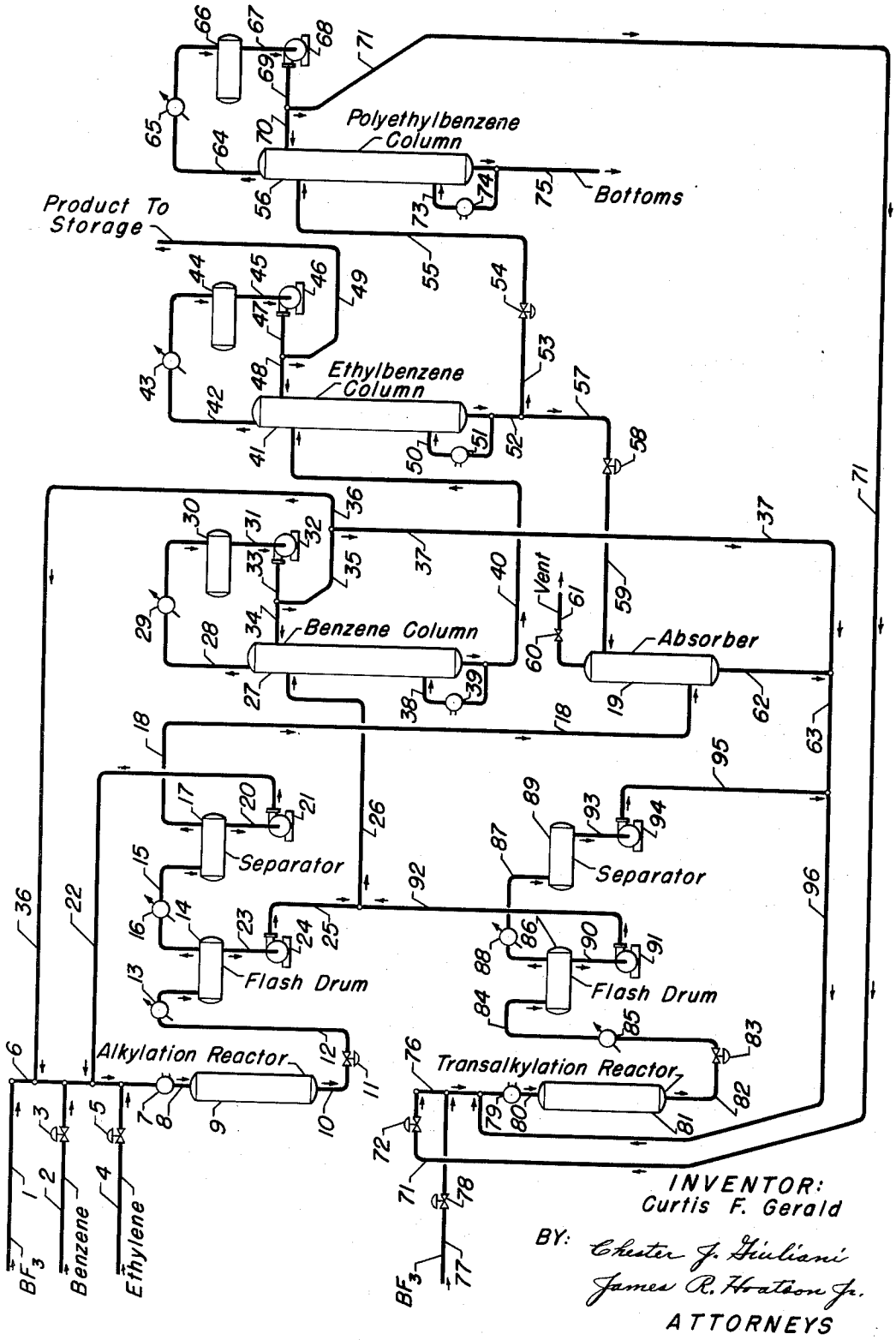
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ALKYLATION-TRANSALKYLATION PROCESS

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ALKYLATION-TRANSALKYLATION PROCESS

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This application is a continuation-in-part of my copending application Serial No. 141,498, filed September 28, 1961, now abandoned.

This invention relates to a process for the production of an aromatic compound, and more particularly relates to a process for the alkylation of an alkylatable aromatic compound with an olefin-acting compound, and still more particularly relates to the alkylation of an aromatic hydrocarbon with an olefinic hydrocarbon which may be in combination with other gases which are unreactive at the process conditions utilized. Further, this invention relates to a combination process including the steps of alkylation, transalkylation, gas-liquid separation, fractionation, and gas-liquid absorption.

An object of this invention is to produce alkylated aromatic hydrocarbons, and more particularly, to produce monoalkylated benzene hydrocarbons. A specific object of this invention is a process for the production of ethylbenzene, a desired chemical intermediate, which ethylbenzene is utilized in large quantities in dehydrogenation processes for the manufacture of styrene, one of the starting materials for the production of resins and some synthetic rubber. Another specific object of this invention is to produce alkylated aromatic hydrocarbons boiling within the gasoline boiling range having high anti-knock value and which may be used as such or as a component of gasoline suitable for use in automobile and airplane engines. A further specific object of this invention is a process for the production of cumene by the reaction of benzene with propylene, which cumene product is oxidized in large quantities to form cumene hydroperoxide which is readily decomposed into phenol and acetone. Another object of this invention is to provide a process for the introduction of alkyl groups into aromatic hydrocarbons of high vapor pressure at normal conditions with minimum loss of said high vapor pressure aromatic hydrocarbons and maximum utilization thereof in the process. Still another object of this invention is a process in which molar execesses of aromatic hydrocarbons to be alkylated are utilized, and in which process the yield of monoalkylated aromatic hydrocarbon product is exceptionally high due to maximum consumption of polyalkylated aromatic hydrocarbon by-products in the process. The further object of maximum boron trifluoride utilization as a catalyst in this process, along with other objects of this invention, will be set forth hereinafter as part of the accompanying specification.

In prior art processes for the alkylation of aromatic hydrocarbons with alkylating agents, it has been disclosed that it is preferable to utilize molar execesses of such aromatic hydrocarbons. In such processess, it is generally preferable to utilize greater than two mols of aromatic hydrocarbon per mol of alkylating agent and in many cases, for best reaction, it is preferred to utilize four or more mols of aromatic hydrocarbon per mol of alkylating agent. When the alkylating agent is an olefin hydrocarbon, this has been found to be particularly necessary to prevent polymerization of the olefin hydrocarbon from taking place prior to the reaction of the olefin hydrocarbon with the aromatic hydrocarbon. Further, it has been found advantageous for maximum olefin utilization in the process. Due to the effect of the Law of Mass Action, one might expect that the yield of polyalkylated aromatic hydrocarbons which results therefrom would

be minimized. While this is generally true, substantial yields of polyalkylated aromatic hydrocarbons are formed even when utilizing such molar excess of aromatic hydrocarbon reactant. Formation of these polyalkylated aromatic hydrocarbons naturally increases the consumption of aromatic hydrocarbon in the process based on the yield of desired alkylated aromatic hydrocarbon. This is an obvious economic disadvantage since one seeks to achieve not only maximum alkylating agent consumption in the process, but also, maximum utilization of aromatic hydrocarbon to desired product. The prior art teaches that maximum utilization of aromatic hydrocarbon to desired product may be increased by recycling of the by-product polyalkylated aromatic hydrocarbons formed in the process back to the alkylation zone so that simultaneous transalkylation may occur. This is to be the usual procedure if the reaction is equilibrium limited, since equilibrium is achieved by placing all components into one reactor and then utilizing conditions of temperature, pressure, and residence time in the reactor to obtain equilibrium. However, it has been found that transalkylation is favored by higher aromatic hydrocarbon to alkylating agent ratios than are necessary for alkylation, indicating that kinetics as well as equilibrium must be involved in obtaining maximum aromatic hydrocarbon utilization. Since the higher aromatic hydrocarbon to polyalkylated aromatic hydrocarbon ratio favors transalkylation, it would appear to be logical to recycle polyalkylated aromatic hydrocarbons formed in the process plus recycle and fresh aromatic hydrocarbon to a first transalkylation reactor or zone followed by addition of the alkylating agent to the effluent therefrom prior to passage thereof to the alkylation zone. This procedure would insure the highest ratio in the transalkylation zone. Then, this effluent plus alkylating agent could be reacted in an alkylation zone at a lower aromatic hydrocarbon to alkylating agent ratio since some aromatic hydrocarbon would have been consumed in the transalkylation zone. However, when attempting to operate in this manner, the alkylation or second reaction zone catalyst rapidly loses activity so that unreacted alkylating agent is found in the effluent. It has unexpectedly been found that if alkylatable aromatic compound plus olefin-acting compound are reacted in an alkylation zone, and simultaneously additional alkylatable aromatic compound and polyalkylated aromatic compound containing substantially no monoalkylated aromatic compound are reacted in a transalkylation zone, and if the effluents from said reaction zones are flashed by reduction in pressure prior to gas-liquid separation, and the gas-free liquid phases from the alkylation zone and the transalkylation zone are commingled and then passed to further separation means, the catalyst in both zones achieves very long life and extremely high conversion of the aromatic compound to monoalkylated derivative thereof is obtained. This occurs in spite of the fact that no olefin-acting compound and substantially no monoalkylated aromatic compound are present as feed to the transalkylation zone. In addition, neither inert gas nor ethane is present in the transalkylation reaction zone making boron trifluoride recovery much easier and more efficient inasmuch as the boron trifluoride becomes part of the flashed liquid aromatic compound recycle. By operating according to the process of this invention, a vastly superior, improved processing scheme results, and, as compared to prior art processes, not only is this design more economical due to lower heat input requirements, better reaction control and more efficient catalyst utilization, but such an improved processing scheme gives great versatility of operation since said processing scheme is more flexible in an integrated refinery or petrochemical complex than previous prior art processes.

A further problem arises in connection with the utilization of molar excesses of aromatic compound to be alkylated. This problem is related to the use of such molar excesses in connection with the alkylation of aromatic hydrocarbons of high vapor pressure at normal conditions, particularly when the olefin-acting compound utilized is a normally gaseous olefin hydrocarbon such as ethylene, propylene, 1-butene, 2-butene, or isobutylene, and this problem is further accentuated when the alkylation is carried out in the presence of a gaseous acidic catalyst such as exemplified by boron trifluoride. The above-mentioned olefin hydrocarbons are often present as minor quantities in various refinery gas streams containing major quantities of other gases such as hydrogen, nitrogen, hydrogen sulfide, and hydrocarbons such as methane, ethane, propane, n-butane and isobutane. It has become very desirable to utilize such gas streams for their olefin content and a problem has arisen therewith which is related thereto and to the utilization of the gaseous acidic catalyst, such as boron trifluoride. This problem is also solved by the utilization of the process of the present invention, which process results in maximum yield of desired alkylated aromatic hydrocarbon and minimum loss of alkylating agent, alkylatable aromatic hydrocarbon, and gaseous acidic catalyst thereby further yielding a considerable economic advantage over prior art processes.

One embodiment of the present invention relates to a process for the production of an alkylaromatic compound which comprises alkylating and alkylatable aromatic compound with an olefin-acting compound in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous inorganic oxide, withdrawing and separating from said alkylation zone a gas phase and a gas-free liquid phase, commingling the gas-free liquid phase from said alkylation zone with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, separating from the resultant gas-free liquid phase mixture unreacted aromatic compound, desired monoalkylated aromatic compound, and higher molecular weight polyalkylated aromatic compound, recycling at least a portion of said unreacted aromatic compound to the alkylation zone, removing desired monoalkylated aromatic compound as product from the process, passing said polyalkylated aromatic compound in admixture with alkylatable aromatic compound and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous inorganic oxide and therein reacting the polyalkylated aromatic compound with the alkylatable aromatic compound, withdrawing and separating from said transalkylation zone a gas phase and a gas-free liquid phase, and passing said gas-free liquid phase to said commingling step as aforesaid.

Another embodiment of the present invention relates to a process for the production of an alkylaromatic hydrocarbon which comprises alkylating a benzene hydrocarbon with an olefinic hydrocarbon in the presence of not more than 1.0 gram of boron trifluoride per gram mol of olefinic hydrocarbon in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous inorganic oxide, withdrawing and separating from said alkylation zone a gas phase and a gas-free liquid phase, commingling the gas-free liquid phase from said alkylation zone with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, separating from the resultant gas-free liquid phase mixture unreacted benzene hydrocarbon, desired monoalkylated benzene hydrocarbon, and higher molecular weight polyalkylated benzene hydrocarbon, recycling at least a portion of said unreacted benzene hydrocarbon to the alkylation zone, removing desired monoalkylated benzene hydrocarbon as product from the process, passing said polyalkylated benzene hydrocarbon in admixture with alkylatable aromatic compound and from 0.002 to about 1.2 grams of boron trifluoride per gram mol of polyalkylated benzene hydrocarbon to a transalkylation zone containing boron trifluoride-modified substantially anhydrous inorganic oxide and therein reacting the polyalkylated benzene hydrocarbon with the alkylatable benzene hydrocarbon, withdrawing and separating from said transalkylation zone a gas phase and a gas-free liquid phase, and passing said gas-free liquid phase to said commingling step as aforesaid.

A further embodiment of the present invention relates to a process for the production of an alkylaromatic hydrocarbon which comprises alkylating a benzene hydrocarbon with a normally gaseous olefin in the presence of not more than 1.0 gram of boron trifluoride per gram mol of olefin in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous gamma-alumina, withdrawing and separating from said alkylation zone a gas phase and a gas-free liquid phase, commingling the gas-free liquid phase from said alkylation zone with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, separating from the resultant gas-free liquid phase mixture unreacted benzene hydrocarbon, desired monoalkylated benzene hydrocarbon and higher molecular weight polyalkylated benzene hydrocarbon, recycling at least a portion of said unreacted benzene hydrocarbon to the alkylation zone, removing desired monoalkylated benzene hydrocarbon as product from the process, passing said polyalkylated benzene hydrocarbon in admixture with alkylatable benzene hydrocarbon and from about 0.002 to about 1.2 grams of boron trifluoride per gram mol of polyalkylated benzene hydrocarbon to a transalkylation zone containing boron trifluoride-modified substantially anhydrous gamma-alumina and therein reacting the polyalkylated benzene hydrocarbon with the alkylatable benzene hydrocarbon, with drawing and separating from said transalkylation zone a gas phase and a gas-free liquid phase, and passing said gas-free liquid phase to said commingling step as aforesaid.

A specific embodiment of the present invention relates to a process for the production of ethylbenzene which comprises alkylating benzene with ethylene in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous alumina, withdrawing and separating from said alkylation zone a gas phase and a gas-free liquid phase, commingling the gas-free liquid phase from said alkylation zone with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, separating from the resultant gas-free liquid phase mixture unreacted benzene, desired ethylbenzene, and high molecular weight polyethylbenzenes, recycling at last a portion of said benzene to the alkylation zone, removing desired ethylebenzene as product from the process, passing said polyethylbenzenes in admixture with benzene and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous alumina and therein reacting the polyethylbenzenes with the benzene, withdrawing and separating from said transalkylation zone a gas phase and a gas-free liquid phase, and passing said gas-free liquid phase to said commingling step as aforesaid.

This invention can be most clearly described and illustrated with reference to the attached drawing. While of necessity, certain limitations must be present in such a schematic description, no intention is meant thereby to limit the generally broad scope of this invention. As stated hereinabove, the first step of the process of the present invention comprises alkylating an alkylatable aromatic compound with an olefin-acting compound in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous inorganic oxide. In the drawing, this first step is represented as taking place in

reaction zone 9, labeled alkylation reactor. However, the mixture of boron trifluoride, alkylatable aromatic compound, and olefin-acting compound must be furnished to this reaction zone. In the drawing, the boron trifluoride is represented as being furnished to reaction zone 9 through line 1. The alkylatable aromatic compound, labeled benzene, is combined therewith in line 6 by passage through line 2 through pressure control valve 3. The olefin-acting compound, labeled ethylene, is combined therewith in line 6 by passage through line 4 through pressure control valve 5. The combined feed passes to reaction zone 9 via line 8 containing heater 7 and is distributed to alkylation zone 9 by conventional distributing means not shown in the drawing but contained in the upper portion of zone 9.

The olefin-acting compound, particularly olefin hydrocarbon, which may be charged to reaction zone 9 via lines 4 and 8, may be selected from diverse materials including monoolefins, diolefins, polyolefins, acetylenic hydrocarbons, and also alcohols, ethers, and esters, the latter including alkyl halides, alkyl sulfates, alkyl phosphates, and various esters of carboxylic acids. The preferred olefin-acting compounds are olefinic hydrocarbons which comprise monoolefins containing one double bond per molecule and polyolefins which contain more than one double bond per molecule. Monoolefins which are utilized as olefin-acting compounds in the process of the present invention are either normally gaseous or normally liquid and include ethylene, propylene, 1-butene, 2-butene, isobutylene, and higher molecular weight normally liquid olefins such as the various pentenes, hexenes, heptenes, octenes, and mixtures thereof, and still higher molecular weight liquid olefins, the latter including various olefin polymers having from about 9 to about 18 carbon atoms per molecule including propylene trimer, propylene tetramer, propylene pentamer, etc. Cycloolefins such as cyclopentene, methylcyclopentene, cyclohexene, methylcyclohexene, etc., may also be utilized. Also included within the scope of the olefin-acting compound are certain substances capable of producing olefinic hydrocarbons or intermediates thereof under the conditions of operation utilized in the process. Typical olefin-producing substances or olefin-acting compounds capable of use include alkyl halides capable of undergoing dehydrohalogenation to form olefinic hydrocarbons and thus containing at least two carbon atoms per molecule. Examples of such alkyl halides include ethyl fluoride, n-propyl fluoride, isopropyl fluoride, n-butyl fluoride, isobutyl fluoride, sec-butyl fluoride, tert-butyl fluoride, etc., ethyl chloride, n-propyl chloride, isopropyl chloride, n-butyl chloride, isobutyl chloride, sec-butyl chloride, tert-butyl chloride, etc., ethyl bromide, n-propyl bromide, isopropyl bromide, n-butyl bromide, isobutyl bromide, sec-butyl bromide, tert-butyl bromide, etc. As stated hereinabove, other esters such as alkyl sulfates including ethyl sulfate, propyl sulfate, etc., and alkyl phosphates including ethyl phosphate, etc., may be utilized. Ethers such as diethyl ether, ethyl propyl ether, dipropyl ether, etc., are also included within the generally broad scope of the term olefin-acting compound and may be successfully utilized as alkylating agents in the process of this invention.

Olefin hydrocarbons, particularly normally-gaseous hydrocarbons, are olefin-acting compounds for the use in the process of this invention and for passage by means of lines 4 and 8 to reaction zone 9. The process of this invention may be successfully applied to and utilized for complete conversion of olefin hydrocarbons when these olefin hydrocarbons are present in minor quantities in various gas streams. Thus, in contrast to prior art processes, the normally gaseous olefin for use in the process of this invention need not be concentrated. Such normally gaseous olefin hydrocarbons appear in minor quantities in various refinery gas streams, usually diluted with gases such as hydrogen, nitrogen, methane, ethane, propane, etc. These gas streams containing minor quantities of olefin hydrocarbons are obtained in petroleum refineries from various refinery installations including thermal cracking units, catalytic cracking units, thermal reforming units, coking units, polymerization units, dehydrogenation units, etc. Such refinery gas streams have in the past often been burned for fuel value, since an economical process for the utilization of their olefin hydrocarbon content has not been available, or processes which have been suggested by the prior art utilize such large quantities of alkylatable aromatic compound that they have not been economically feasible. This is particularly true for refinery gas streams known as off-gas streams containing relatively minor quantities of olefin hydrocarbons such as ethylene. Thus, it has been possible to catalytically polymerize propylene and/or butenes in various refinery gas streams, but the off-gases from such processes still contain the utilizable olefin hydrocarbon, ethylene. In addition to containing ethylene in minor quantities, these off-gas streams contain other olefin hydrocarbons, depending upon their source, including propylene and butenes. A refinery off-gas ethylene stream may contain varying quantities of hydrogen, nitrogen, methane and ethane with the ethylene in minor proportion, while the refinery off-gas propylene stream is normally diluted with propane and contains the propylene in minor quantity, and a refinery off-gas butene stream is normally diluted with butanes and contains the butenes in minor quantities. A typical analysis in mol percent for utilizable refinery off-gas from a catalytic cracking unit is as follows: nitrogen, 4.0%; carbon monoxide, 0.2%; hydrogen, 5.4%; methane, 37.8%; ethylene, 10.3%; ethane, 24.7%; propylene, 6.4%; propane, 10.7% and C_4 hydrocarbons, 0.5%. It is readily observed that the total olefin content of this gas stream is 16.7 mol percent and the ethylene content is even lower, namely 10.3 percent. Such gas streams containing olefin hydrocarbons in minor or dilute quantities are particularly preferred alkylating agents within the broad scope of this invention. It is readily apparent that only the olefin content of such streams undergoes reaction at alkylation conditions in the process, and that the remaining gases free from olefin hydrocarbons are vented from the process. It is one of the features of this invention that the gases which do not react are vented from the process with minimum loss of boron trifluoride and alkylatable aromatic compounds due to their vapor pressure at the conditions of temperature and pressure utilized for venting the non-reactive gases.

The olefin-acting compound, acting as the alkylating agent, has combined therewith in line 8 alkylatable aromatic compound from line 2 with boron trifluoride combined therewith from line 1 as will be set forth hereinafter. Many aromatic compounds are utilizable as alkylatable aromatic compounds within the process of this invention. The preferred aromatic compounds are aromatic hydrocarbons, and the preferred aromatic hydrocarbons are monocyclic aromatic hydrocarbons, that is, benzene hydrocarbons. Suitable aromatic hydrocarbons include benzene, toluene, ortho-xylene, meta-xylene, para-xylene, ethylbenzene, ortho-ethyltoluene, meta-ethyltoluene, paraethyltoluene, 1,2,3-trimethylbenzene, 1,2,4-trimethylbenzene, 1,3,5 - trimethylbenzene, normal propylbenzene, isopropylbenzene or cumene, normal butylbenzene, etc. Higher molecular weight alkylaromatic hydrocarbons are also suitable as starting materials and include aromatic hydrocarbons such as are produced by the alkylation of the aromatic hydrocarbons with olefin polymers. Such products are referred to in the art as alkylate and include hexylbenzene, hexyltoluene, nonylbenzene, nonyltoluene, dodecylbenzene, dodecyltoluene, pentadecylbenzene, pentadecyltoluene, etc. Other suitable alkylatable aromatic hydrocarbons include those with two or more aryl groups such as diphenyl, diphenylmethane, triphenylmethane, fluorene, stilbene, etc. Examples of alkylatable aromatic hydrocarbons within the scope of this invention utilizable as starting materials and con-

taining condensed benzene rings include naphthalene, alpha-methylnaphthalene, beta-methylnaphthalene, etc., anthracene, phenanthrene, naphthacene, rubrene, etc. When the selected alkylated aromatic hydrocarbon is a solid, it may be heated by means not shown so that it passes as a liquid through line 2 or 8 as hereinafter described. Of the alkylatable aromatic hydrocarbons for use as starting materials in the process of this invention, the benzene hydrocarbons are preferred, and of the benzene hydrocarbons, benzene itself is particularly preferred.

As stated hereinabove, boron trifluoride is added in admixture with the alkylatable aromatic compound prior to passage thereof to line 6. This is conveniently accomplished by passage of boron trifluoride from line 1 through line 6. Boron trifluoride is a gas, boiling point −101° C., melting point −126° C., and is somewhat soluble in most organic solvents. It may be and generally is utilized per se by mere passage thereof as a gas through lines 1 and 6 so that it dissolves at least partially in the alkylatable aromatic compound passing concurrently therewith through line 6. The boron trifluoride may also be added as a solution of the gas in a suitable organic solvent. However, in the utilization of such solutions, care must be exercised so that the selected solvent is unreactive with the alkylating agent or normally gaseous olefin hydrocarbon utilized in the process. Furthermore, boron trifluoride complexes with many many organic compounds, particularly those containing sulfur or oxygen atoms. These complexes, while utilizable as catalysts, are very stable and thus will interfere with the recovery of boron trifluoride in the gas-liquid absorption zone hereinafter set forth. Therefore, a further limitation upon the selection of such a solvent is that it be free from atoms or groups which from complexes with boron trifluoride. The amount of boron trifluoride which is utilized is relatively small. It has been found that the amount necessary can be conveniently expressed as grams of boron trifluoride per gram mol of olefin-acting compound, preferably olefin. This amount of boron trifluoride will contain not more than 1.0 gram of boron trifluoride per gram mol of olefin utilized. When the amount of boron trifluoride present in the alkylation zone is within the above expressed limit, substantially complete conversion of the olefin-acting compound is obtained even when the olefin-acting compound is present in what might seem to be minor or dilute quantities in the gas stream. Furthermore, a portion of boron trifluoride then carries over from the alkylation reaction zone to the transalkylation reaction zone as hereinafter described wherein that amount will be utilized again, or in combination with further added boron trifluoride, to cause the transalkylation reaction to go forward, thus, double use of the originally added quantity of boron trifluoride is obtained in this process.

Prior to passage to the alkylation zone, the fresh aromatic compound, olefin-acting compound, and boron tri-fluoride have combined therewith recycled fractionated unreacted aromatic compound via line 36 and recycle flashed unreacted aromatic compound containing boron trifluoride via line 22 as hereinafter set forth. Recycle unreacted aromatic compound is available in the process since it is preferred to utilize a molar excess of alkylatable aromatic compound over olefin-acting compound, preferably olefin. This, as is disclosed in the prior art, has been found necessary to prevent side reactions from taking place such as, for example, polymerization of the olefin-acting compound prior to reaction thereof with the alkylatable aromatic compound and to direct the reaction principally to monoalkylation. Any molar excess of alkylatable aromatic compound may be utilized, although best results are obtained when the alkylatable aromatic compound to olefin-acting compound molar ratio is from about 4:1 to about 20:1 or more.

The combined feed to the alkylation reaction zone comprising alkylatable aromatic compound in a molar excess based on the alkylating agent, and boron trifluoride provided in a manner hereinabove specified is passed to reactor 9 containing the alkylation zone. Reactor 9 is of the conventional type with a boron trifluoride-modified inorganic oxide disposed therein in the reaction zone. In addition, the reactor may be equipped with heat transfer means, baffles, trays, heating means, etc. The reactor is preferably of the adiabatic type and thus the feed to the reactor will preferably be provided with the requisite amount of heat prior to passage thereof to said reactor. As set forth hereinabove, the alkylation reaction zone is packed with a boron trifluoride-modified inorganic oxide. The inorganic oxide with which the zone in the reactor is packed may be selected from among diverse inorganic oxides including alumina, silica, boria, oxides of phosphorus, titanium dioxide, zirconium dioxide, chromia, zinc oxide, magnesia, calcium oxide, silica-alumina, silica-magnesia, silica-alumina-magnesia, silica-alumina-zirconia, chromia-alumina, alumina-boria, silica-zirconia, etc., and various naturally occurring inorganic oxides of various states of purity such as bauxite, clay (which may or may not have been previously acid treated), diatomaceous earth, etc. Of the above-mentioned inorganic oxides, gamma-alumina and theta-alumina are most readily modified by boron trifluoride, and thus the use of one or both of these boron trifluoride-modified aluminas is preferred. The modification of the inorganic oxide, particularly alumina, may be carried out prior to or simultaneous with the passage of the reactants containing boron trifluoride to the reactor. The exact manner in which the inorganic oxides are modified by boron trifluoride is not completely understood. However, it has been found that the modification is preferably carried out at a temperature at least as high as that selected for use in the particular zone, so that the catalyst in said zone will not exhibit an activity induction period. If the inoragnic oxide is modified prior to use, this modification may be carried out in situ in the reactor or in a separate catalyst preparation step. More simply, this modification is accomplished by mere passage of boron trifluoride gas over a bed of the inorganic oxide maintained at the desired temperature. If the modification of the inorganic oxide with boron trifluoride is carried out during the passage of the reactants thereover, the catalyst will exhibit an induction period and thus complete reaction of the alkylating agent with the alkylatable aromatic compound, and transalkylation of the recycled polyalkylated aromatic compounds will not take place for some hours, say up to 12 or more.

The conditions utilized in reaction zone 9 may be varied over a relatively wide range. Thus, the desired alkylation reaction in the presence of the above indicated catalyst may be effected at a temperature of from about 0° or lower to about 250° C. or higher. The alkylation reaction is usually carried out at a pressure of from about substantially atmospheric, preferably from about 15 to about 200 atmospheres or more. The pressure utilized is usually selected to maintain the alkylatable aromatic compound in substantially liquid phase which is readily accomplished when utilizing relatively pure, concentrated components. However, within the above-mentioned temperature and pressure ranges, it is not always possible to maintain the olefin-acting compound in completely liquid phase. Thus, when utilizing a refinery off-gas containing ethylene as the olefin-acting compound, the dilute ethylene stream will be dissolved in the liquid phase alkylatable aromatic compound (and alkylated aromatic compound as formed) to the extent governed by temperature, pressure, and solubility considerations. However, a small portion thereof will always be in the vapor phase. The hourly liquid space velocity of the liquid through the alkylation zone may be varied over relatively wide range of from about 0.1 to about 20 or more.

When the alkylation reaction has proceeded to the de-

sired extent, preferably with 100% conversion of the olefin-acting compound, the products from the alkylation zone which may be termed alkylation zone effluent, pass from alkylation reaction zone 9 via line 10 through pressure control valve 11 to line 12, where the effluent is cooled and condensed in condenser 13 from which the gases and liquids pass to flash drum 14. Pressure control valve 11 generally is set so that a substantial pressure drop, preferably of about 200 p.s.i.g., occurs from line 10 to line 12. This accomplishes the purpose of allowing condensation of the heavier components of the effluent while still allowing a flashing of the gases and a portion of the alkylatable aromatic compound from flash drum 14 through line 15. In line 15 there is another cooler or condenser 16 which is maintained at a lower temperature than condenser 13 and further accomplishes the condensation of uncondensed alkylatable aromatic compound which passes through line 15 along with the gases from flash drum 14. By the utilization of the low pressure flash drum separation, all of the products in the effluent which are gaseous at atmospheric conditions can be separated in separator 17 and passed therefrom through line 18 to countercurrent gas-liquid absorber 19, hereinafter described. The condensable materials in separator 17, comprising mainly alkylatable aromatic compounds are withdrawn therefrom from line 20 by pump 21 which passes the same via line 22 for recycle to the alkylation zone. The flash drum also frees the liquid effluent therein from gaseous components in the effluent and thus a gas-free liquid phase is withdrawn from flash drum 14 through line 23 by pump 24 which passes the same via line 25 to a commingling step. The thereinabove mentioned gas-free liquid phase is then passed from the commingling step, hereinafter described, though line 26 to fractionation zone 27, labeled benzene column.

Fractionation zone 27 is a conventional fractionator distillation column or tower and utilized for the purpose of recovering excess unreacted alkylatable aromatic compound from the alkylation reaction zone effluents for recycle. The recovered unreacted alkylatable aromatic compound passes overhead from fractionation zone 27 through line 28 containing condenser 29 to overhead receiver 30. The unreacted alkylatable aromatic compound recovered overhead from fractionation zone 27 is withdrawn from overhead receiver 30 through line 31 by pump 32 which provides reflux to fractionation zone 27 by means of lines 33 and 34 and which also recycles the remainder or net amount of the recovered alkylatable aromatic compound via lines 33 and 35 to line 36 which recycles the same to the alkylation reaction zone 9. The higher boiling alkylated aromatic compounds are withdrawn from fractionation zone 27 through line 40 and passed to fractionation zone 41, labeled ethylbenzene column. The requisite amount of heat is furnished to fractionation zone 27 by reboiler 39 which heats and recycles a portion of the high boiling alkylated aromatic compounds back to fractionation zone 27 through line 38.

Second fractionation zone 41 is of the conventional type and is utilized for recovering of desired alkylated aromatic compound from higher boiling homologs thereof. The desired alkylated aromatic compound is withdrawn overhead from fractionation zone 41 through line 42 containing condenser 43 and is passed to overhead receiver 44. The liquid product from overhead receiver 44 comprises desired alkylated aromatic compound which is withdrawn therefrom through line 45 by pump 46 which provides reflux to fractionation zone 41 by means of lines 47 and 48. Pump 46 also provides a means for passage of the desired alkylated aromatic compound from the process by means of line 49, labeled product to storage. The still higher boiling polyalkylated aromatic compounds are withdrawn from fractionation zone 41 by means of line 52. The requisite amount of heat is provided to fractionation zone 41 by reboiler 51 through which a portion of the higher boiling polyalkylated aromatic compounds is passed by means of line 50.

The higher boiling polyalkylated aromatic compounds may be passed directly from fractionation zone 41 for use as absorber oil in gas-liquid absorption zone 19, or they may be passed to further fractionation as hereinafter described. When direct passage of these higher boiling polyalkylated aromatic compounds to the absorption zone is desired, control valve 58 is opened and control valve 54 is closed. The higher boiling polyalkylated aromatic compounds pass from fractionation zone 41 via lines 52 and 57 through control valve 58 through line 59 into an upper region of gas-liquid absorption zone 19.

Absorption zone 19, labeled absorber, is a countercurrent contacting zone, of conventional design, the size of which is varied depending upon the quantity of polyalkylated aromatic compounds passed thereto and upon the quantity of unreacted aromatic compound, boron trifluoride, and unreacted gases passed to a lower region thereof. In gas-liquid absorber 19, the higher boiling polyalkylated aromatic compounds flowing through line 59 flow downward in a countercurrent manner to the ascending gases which are introduced thereto in a lower region thereof, for example, via line 18. The unreactive gases are vented from absorption zone 19 through line 61 containing valve 60. The polyalkylated aromatic compound and at least a portion of the boron trifluoride and any alkylatable aromatic compound which may have been passed thereto from separator 17 are withdrawn from the bottom of absorption zone 19 through line 62 and passed to transalkylation zone 81 via lines 63 and 96 as hereinafter described.

When direct passage of these higher boiling polyalkylated aromatic compounds to the next fractionation zone is desired, pressure control valve 58 is closed and pressure control valve 54 is opened. The higher boiling polyalkylated aromatic compounds pass from fractionation zone 41 via lines 52 and 53 through pressure control valve 54 through line 55 to fractionation zone 56, labeled polyethylbenzene column. Fractionation zone 56 is of the conventional type and the polyalkylated aromatic compounds are fractionated therein to remove the desired recycle as overhead therefrom. The desired polyalkylated aromatic compound recycle passes overhead from column 65 through line 64 containing condenser 65 to overhead receiver 66. The liquid is withdrawn from overhead receiver 66 through line 67 by pump 68 which provides reflux for fractionation zone 56 by means of lines 69 and 70. The net amount of polyalkylated aromatic compound is passed by pump 68 through line 69 and 71 through pressure control valve 72 to line 76 and then to transalkylation zone 81 are hereinafter described. The column is supplied with the necessary amount of heat by reboiler 74 which recycles a portion of the higher boiling bottoms back to the column through line 73. A portion of the higher boiling bottoms may be withdrawn through line 75.

The polyalkylated aromatic compound from overhead receiver 66, as hereinbefore described, is passed to transalkylation zone 81 via line 76. However, prior to passage of the polyalkylated aromatic compound to transalkylation zone 81, these compounds have combined therewith unreacted fractionated alkylatable aromatic compound via lines 37 and 63, and with unreacted flashed alkylatable aromatic compound via lines 95 and 96 as hereinafter described to provide a molar excess thereof in relation to the alkyl groups contained in the polyalkylated aromatic compounds passed to said transalkylation zone. Furthermore, a quantity of boron trifluoride in the amount of 0.002 gram to about 1.2 grams of boron trifluoride per gram mol of polyalkylated aromatic compound is added to the transalkylation zone via lines 62 and 63 from gas-liquid absorption zone 19 as hereinbefore described as well as from line 95. If necessary, and/or desirable an additional amount of boron trifluo-

ride may be added through line 77 containing pressure control valve 78 via line 76. Thus, a transalkylation reaction zone combined feed of polyalkylated aromatic compounds, alkylatable aromatic compound and boron trifluoride passes through mixing line 76 through heater 79 via line 80 to transalkylation reaction zone 81.

Transalkylation reaction zone 81 is of the conventional type and may be equipped with heat transfer means, baffles, trays, heating means, etc. The reactor is preferably of the adiabatic type and thus the feed to the reactor will preferably be provided with the requisite amount of heat prior to passage thereof to said reactor. As set forth hereinabove, the reaction zone is packed with a boron trifluoride-modified inorganic oxide. The particular boron trifluoride-modified inorganic oxide is generally selected so that the same material is utilized in both the alkylation reaction zone and the transalkylation reaction zone. Since the conditions necessary for transalkylation are generally more severe than for alkylation, one effective means for increasing severity is by utilization of a bed of boron trifluoride-modified inorganic oxide in transalkylation zone 81 of greater depth than was utilized as in the alkylation zone 9. By the utilization of such greater bed depth, one effectively decreases the liquid hourly space velocity of the combined feed therethrough and thus increases reaction zone severity. As was the case with the conditions utilized in the alkylation reaction zone, the conditions utilized in transalkylation reaction zone 81 may be varied over a relatively wide range, but, as set forth hereinabove, are usually of greater severity than prevail in the alkylation reaction zone. Various means other than increasing catalyst bed depth and decreasing liquid hourly space velocity may be utilized for increasing this reaction zone severity. For example, the mol concentration of boron trifluoride in transalkylation zone 81 may be greater than for alkylation zone 9 by passage of additional boron trifluoride thereto via line 77. Also, when the alkylation reaction zone and transalkylation reaction zone are separated as shown in the drawing, one may effectively increase the temperature by proper placement of heating means before each reactor. The transalkylation reaction may be effected at temperatures of from about 50° to about 300° C. or higher and at a pressure of from about substantially atmospheric, preferably from about 15 to about 200 atmospheres. The pressure utilized is selected to maintain the alkylatable aromatic compound and polyalkylated aromatic compounds in liquid phase. Referring to the alkylatable aromatic compound, it is preferable to have present in the transalkylation reaction zone from about 1 to about 10 or more, sometimes up to 20, molar proportions per molar proportion of alkyl group in the polyalkylated aromatic hydrocarbon introduced therewith. The hourly liquid space velocity of the liquid through transalkylation reaction zone 81 may be varied over a relatively wide range of from about 0.1 to about 20 or more. It is a feature of the present invention that the alkylatable aromatic compound to polyalkylated aromatic compound ratio in the transalkylation reaction zone can be varied independently of the alkylation reactor rates. When the transalkylation reaction has proceeded to the desired extent so that a sufficient quantity of polyalkylated aromatic compounds are converted to monoalkylated aromatic compounds by reaction with alkylatable aromatic compound, the products from transalkylation zone 81 are withdrawn through line 82 for recovery of the desired components therefrom.

The transalkylation reaction zone effluent passes through line 82 through line 84 containing pressure control valve 83 where the effluent is cooled and condensed in condenser 85 from which the gases and liquids pass to flash drum 86. Pressure control valve 83 generally is set so that a substantial pressure drop, preferably of about 200 p.s.i.g., occurs from line 82 to line 84. This accomplishes the purpose of allowing condensation of the heavier components of the effluent while still allowing the flashing of the gases and a portion of the alkylatable aromatic compound from flash drum 86 through line 87. In line 87, there is another cooler or condenser 88 which is maintained at a lower temperature than condenser 85 and further accomplishes the condensation of uncondensed alkylatable aromatic compound which passes through line 87 along with the gases from flash drum 86. The flash drum also frees the liquid effluent therein from gaseous components in the effluent and thus a gas-free liquid phase is withdrawn from flash drum 86 through line 90 by pump 91 which passes the same via line 92 to a commingling step as hereinbefore mentioned.

By the utilization of the low-pressure flash drum separation, the condensible materials in separator 89, comprising mainly alkylatable aromatic compound in admixture with boron trifluoride, are withdrawn therefrom via line 93 by pump 94 to line 95 for recycle to the transalkylation reaction zone via line 96 as hereinabove set forth.

The commingling step comprises withdrawing the gas-free liquid phase from alkylation reaction zone flash drum 14 via line 25 and withdrawing the gas-free liquid phase from transalkylation reaction zone flash drum 86 via line 92 and passing the resultant gas-free liquid phase mixture via line 26 to fractionation zone 27. By the utilization of the commingling step, the unreacted aromatic compound, monoalkylated aromatic compound, and polyalkylated aromatic compounds are fed directly to the fractionators for separation into the desired components as hereinabove described.

The following example is introduced for the purpose of illustration with no intention of unduly limiting the generally broad scope of the present invention. This example is carried out in a bench scale pilot plant equipped with two separate reactors, gas-liquid separation means, fractionation means for separation and recycle of excess alkylatable aromatic compound, fractionation means for separation and recovery of monoalkylated aromatic compound, and recycle means for polyalkylated aromatic compound. The reactors are equipped with separate heating means so that the temperature in each can be maintained at different levels. The alkylation reactor is of sufficient size so that a bed of approximately 175 cc. of boron trifluoride-modified gamma-alumina could be utilized therein. The transalkylation reactor is of sufficient size so that a bed of approximately 200 cc. of boron trifluoride-modified gamma-alumina could be utilized therein.

The boron trifluoride-modified gamma-alumina utilized is prepared by treating gamma- alumina with a mixture of boron trifluoride and nitrogen at a temperature of approximately 300° F. After loading the thus modified alumina into the separate reactors, its temperature is raised to 300° C. and it is again treated with 22% boron trifluoride and nitrogen to insure modification thereof. The feed stocks utilized consisted of benzene, a synthetic off-gas consisting of about 11% ethylene in nitrogen, and polyethylated benzene hydrocarbons produced in the process.

This example illustrates the process of the present invention for the production of ethylbenzene utilizing the flow scheme as shown in the drawing. In this example, benzene and ethylene are fed to the alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous gamma-alumina along with a catalytic amount of boron trifluoride. The gas-free liquid phase that is withdrawn and separated from the alkylation zone is commingled with the gas-free liquid phase that is withdrawn and separated from the transalkylation zone and from the commingling step, unreacted benzene, desired ethylbenzene, and higher molecular weight polyethylbenzenes are separated. A portion of the unreacted benzene is recycled to the alkylation zone, and the desired ethylbenzene is removed as product from the process.

The polyethylbenzenes are passed in admixtures with unreacted benzene and boron trifluoride to the transalkylation reaction zone also containing a boron trifluoride-modified substantially anhydrous gamma-alumina and therein reacts the polyethylbenzenes with the benzene, and a gas-free liquid phase is withdrawn and separated from the transalkylation zone and recycled to the commingling step as hereinabove set forth.

The test is carried out at pressure of 530 p.s.ig. for both reactors with a maximum temperature of 235° C. in the alkylation reaction zone and a maximum temperature of 258° C. in the transalkylation reaction zone. In the test, 525 cc. per hour of benzene and 28.0 grams per hour of ethylene in the form of a synthetic off-gas is passed to the alkylation reactor. Along with this feed, there is also passed to this reactor 0.05 gram per hour of boron trifluoride. The benzene to olefin mol ratio in the alkylation reactor is kept at about 6:1. The effluent from the alkylation reactor is flashed by a reduction in pressure prior to gas-liquid separation, and a total of 223 grams of unreacted benzene is recycled back to the reactor while a total of 180 grams of unreatced benzene, ethylbenzene, and polyethylbenzenes pass to the fractionation zones for separation into the desired components. All of the gaseous products are passed to the absorber where the polyethylbenzenes and the boron trifluoride and any unreacted benzene passed thereto is passed to the transalkylation zone. A total of approximately 125 cc. of benzene and 25 grams of polyethylbenzenes is passed to the transalkylation reaction zone so that a benzene to ethyl group ratio of about 2 is maintained. In addition, there is added an additional amount of boron trifluoride so that the total quantity of boron trifluoride passing to the transalkylation reaction zone is maintained at approximately 0.20 gram per hour. The effluent from the transalkylation reaction zone is flashed by a reduction in pressure prior to gas-liquid separation and a total of 85 grams of the gas-free liquid phase is passed to the commingling step where the gas-free liquid phases from both reactors join and then pass as a gas-free liquid phase mixture to the fractionation zones for separation into the desired components.

In this twenty-four hour test period illustrating the process of the present invention, ethylene conversion starts out at 100% and continues at 100% throughout the entire test. During the test, complete transalkylation of the polyethylbenzenes continues so that ethylbenzenes yields based on benzene reacted approach the stoichiometric yields and continue at this high level. Simultaneously, no catalyst deactivation is observed for the entire test period.

I claim as my invention:

1. A process for the production of an alkylaromatic compound which comprises alklating an alkylatable aromatic compound with an olefin-acting compound in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous inorganic oxide, flashing the effluent from said alkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, commingling said gas-free liquid phase with a gas-free liquid phase from a transalkylation zone as hereinafter set forth, fractionating the resultant gas-free liquid phase mixture and separating therefrom unreacted aromatic compound, desired monoalkylated aromatic compound, and higher molecular weight polyalkylated aromatic compound, recycling at least a portion of said unreacted aromatic compound to the alkylation zone, removing desired monoalkylated aromatic compound as product from the process, passing said polyalkylated aromatic compound in admixture with alkylatable aromatic compound and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous inorganic oxide and therein reacting the polyalkylated aromatic compound with the alkylatable aromatic compound, flashing the effluent from said transalkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, and passing the last-named gas-free liquid phase to said commingling step as aforesaid.

2. The process of claim 1 further characterized in that said alkylatable aromatic compound is an alkylatable aromatic hydrocarbon.

3. The process of claim 1 further characterized in that said alkylatable aromatic compound is a benzene hydrocarbon.

4. The process of claim 3 further characterized in that said transalkylation reaction zone contains from about 0.0002 to about 1.2 grams of boron trifluoride per gram mol of polyalkylated aromatic compound and in that said alkylation reaction zone contains not more than 1.0 gram of boron trifluoride per gram mol of olefin-acting compound.

5. The process of claim 4 further characterized in that said olefin-acting compound is an olefinic hydrocarbon.

6. The process of claim 4 further characterized in that said olefin-acting compound is a normally gaseous olefin.

7. The process of claim 6 further characterized in that said inorganic oxide is a substantially anhydrous alumina.

8. The process of claim 6 further characterized in that said substantially anhydrous inorganic oxide is gamma-alumina.

9. The process of claim 6 further characterized in that said substantially anhydrous inorganic oxide is theta-alumina.

10. A process for the production of ethylbenzene which comprises alkylating benzene with ethylene in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous alumina, flashing the effluent from said alkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, commingling said gas-free liquid phase with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, fractionating the resultant gas-free phase mixture and separating therefrom unreacted benzene, desired ethylbenzene, and higher molecular weight polyethylbenzenes, recycling at least a portion of said unreacted benzene to the alkylation zone, removing desired ethylbenzene as product from the process, passing said polyethylbenzenes in admixture with benzene and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous alumina and therein reacting the polyethylbenzenes with the benzene, flashing the effluent from said transalkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, and passing the last-named gas-free liquid phase to said commingling step as aforesaid.

11. A process for the production of cumene which comprises alkylating benzene with propylene in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous gamma-alumina, flashing the effluent from said alkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, commingling said gas-free liquid phase with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, fractionating the resultant gas-free liquid phase mixture and separating therefrom unreacted benzene, desired cumene, and higher molecular weight polypropylbenzenes, recycling at least a portion of said unreacted benzene to the alkylation zone, removing desired cumene as product from the process, passing said polypropylbenzenes in admixture with benzene and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous gamma-alumina and therein reacting the polypropylbenzenes with the benzene, flashing the effluent

from said transalkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, and passing the last-named gas-free liquid phase to said commingling step as aforesaid.

12. A process for the production of butylbenzene which comprises alkylating benzene with a butene in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous alumina, flashing the effluent from said alkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, commingling said gas-free liquid phase with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth, fractionating the resultant gas-free liquid phase mixture and separating therefrom unreacted benzene, desired butylbenzene, and higher molecular weight polybutylbenzenes, recycling at least a portion of said unreacted benzene to the alkylation zone, removing desired butylbenzene as product from the process, passing said polybutylbenzenes in admixture with benzene and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous alumina and therein reacting the polybutylbenzenes with the benzene, flashing the effluent from said transalkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, and passing the last-named gas-free liquid phase to said commingling step as aforesaid.

13. A process for the production of ethylbenzene which comprises alkylating benzene with a refinery off-gas containing a minor quantity of ethylene in the presence of a catalytic amount of boron trifluoride in an alkylation reaction zone containing a boron trifluoride-modified substantially anhydrous alumina, flashing the effluent from said alkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, commingling said gas-free liquid phase with a gas-free liquid phase from a transalkylation reaction zone as hereinafter set forth fractionating the resultant gas-free liquid phase mixture and separating therefrom unreacted benzene, desired ethylbenzene, and higher molecular weight polyethylbenzenes, recycling at least a portion of said unreacted benzene to the alkylation zone, removing desired ethylbenzene as product from the process, passing said polyethylbenzenes in admixture with benzene and boron trifluoride to a transalkylation zone containing boron trifluoride-modified substantially anhydrous alumina and therein reacting the polyethylbenzenes with the benzene, flashing the effluent from said transalkylation zone by pressure reduction and then separating the same into a gas phase and a gas-free liquid phase, and passing the last-named gas-free liquid phase to said commingling step as aforesaid.

14. The process of claim 1 further characterized in that the alkylation conditions are a temperature of from about 0° to about 250° C., a pressure of from about atmospheric to about 200 atmospheres, and a liquid hourly space velocity of from about 0.1 to about 20.

15. The process of claim 1 further characterized in that the transalkylation conditions are a temperature of from about 50° to 300° C., a pressure of from about atmospheric to about 200 atmospheres, and a liquid hourly space velocity of from about 0.1 to about 20.

16. The process of claim 1 further characterized in that the alkylation and transalkylation zones are confined within separate reaction vessels.

17. The process of claim 1 further characterized in that the quantity of boron trifluoride present in the alkylation zone is less than that present in the transalkylation zone.

18. The process of claim 1 further characterized in that the boron trifluoride furnished to the transalkylation zone is supplied solely from the alkylation zone.

19. The process of claim 1 further characterized in that both the alkylation zone and transalkylation zone effluents are flashed by a reduction in pressure of about 200 pounds per square inch.

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