

Nov. 27, 1973

H. R. DEBUS ET AL
HYDROCARBON CONVERSION

3,775,499

Filed May 10, 1972

2 Sheets-Sheet 1

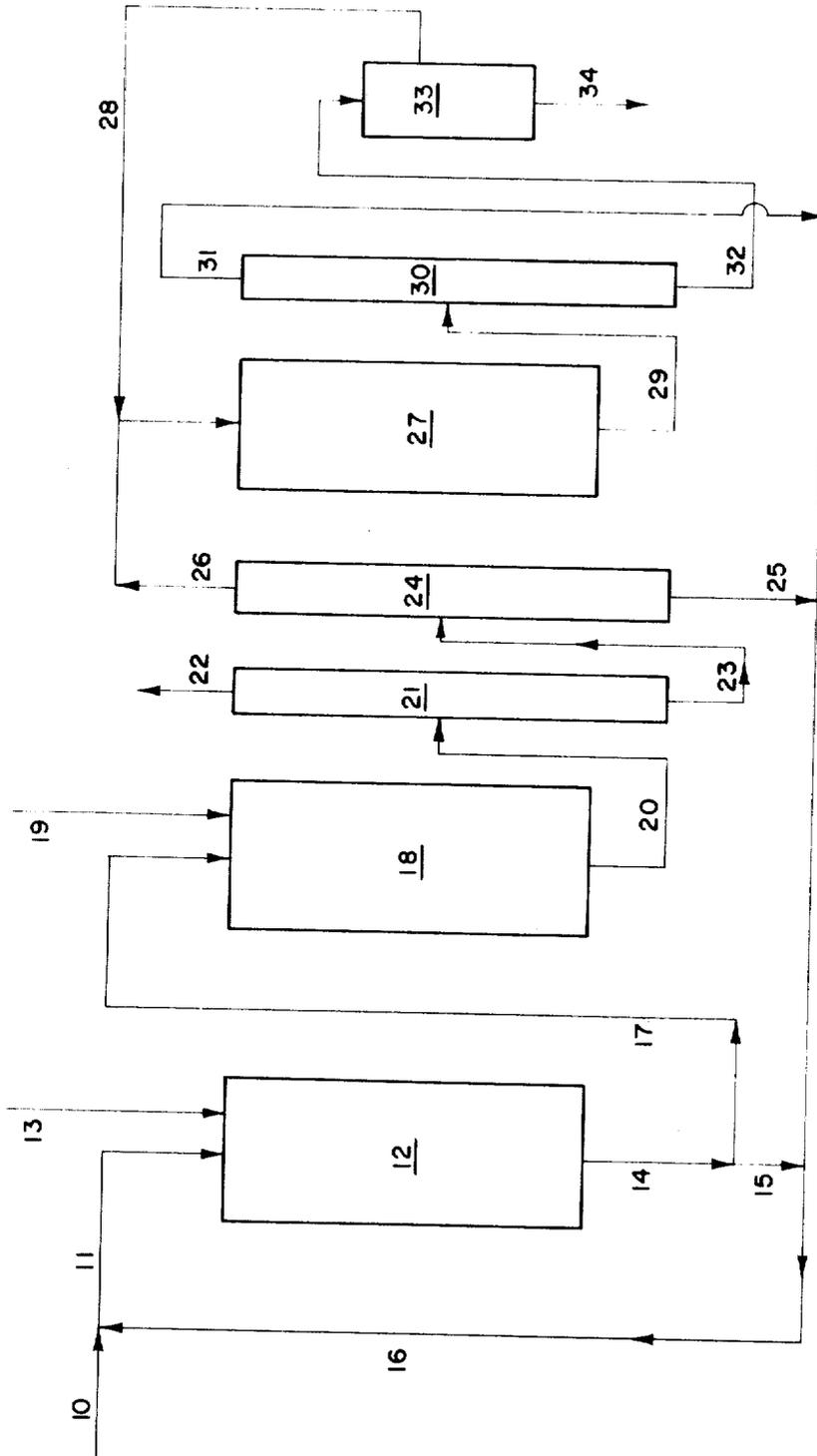


FIGURE 1

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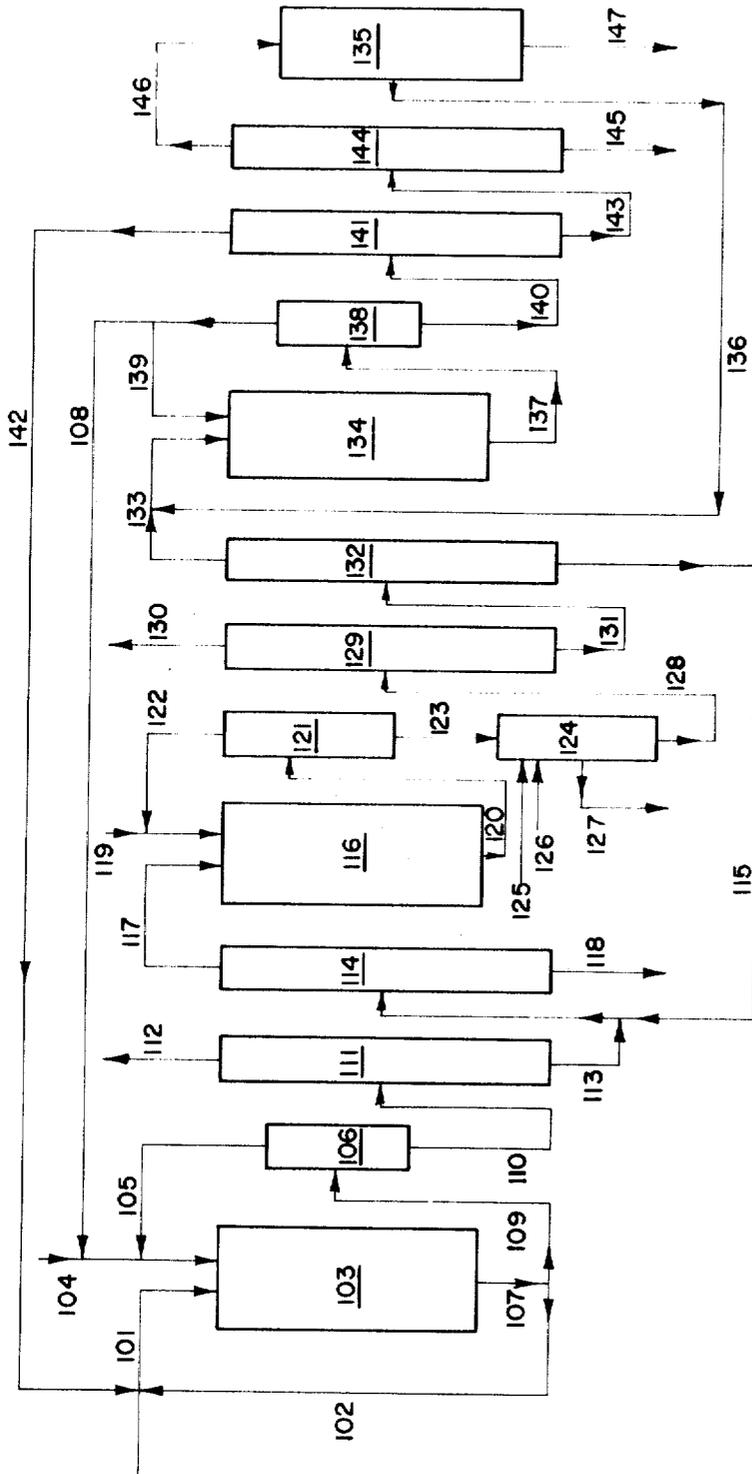


FIGURE 2

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HYDROCARBON CONVERSION

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14 Claims

ABSTRACT OF THE DISCLOSURE

A process for the production of polymethylbenzenes from alkyl aromatic hydrocarbons having at least one alkyl radical with more than one carbon atom, said process comprising hydrogenating said alkyl aromatic hydrocarbons into the corresponding alkylcycloalkanes, isomerizing said alkylcycloalkanes in the presence of Friedel Crafts catalyst to provide polymethylcyclohexanes isomers of said alkylcycloalkanes and then subjecting said polymethylcyclohexanes isomers to a combined dehydrogenation-isomerization step to thereby obtain polymethylbenzenes.

BACKGROUND OF THE INVENTION

This invention relates to a process for the production of polymethylbenzenes from alkylaromatic hydrocarbons having at least one alkyl radical comprising more than 1 carbon atom.

Polymethylbenzenes are interesting materials which are frequently used as feed materials for the production of aromatic polycarboxylic acids, for example phthalic acids from xylenes, pyromellitic acid or anhydride from durene, trimesic acid from trimethylbenzene, etc. The commercial importance of these acids has been increasing over the last few years since they are useful raw materials for the manufacture of synthetic resins. At present, large amounts of alkylaromatic hydrocarbon are now available and are obtained by such methods as for example, by treatment of particular petroleum fractions or as by-products resulting from the manufacture of other widely used compounds. Therefore, it would be particularly advantageous to be able to convert these alkylaromatic hydrocarbons into polymethylbenzenes in an effective manner.

Various processes are known for converting alkylaromatic hydrocarbons having an alkyl radical of more than 1 carbon to polymethylaromatic hydrocarbons. In such processes, the alkyl radicals of more than 1 carbon atom are converted into methyl radicals, a portion of which attached to the aromatic nucleus such that the number of methyl radicals linked to the aromatic nucleus increases. For example, in this manner, ethylbenzene may be converted into dimethylbenzenes. The reaction may be carried out in a 1 step or 2 step process. In the latter case, the starting alkylaromatic hydrocarbons are first hydrogenated and partially isomerized in one step and the resulting products are then further isomerized and dehydrogenated in a second step. The operating conditions of these prior art processes are such that the above defined reactions are always impaired by secondary reactions, more particularly by cracking reactions. Moreover, some of these processes are only available for a single type of alkylaromatic hydrocarbon, so that the results obtained with homologs are distinctly less favorable.

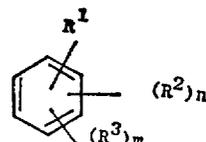
An object of the present invention is to provide a particularly selective process for the production of polymethylbenzenes, with a minimum of secondary reactions, from alkylaromatic hydrocarbons having at least one alkyl radical of more than one carbon atom. Another

object of the present invention is the production of polymethylbenzenes from fractions having a high content of polymethylbenzene precursors. A further object is to provide a process wherein polymethylbenzene precursors are produced with good yields and in an easily recoverable form. Still another object of the invention is to provide a flexible process for the production of polymethylbenzenes which permits the treatment of a wide variety of alkylaromatic hydrocarbons.

In accomplishment of the above and other objects, the present invention provides a process for the production of polymethylbenzenes from alkylaromatic hydrocarbons having at least one alkyl radical with more than one carbon atom, said process comprising hydrogenating said alkylaromatic hydrocarbons into the corresponding alkylcycloalkanes, isomerizing said alkylcycloalkanes in the presence of Friedel Crafts catalyst to provide polymethylcyclohexane isomers and then subjecting said polymethylcyclohexane isomers to a combined dehydrogenation isomerization step to thereby obtain polymethylbenzenes.

To further describe the process of the present invention, reference will hereinafter be made to the accompanying drawings of which FIG. 1 is a diagrammatic representation of an embodiment of the process of the present invention and FIG. 2 is a diagrammatic flow diagram of another and more specific embodiment of the process of the present invention.

The starting alkylaromatic hydrocarbons of the process of the present invention have the general formula



Wherein R^1 is an aliphatic radical containing two to six, preferably two to three, carbon atoms, R^2 is an aliphatic radical containing from one to three carbon atoms, R^3 is a methyl radical, n is an integer from zero to three inclusive, and m is 0 or 1, the total number of carbon atoms of the alkylaromatic hydrocarbon being at least eight.

The alkylaromatic hydrocarbons may be introduced into the first step of the present process either alone or in admixture with one another or with their hydrogenated products. The feed desirably is substantially water-free if the product of the first step is to be subjected to the second step isomerization without further processing since the isomerization reaction is impaired by the presence of water. Among the hydrocarbons having the above formula, ethylbenzene, cumene, diethylbenzenes, cymenes and triethylbenzenes preferably are used. These materials are readily available and are converted in the present process to the di-, tri-, tetra- and hexa-methylbenzenes which are in turn used for the production of the corresponding benzene di-, tri-, tetra- and hexacarboxylic acids.

The hydrogenation of the alkylaromatic hydrocarbon feed in accordance with the first step of the present process is carried out in the presence of a catalyst containing a metal of group VIII of the Periodic Table such as nickel, or a metal of the platinum group such as platinum, palladium, ruthenium, osmium, rhodium and the like or a platinum alloy, such as an alloy with rhenium. The catalyst generally includes a carrier, a support, such as silica, alumina, a mixture of silica and alumina or a zeolitic compound. It can be employed in any one of various possible forms, that is in granules or pellets or in fine powder form. The catalyst can be employed in a fixed bed or fluidized or other moving bed. Catalysts comprising nickel on silica and containing from 5 to 60% by weight of

nickel, and catalyst comprising platinum on a carrier and containing 0.1 to 1.5% by weight of platinum, generally are used.

The hydrogenation reaction being highly exothermic, it is preferable to dilute the feed with recycled products issuing from a prior hydrogenation and/or the second step isomerization. The ratio by weight of fresh feed to recycled products may be varied between wide limits, for example between 1:1 and 1:50, but is preferably between 1:2 and 1:10. The reaction temperature generally is between 150 and 350° C. and particularly, between 200 and 300° C.

In order to avoid any deactivation of the isomerization catalyst during the second step of the process, the hydrogenation of the starting hydrocarbons should be as complete as possible. For this purpose, the partial pressure of these hydrocarbons may be reduced by using a high ratio of hydrogen to hydrocarbons. However, the use of large amounts of hydrogen is not economical and, generally, the ratio of hydrogen to hydrocarbons (fresh feed plus recycled hydrocarbons) may be varied from 2:1 to 30:1 and, preferably, between 3:1 and 20:1.

The hydrogenation yields may be increased by using higher pressures but, generally, the pressure is kept between 20 and 70 kg./cm.². Indeed, higher pressures do not result in an improvement in the results such as to justify additional cost of the such higher pressures.

The space velocity of the feed in the hydrogenation step depends on many factors, such as dilution ratio, temperature and pressure. A liquid space velocity with respect to the total feed on the order of 0.5 to 50 will be found suitable and generally the preferred residence time may be varied between 1 and 12 volumes per hour per volume of catalyst.

The interdependence of the operating conditions of the hydrogenation reaction is shown in Table I below, which gives the results of hydrogenation experiments with a mixture comprising equal parts of diethylbenzenes and diethylcyclohexanes in the presence of a catalyst consisting of platinum (0.9% by weight) on a silica-alumina carrier, under a pressure of 50 kg./cm.². The explored variables were: temperature, molar ratio of hydrogen to hydrocarbons of the feed (H₂/HC) and space velocity (S.V.) in litres per hour per litre of catalyst.

The operating conditions were:

Experiment A: H₂/HC=3.5:1, S.V.=2

Experiment B: H₂/HC=9:1, S.V.=2

Experiment C: H₂/HC=9:1, S.V.=1

For each experiment, the percentage of nonhydrogenated hydrocarbon in the reaction products is given in Table I.

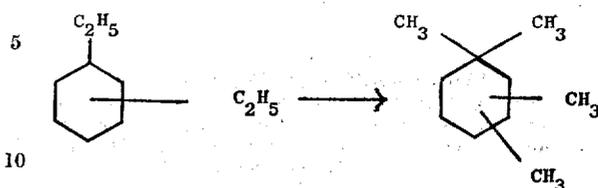
TABLE I

Temperature (° C.)	Experiment		
	A	B	C
305	0.04	0.01
300	0.02	0.01
280	0.01	0.002	0.001
270	0.002
250	0.001	0.001
240	0.001	0.001

Another set of experiments has been performed with a catalyst containing nickel (50% by weight) on silica, at 240° C. and 50 kg./cm.², with a H₂/HC of 6:1 and a S.V. of 4. Under these conditions, the percentage of unreacted hydrocarbon is lower than 0.001.

The second step of the process comprises isomerizing the alkylcycloalkanes obtained from the hydrogenation reaction into polymethylcycloalkanes. For example, diethylcyclohexane is isomerized with breakdown of the ethyl groups and formation of methyl groups, a substan-

tial portion of which are gem methyl groups, according to the following reaction:



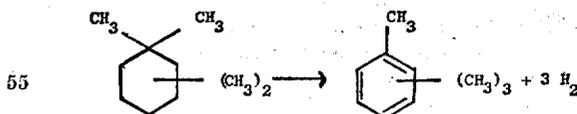
This reaction is carried out with Friedel-Crafts catalysts such as AlCl₃, BF₃, SbCl₃, SnCl₄, ZnCl₂, TiCl₄, HF, GaCl₃, FeCl₃, AlBr₃, and the like.

In order to illustrate the formation of gem-polymethylcycloalkanes from hydrocarbons containing at least 10 carbon atoms, isomerization experiments were performed with diethylcyclohexane in the presence of AlCl₃. The isomerization products were divided into five equal parts having increased boiling points. It was found that, with the C₁₀ hydrocarbons, tetramethylbenzene precursors were found primarily in the fractions having boiling points substantially between 150 and 165° C. These fractions had a high content of gem-tetramethylcyclohexanes. The gem-polymethylcycloalkanes have a lower boiling point than the corresponding polymethylcyclohexanes which are not geminated and consequently they can easily be separated by distillation.

The isomerization reaction is generally carried out at a temperature lower than 150° C., and more particularly at a temperature between about 0 and 70° C., in order to avoid secondary reactions, for example, cracking and disproportionation reactions. A promotor may be added and, in the case of AlCl₃, this promotor can be gaseous HCl, used alone, an olefin or chlorinated hydrocarbon, such as butyl chloride or mixtures thereof. Pressure may be atmospheric or superatmospheric, the choice depending largely on economic and practical considerations.

The space velocity used during the isomerization step depends on other factors, such as temperature, type and amount of catalyst, presence or absence of promotors, etc. However, most of the experiments have shown that the most useful contact times between the feed and the catalyst will be between 0.1 and 10 hours.

The polymethylcycloalkanes issuing from the second step of the process are next subjected to a combination dehydrogenation isomerization step to thereby form polymethylbenzenes. In this step, the reaction is illustrated by the following:



The polymethylcycloalkanes of the second step can be sent directly to the third step of the process and such a procedure usually is applied when the hydrocarbons contain 8 or 9 carbon atoms. However, in the case of hydrocarbons with more than 9 carbon atoms, the polymethylcycloalkanes desirably are first distilled in order to separate the gem. compounds from the other products. The gem. compounds are then simultaneously dehydrogenated and isomerized, while the higher boiling hydrocarbons may be recycled to the second step of the process.

The catalyst which is used for the third step of the present invention is difunctional with acidic sites to promote the isomerization reaction and other sites to promote dehydrogenation. The catalyst for the third step generally is a metal of the platinum group or an alloy of platinum, i.e., with rhenium, on a carrier consisting of silica and/or alumina or a zeolitic compound. Such a difunctional catalyst contains preferably between 0.1 and 5% and more

particularly from about 0.2% to 3% by weight, of platinum on an amorphous or crystalline silica alumina carrier consisting of 5% to 95% of silica and preferably 10 to 50% by weight of silica for amorphous silica-alumina and 30 to 90% by weight of silica for crystalline silica-alumina.

The dehydrogenation reaction is endothermic and thus is promoted by high temperatures. However, in order to prevent cracking, it is preferable to work at lower temperatures. A practical compromise between temperature and rate of reaction most often is employed and will vary depending upon the choice of the operator. Generally, however, the temperature employed is between 200° and 600° C. and more preferably between 300° and 500° C. The pressure usually will be less than 50 kg./cm.², more particularly between atmospheric and 35 kg./cm.² while the space velocity of the hydrocarbons will be between 0.5 and 30 volumes of hydrocarbons per hour per volume of catalyst.

Comparative experiments have shown that with the process of the present invention, alkylaromatic hydrocarbons as defined above, are converted into polymethylbenzenes with yields and selectivities distinctly higher than those obtained with prior art processes.

For example, a feed containing 100% ethylbenzene was hydrogenated and the ethylcyclohexane obtained was isomerized in the presence of AlCl₃. The isomerization products were then dehydrogenated without prior distillation. The analysis of the products was the following:

	By weight, percent
Light products (gaseous products and hydrocarbons having up to 7 C) -----	13.5
Ethylbenzene -----	6.1
p-Xylene -----	19.5
m-Xylene -----	47.8
o-Xylene -----	13.1

With the process of the present invention, the conversion yield of ethylbenzene into xylenes is thus 80.4% by weight, while prior processes give yields within the range of 40 to 45% by weight.

In another run, cumene was treated according to the process of the present invention. On analysis, it was found that the products contained 68.6% by weight of trimethylbenzenes. A yield of about 45% generally is obtained from conventional known processes.

The features and other characteristics of the process of the present invention will be further described and exemplified by the following non-limiting examples, which are described with reference to the accompanying drawings.

EXAMPLE 1

A feedstock consisting mainly of diethylbenzenes was treated in accordance with the process of the present invention as shown in FIG. 1. This alkylaromatic feed had been pre-dried. The feed mixture, 100 parts by weight, was passed through line 10 into admixture with a recycle stream containing material from the hydrogenation from the isomerization and from the dehydrogenation-isomerization steps of the process of the present invention. The resulting mixture representing 576.8 parts by weight, was sent by line 11 into hydrogenation reactor 12.

In hydrogenation reactor 12, the mixture of fresh alkylaromatic feed and recycle was passed over a catalyst containing 50% by weight of nickel and the remainder being a silica carrier upon which the nickel was distributed. Hydrogen was introduced into hydrogenation reactor 12 by means of line 13 concurrently with the hydrocarbon feed material being introduced via line 11. The molar ratio of hydrogen introduced to hydrocarbon feed was 6:1. The liquid hourly space velocity employed was 4 and the pressure was 50 kg./cm.² while the temperature was maintained at about 240° C.

The hydrogenated product exited hydrogenation reactor 12 by means of line 14 and was separated into a recycle stream (301 parts by weight) and an isomerization feed stream (275.8 parts by weight). The recycle stream passes by line 15 into a common recycle line 16 through which the combined recycle stream of the hydrogenation, the isomerization and the dehydrogenation-isomerization steps pass into line 10 and admixture with fresh feed to the hydrogenation reactor 12. The isomerization feed passes from line 14 via line 17 into an isomerization reaction zone 18.

Within reaction zone 18, the hydrogenated hydrocarbons were contacted with an AlCl₃ catalyst. The concentration of AlCl₃ catalyst in the resulting reaction mass was 30% by weight. Concurrently with the introduction of the hydrogenated hydrocarbons into isomerization reaction zone 18, anhydrous HCl is introduced into reaction zone 18 via line 19. In an alternate arrangement, the anhydrous HCl is admixed with the hydrogenated hydrocarbons in line 17 and the resulting mixture introduced into reaction zone 18.

The operating conditions within reaction zone 18 were a temperature of 30° C.±5° C., atmospheric pressure and a contacting time of 2 hours. The amount of HCl added was sufficient to keep the hydrogenated hydrocarbon feed substantially saturated under the conditions employed. Used AlCl₃ was periodically withdrawn and fresh AlCl₃ added to insure that its activity remained substantially constant.

The products of the isomerization reaction zone 18 were removed and passed by means of line 20 to a first fractionation column 21. The more volatile components boiling below 150° C. were taken overhead from column 21 via line 22. This fraction representing 14.1 parts by weight, may be sent to any utility or further processing such as reforming or may be sent to waste disposal. The bottoms fraction of column 21 was sent by line 23 to a second distillation column 24. From second distillation column 24, a 150–165° C. boiling fraction representing 155.4 parts by weight, was taken overhead via line 26. The heavier boiling bottoms fraction representing 106.3 parts by weight was removed from the bottom of column 24 by means of line 25 and recycled to the hydrogenation reactor via line 16.

The overhead of column 24 is passed through line 26 into a dehydrogenation-isomerization zone 27. The residual liquor from a crystallization step following the dehydrogenation-isomerization step is introduced via recycle line 28 into line 26 and into admixture with the overhead from column 24. The amount of such residual liquor represented 228.5 parts by weight.

Within the dehydrogenation-isomerization zone 27, the hydrocarbon reactant mixture was contacted with a catalyst containing 0.5% by weight of platinum deposited on a silica-alumina support. The operating conditions within dehydrogenation-isomerization zone 27 were a temperature of 425° C., a pressure of 15 kg./cm.² and a liquid hourly space velocity of 6. A hydrogen to hydrocarbon ratio of 9:1 was employed within the dehydrogenation-isomerization zone 27.

The products of dehydrogenation-isomerization zone 27 are removed via line 29 and thereby introduced into a fractionating column 30. From fractionating column 30, lighter components (69.5 parts by weight) having a boiling point lower than 192° C. were taken overhead by line 31 and introduced into line 16 whereby such components are recycled to hydrogenation reactor 12. The bottoms fraction (314.3 parts by weight) from fractionating column 30 was passed by means of line 32 to crystallization zone 33. In crystallization zone 33, durene (85.8 parts by weight) was crystallized from the liquor. The durene was removed from crystallization zone 33 by line 34. The remaining mother liquor within crystallization zone 33 was removed by line 28 as above discussed for recycle to dehydrogenation-isomerization zone 27.

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EXAMPLE 2

A second embodiment of the process of the present invention was carried out employing the scheme of FIG. 2. In this embodiment, a feedstock containing approximately 80% diethylbenzenes, the remainder being about equal in lower and higher boiling aromatics, is introduced by line 101 into admixture with a recycle stream (more fully discussed below) from line 102 and the resulting mixture passed into hydrogenation reactor 103. Within hydrogenation reactor 103, the feed-recycle mixture is treated with hydrogen introduced into reactor 103 via line 104. The hydrogen employed includes fresh hydrogen as well as recycle hydrogen introduced into line 104 by line 105 from separating tank 106 and recycle hydrogen introduced by line 108 from separating tank 138. The catalyst and operating conditions utilized in hydrogenation reactor 103 are substantially the same as those employed in the like step of Example 1 above.

The products of hydrogenation reactor 103 are withdrawn through line 107 and separated into two parts. One part is passed into recycle line 102 for reintroduction into hydrogenation reactor 103. The remaining portion of the product of hydrogenation reactor 103 is conducted via line 109 to separating tank 106.

In separating tank 106, unreacted hydrogen is separated and recycled by line 105 to hydrogen entry line 104. The remaining liquid product is passed by line 110 to a first distillation column 111.

From distillation column 111, the materials boiling below 150° C. are taken overhead via line 112 to further processing such as to a reforming unit. The heavier bottoms fraction passes from distillation column 111 by means of line 113 to a second distillation column 114. Along with the bottoms fraction withdrawn from column 111 is introduced a recycle stream 115 from the isomerization step of the present process.

A fraction boiling within the range of 150–180° C. is taken overhead from distillation column 114 by line 117 through which it is passed to isomerization reactor 116. The materials boiling above 180° C. are removed from distillation column 114 by means of line 118.

Within isomerization reactor 116, catalyst and operating conditions are substantially those described for the like step of Example 1, HCl being introduced into isomerization reactor 116 via line 119.

The products of the isomerization reaction are removed by line 120 and passed to a stripper column 121. HCl is removed overhead from stripper column 121 and recycled by line 122 into admixture with fresh HCl in line 119 for entry into isomerization reactor 116. From stripper column 121, the stripped isomerization product passes by line 123 into wash tank 124 into which soda is introduced by line 125 and water is introduced by line 126. Within wash tank 124, the isomerized product is first soda washed and then water washed. The wash liquors pass from wash tank 124 by line 127 to waste disposal or other processing. The washed isomerization product passes from wash tank 124 by line 128 through which it is introduced into distillation column 129.

From distillation column 129, materials boiling lower than 150° C. are removed overhead by line 130 and sent to further processing such as reforming or to disposal. The remaining bottoms portion is taken from distillation column 129 by means of line 131 through which it is passed to a second distillation column 132. From distillation column 132, a fraction boiling within the range of 150–165° C. is taken overhead through line 133 and thereby introduced into a dehydrogenation-isomerization reactor 134. The bottoms fraction of distillation column 132 is removed and recycled via line 115 into line 113 and then into distillation column 114.

Along with the overhead fraction from distillation column 132, the mother liquor from crystallization zone 135 is introduced by means of line 136 and line 133 into reactor 134. The catalyst and operating conditions of reactor

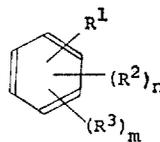
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134 are substantially the same as those employed in the like step of Example 1. Products of reactor 134 pass by line 137 to a separating tank 138 in order to recover hydrogen a portion of which is recycled through line 139 to reactor 134, the remainder being recycled by line 108 to line 104 and hydrogenation reactor 103.

The liquid products of separating tank 138 pass by line 140 to a distillation column 141. In distillation column 141, the fraction is separated into a lighter fraction boiling below 192° C. which is conducted by line 142 to line 101 for recycle to the hydrogenation reaction step of the present process. The heavier or bottoms fraction is taken by line 143 from distillation column 141 to distillation column 144. From this distillation column 144 a heavy fraction boiling above 210° C. is taken by line 145. This fraction may be further processed as desired. Overhead from column 144 is taken by line 146 a 192–210° C. boiling fraction which is passed to crystallization zone 135 wherein durene is crystallized. The durene is removed via line 147 while the mother liquor is recycled by line 136 to line 133 for reentry into reactor 134.

What is claimed is:

1. A process for the production of polymethylbenzenes containing at least three methyl groups from alkylaromatic hydrocarbons having the formula



wherein R¹ is an alkyl radical containing 2 to 6 carbon atoms, R² is an alkyl radical containing from 1 to 3 carbon atoms, R³ is a methyl radical, *n* is an integer from 1 to 3 inclusive and *m* is 0 or 1, said process comprising:

- hydrogenating said alkylaromatic hydrocarbons into the corresponding alkylcycloalkanes, the percentage of unreacted hydrocarbons not being higher than about 0.001;
- isomerizing said alkylcycloalkanes in the presence of AlCl₃ at a temperature not greater than 150° C. for a time sufficient to form a reaction product containing substantially gem structured polymethylcyclohexanes having the same number of carbon atoms as the feed material;
- contacting said reaction product with an acidic, difunctional dehydrogenation-isomerization platinum catalyst in the presence of hydrogen in a reaction zone maintained at a temperature in the range of from 200° to 600° C. and a pressure not greater than 750 p.s.i.; and
- recovering a reaction product containing polymethylbenzenes of the same number of carbon atoms as the feed material.

2. The process of claim 1 wherein R¹ is an aliphatic radical containing from 2 to 3 carbon atoms and R² is an aliphatic radical containing from 2 to 3 carbon atoms.

3. The process of claim 2 wherein the alkyl aromatic hydrocarbon is one selected from the group consisting of diethylbenzenes, cymene and triethylbenzene.

4. The process of claim 1 wherein hydrogenation is carried out in the presence of a catalyst containing a metal of Group VIII of the Periodic Table deposited upon a support selected from the group consisting of silica, alumina, silica-alumina, silica or alumina containing zeolitic compounds and mixtures thereof.

5. The process of claim 4 wherein the group VIII metal is one selected from the group consisting of nickel, platinum, palladium, ruthenium, osmium, rhodium, mixtures thereof and alloys and mixtures with rhenium.

6. The process of claim 1 wherein the hydrogenation reaction is carried out at a temperature within the range of 150 to 360° C.

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7. The process of claim 1 wherein hydrogen employed in the hydrogenation reaction is present in a molar ratio of hydrogen to hydrocarbons within the range of 2 to 1 to 30 to 1.

8. The process of claim 1 wherein the hydrogenation reaction is carried out at a pressure within the range of 20 to 70 kg./cm.².

9. The process of claim 1 wherein the hydrogenation reaction is carried out at a liquid hourly space velocity of 0.5 to 50.

10. The process of claim 1 wherein the isomerization reaction is carried out in the presence of a promoter selected from the group consisting of gaseous HCl, olefins, chlorinated hydrocarbons and mixtures thereof.

11. The process of claim 1 wherein said catalyst used in the dehydrogenation-isomerization reaction is one containing a metal selected from the group consisting of platinum and alloys of platinum with rhenium deposited on the carrier selected from the group consisting of silica, alumina, silica-alumina, silica or alumina containing zeolitic compounds and mixtures thereof.

12. The process of claim 1 wherein the combined dehydrogenation-isomerization reaction is carried out at a pressure within the range of 7 to 50 kg./cm.² and wherein the liquid hourly space velocity is within the range of 0.5 to 30.

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13. The process of claim 1 wherein durene is prepared from an alkyl aromatic hydrocarbon consisting essentially of diethyl benzenes, further comprising the step of separating from the reaction product of isomerization step (b) a fraction essentially pure in gem structured tetramethylcyclohexanes and thereafter conducting only said gem structured tetramethylcyclohexane fraction to the dehydrogenation-isomerization step (c).

14. The process of claim 13, wherein said separation is accomplished by fractional distillation.

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