were aimed at the preparation of products whose C_5^+ fraction had a particular

aromatic content, the gallium silicates gave a higher yield of the C_5^+ fraction, whereas if the processes were aimed at the preparation of products with a given content of the C_5^+ fraction, the gallium silicates gave a C_5^+ fraction with a higher aromatic content.

The present patent application therefore relates to a process for the preparation of an aromatic hydrocarbon mixture, characterized in that a feed of one or more olefins with two to four carbon atoms per molecular or aliphatic hydrocarbons, at least 50 %wt of which consisting of the said olefins, are contacted at a temperature of between 325 and 700°C with a catalyst containing a crystalline gallium silicate, which silicate has the following distinguishing features after calcination for one hour in air at 500°C:

(a) an X-ray powder diffraction pattern that contains as strongest lines the four lines listed in Table A

TABLE A

d(Å)
11.1 ± 0.1
10.0 ± 0.1
3.84 ± 0.07
3.72 ± 0.06

and

(b) in the formula which gives the composition of the silicate expressed in moles of the oxides, the SiO_2/Ga_2O_3 mol. ratio is between 25 and 250,

and that, if the silicate has a SiO_2/Ga_2O_3 mol. ratio of 100-250, the catalyst is subjected one or more times to a two-stage treatment comprising a first stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with a hydrogen-containing reducing gas, followed by a second stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with an oxygen-containing oxidizing gas.

The starting material for the process according to the invention should be one or more mono-olefins having two to four carbon atoms per molecule or aliphatic hydrocarbons, at least 50 %wt of which consists of the said olefins. The mono-olefins with two to four carbon atoms per molecule, which should form at least 50 %wt of the feed, are ethylene,

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propylene, butylene-1, butylene-2 and iso-butylene. If a hydrocarbon is employed that in addition to the said C_4^- mono-olefins also contains other aliphatic hydrocarbons, these other hydrocarbons may be paraffins, di-olefins or C_5^+ mono-olefins. In the process according to the invention a feed is preferably employed which consists of at least 75 %wt and in particular substantially completely of

one or more mono-olefins with three or four carbon atoms per molecule. A very suitable feed for the present process is a hydrocarbon mixture consisting substantially of C_3 and/or C_4 mono-olefins, this mixture being obtained as byproduct of the catalytic or thermal cracking of hydrocarbons, in particular the steam cracking of hydrocarbons for the preparation of ethylene.

The process according to the invention is preferably carried out at a temperature of 350-575 °C, a pressure of 1-20 bar and particularly 1-10 bar, and a space velocity of 0.1-10 $kg.kg^{-1}.hour^{-1}$ and in particular of 0.5-5 $kg.kg^{-1}.hour^{-1}$.

In the process according to the invention the feed is contacted with a catalyst containing a crystalline gallium silicate defined amongst other things by the X-ray powder diffraction pattern of the silicate after calcination for one hour in air at 500 °C. This should contain as strongest lines the four lines given in Table A. The complete X-ray powder diffraction pattern of a typical example of the present crystalline gallium silicates after calcination for one hour in air at 500 °C is represented in Table B.

TABLE B

d(Å)	Rel. int.	d(Å)	Rel. int.
11.1	100	3.84 (D)	57
10.0 (D)	70	3.72 (D)	31
8.93	1	3.63	16
7.99	1	3.47	< 1
7.42	2	3.43	5
6.68	7	3.34	2
6.35	11	3.30	5
5.97	17	3.25	1
5.70	7	3.05	8
5.56	10	2.98	11
5.35	2	2.96	3
4.98 (D)	6	2.86	2
4.60	4	2.73	2
4.35	5	2.60	2
4.25	7	2.48	3
4.07	2	2.40	2
4.00	4		

(D) = doublet

Suitable catalysts for use in the process according to the invention are those containing a crystalline gallium silicate with a SiO_2/Ga_2O_3 mol. ratio of 25-250. If the crystalline gallium silicate has a SiO_2/Ga_2O_3 mol. ratio of 100-250, it should be subjected one or more times to the above-described two-stage treatment. Although crystalline gallium silicates with a SiO_2/Ga_2O_3 mol. ratio of less than 100 are suitable as such for use in the process according to the invention, they are also preferably subjected one or more times to the above-described two-stage treatment. In the process according to the invention,

preferred crystalline gallium silicates have a SiO_2/Ga_2O_3 mol. ratio of 60-220. When employing a crystalline gallium silicate with a maximum SiO_2/Ga_2O_3 mol. ratio of 110, the two-stage treatment is preferably not performed more than three times. When employing a crystalline gallium silicate with a SiO_2/Ga_2O_3 mol. ratio of more than 110, but not more than 130, the two-stage treatment is preferably performed at least three times, but not more than ten times. When employing a crystalline gallium silicate with a SiO_2/Ga_2O_3 mol. ratio of more than 130, but not more than 220, the silicate is preferably first subjected to calcination at a temperature of 600-1000 °C, after which the two-stage treatment is performed on it at least three times, but not more than ten times.

During the first stage of the two-stage treatment the catalyst should be contacted for at least 15 minutes at a temperature of 350-700 °C with a hydrogen-containing reducing gas. The first stage of the two-stage treatment can in principle be performed in two ways. In the first place the hydrogen-containing reducing gas can be fed to the catalyst from outside. In that case a gas is preferably employed that contains at least 20 vol.% and in particular at least 40 vol.% hydrogen. The gas employed can very suitably contain, besides hydrogen, either mainly nitrogen or mainly carbon monoxide or mainly C_4^- hydrocarbons. Suitable gases which, besides hydrogen, contain mainly carbon monoxide can be obtained as synthesis gas from a highly carbonaceous material, such as coal, by gasification, or from light hydrocarbons, such as natural gas, by steam reforming or partial oxidation. Suitable gases which, besides hydrogen, contain mainly C_4^- hydrocarbons can be obtained as byproduct from the catalytic conversion of hydrocarbons in the presence of hydrogen, such as cracking, isomerization and reforming. In the second place the hydrogen-containing reducing gas can be produced in situ by contacting the catalyst for a maximum of five hours with a hydrocarbon or a hydrocarbon mixture. A very

suitable hydrocarbon or hydrocarbon mixture would in this case be the feed employed in the present process.

In the second stage of the two-stage treatment the catalyst should be contacted for at least 15 minutes at a temperature of 350-700 °C with an oxygen-containing oxidizing gas. The gas employed preferably contains at least 5 vol.% and in particular at least 10 vol.% oxygen. The second stage can very suitably be performed using a gas which, besides oxygen, contains either mainly nitrogen or otherwise mainly nitrogen, carbon monoxide and carbon dioxide. A suitable gas that, besides oxygen, contains mainly nitrogen is air. Suitable gases which, besides oxygen contain mainly nitrogen, carbon monoxide and carbon dioxide are offgases obtained during the removal of carbon by means of an excess of air from deactivated hydrocarbon conversion catalysts. Preferably, the two stages of the two-stage treatment are performed at a temperature of 400-650 °C and in particular at a temperature of 475-575 °C. It is also preferable that the two stages be performed at the same temperature.

The crystalline gallium silicates employed in the process according to the invention can very suitably be prepared from an aqueous mixture containing the following compounds: one or more compounds of an alkali metal (M), one or more organic nitrogen compounds (RN) containing an organic cation or from which an organic cation is formed during the preparation of the silicate, one or more silicon compounds and one or more gallium compounds. Preparation is effected by maintaining the mixture at an elevated temperature until the silicate has been formed and then separating the silicate crystals from the mother liquor and washing, drying and calcining the crystals. The aqueous mixture from which the silicates are prepared should contain the various compounds in the following molar ratios, except for the organic nitrogen compounds, expressed in moles of the oxides:

M_2O : SiO_2 = 0.01-0.35,
RN : SiO_2 = 0.02-1.0,
SiO_2 : Ga_2O_3 = 25-400, and
H_2O : SiO_2 = 5-65.

The silicates can very suitably be prepared from a basis mixture containing a quaternary ammonium compound as organic nitrogen compound, a sodium compound as alkali metal compound and amorphous silica as silicon compound.

The silicates prepared according to the above method contain alkali metal ions. By means of suitable exchange techniques these can be replaced by other cations such as hydrogen ions or ammonium ions. The crystalline gallium silicates employed in the process according to the invention preferably have an alkali metal content of less than 0.05 %wt. In the process according to the invention the crystalline gallium silicates can be used as such or in combination with a binding material, such as kaolin or bentonite.

The invention will now be illustrated with the aid of the following example.

EXAMPLE

Two crystalline aluminium silicates (silicates 1 and 2) and three crystalline gallium silicates (silicates 3-5) were prepared by heating mixtures of NaOH, amorphous silica, $(C_3H_7)_4NOH$ and either $Al(NO_3)_3$, or $Ga(NO_3)_3$ in water in an autoclave under autogenous pressure for 24 hours at 150 °C. After the reactants had cooled down, the resulting silicates were filtered off, washed with water until the pH of the wash water was about 8, and dried at 120 °C. After calcination for one hour in air at 500 °C, silicates 1-5 had the following properties:

a) an X-ray powder diffraction pattern corresponding substantially with that given in Table B, and
b) a value for the SiO_2/Al_2O_3 or SiO_2/Ga_2O_3 mol. ratio as given in Table C.

TABLE C

Silicate No.	SiO_2/Al_2O_3 mol. ratio	SiO_2/Ga_2O_3 mol. ratio
1	77	-
2	250	-
3	-	70
4	-	130
5	-	350

From silicates 1-5 were prepared silicates I-V by boiling silicates 1-5 with 1.0 molar NH_4NO_3 solution, washing with water, again boiling with 1.0 molar NH_4NO_3 solution, and washing, drying at 120 °C and calcining at 500 °C.

Samples of silicates III-V were repeatedly subjected to a two-stage treatment comprising a first stage in which the silicate was contacted for 30 minutes with n-butane at a temperature of 550 °C, a pressure of 1.5 bar and a space velocity of 8 $g.g^{-1}.hour^{-1}$, followed by a second stage in which the silicate was contacted for one hour with air at a temperature of 550 °C and a pressure of 1.5 bar. By performing this two-stage treatment three times on silicate III and ten times on silicates IV and V, silicates IIIA-VA were obtained from silicates III-V respectively.

Silicates I, II, IIIA, IV, IVA and VA were tested in twelve experiments (experiments 1-12) for the preparation of C_5^+ fractions from iso-butylene (experiments 1-11) and from propylene (experiment 12). All experiments were performed at a pressure of 1.5 bar and a space velocity of 2 $kg.kg^{-1}.hour^{-1}$ in a reactor containing a solid catalyst. Conversion of the feed was practically complete in all the experiments. The tempera-

tures at which the experiments were carried out and the results of these experiments are given in Table D.

Of experiments 1-12, only experiments 5-7, 9 and 12 are experiments according to the invention. The other experiments fall outside the scope of the invention. They are included in the patent application for the purpose of comparison. No crystalline gallium silicate was used in the comparative experiments 1-4. In the comparative experiments 8, 10 and 11, despite the fact that a crystalline gallium silicate was used, the results were nevertheless unsatisfactory, the reasons being that the silicate was not pre-treated in experiment 8, the reaction temperature was too low in experiment 10, and the gallium silicate had a too high SiO_2/Ga_2O_3 mol. ratio in experiment 11.

A comparison of experiment 7 (with gallium silicate) and experiment 3 (with aluminium silicate) shows that for an equal yield of C_5^+ fraction (63 %wt) the gallium silicate produces a C_5^+ fraction with a much higher aromatic content (83 %wt) than the aluminium silicate (48 %wt).

A comparison of experiment 7 (with gallium silicate) and experiment 4 (with aluminium silicate) shows that for an almost identical aromatic content of the C_5^+ fraction (81-83 %wt) the gallium silicate gives a much higher yield of C_5^+ fraction (63 %wt) than the aluminium silicate (38 %wt). This was also shown by a comparison of experiments 5 and 3.

TABLE D

Experiment No.	1	2	3	4	5	6	7	8	9	10	11	12
Silicate No.	II	I	II	II	IIIA	IIIA	IIIA	IV	IVA	IIIA	VA	IIIA
Temperature, °C	400	400	450	575	350	425	550	425	425	315	500	550
Yield of C_4^- fraction, %wt	16	37	37	62	28	33	37	36	30	3	67	39
Yield of C_5^+ fraction, %wt	84	63	63	38	72	67	63	64	70	97	33	61
Aromatic content of C_5^+ fraction, %wt	19	49	48	81	48	65	83	48	52	2	11	85

The claims defining the invention are as follows:

1. Process for the preparation of an aromatic hydrocarbon mixture, characterized in that a feed of one or more olefins with two to four carbon atoms per molecular or aliphatic hydrocarbons, at least 50 %wt of which consisting of the said olefins, are contacted at a temperature of between 325 and 700°C with a catalyst containing a crystalline gallium silicate, which silicate has the following distinguishing features after calcination for one hour in air at 500°C:

(a) an X-ray powder diffraction pattern that contains as strongest lines the four lines listed in Table A

TABLE A

d(Å)
11.1 ± 0.1
10.0 ± 0.1
3.84 ± 0.07
3.72 ± 0.06

and

(b) in the formula which gives the composition of the silicate expressed in moles of the oxides, the SiO_2/Ga_2O_3 mol. ratio is between 25 and 250,

and that, if the silicate has a SiO_2/Ga_2O_3 mol. ratio of 100-250, the catalyst is subjected one or more times to a two-stage treatment comprising a first stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with a hydrogen-containing reducing gas, followed by a second stage in which the catalyst is contacted for at least 15 minutes at a temperature of 350-700°C with an oxygen-containing oxidizing gas.

2. Process according to claim 1, characterized in that 75 %wt of the feed consists of one or more mono-olefins with three or four carbon atoms per molecule.

3. Process according to claim 2, characterized in that the feed consists substantially completely of one or more mono-olefins with three or four carbon atoms per molecule.

4. Process according to claim 3, characterized in that the feed is a hydrocarbon mixture consisting substantially of C_3 and/or mono-olefins, the mixture being obtained as byproduct of the catalytic or thermal cracking of hydrocarbons.

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5. Process according to one of claims 1-4, characterized in that it is carried out at a pressure of 1-20 bar and a space velocity of 0.1-10 $kg.kg^{-1}.hour^{-1}$.

6. Process according to one of claims 1-5, characterized in that a crystalline gallium silicate is used with a SiO_2/Ga_2O_3 mol. ratio of 60-220.

7. Process according to one or more of claims 1-6, characterized in that the crystalline gallium silicate is prepared from an aqueous mixture containing the following compounds: one or more compounds of an alkali metal (M), one or more organic nitrogen compounds (RN) containing an organic cation or from which an organic cation is formed during the preparation of the silicate, one or more silicon compounds and one or more gallium compounds, and in which mixture the various compounds are present in the following molar ratios, except for the organic nitrogen compounds, expressed in moles of the oxides:

$M_2O : SiO_2 = 0.01-0.35$,

$RN : SiO_2 = 0.02-1.0$,

$SiO_2 : Ga_2O_3 = 25-400$, and

$H_2O : SiO_2 = 5-65$

by maintaining the aqueous mixture at an elevated temperature until the silicate has been formed and then separating the silicate crystals from the mother liquor and calcining them.

8. Process according to one of claims 1-7, characterized in that the crystalline gallium silicate has an alkali metal content of less than 0.05 %wt.

9. Process for the preparation of an aromatic hydrocarbon mixture according to claim 1, substantially as described above and with particular reference to the example.

10. Aromatic hydrocarbon mixtures prepared in accordance with a process as described in claim 9.

DATED this TWENTY FOURTH day of APRIL 1985

SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ B.V.

ESRH04/MF

Patent Attorneys for the Applicant
SPRUSON & FERGUSON