

Nov. 29, 1966

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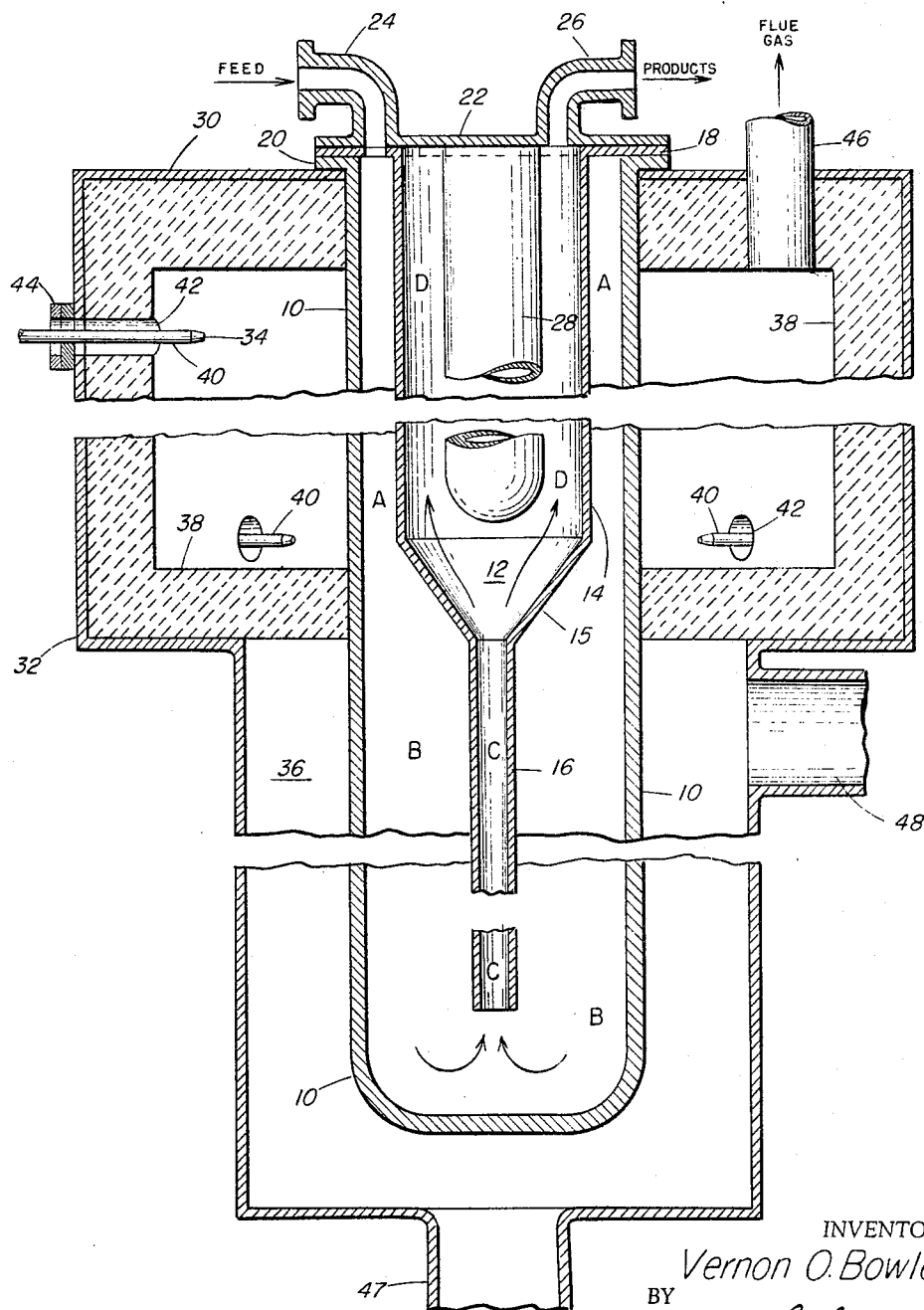
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APPARATUS AND METHOD FOR HIGH TEMPERATURE REACTIONS

Filed June 27, 1963

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FIG. 1



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2 Sheets-Sheet 2

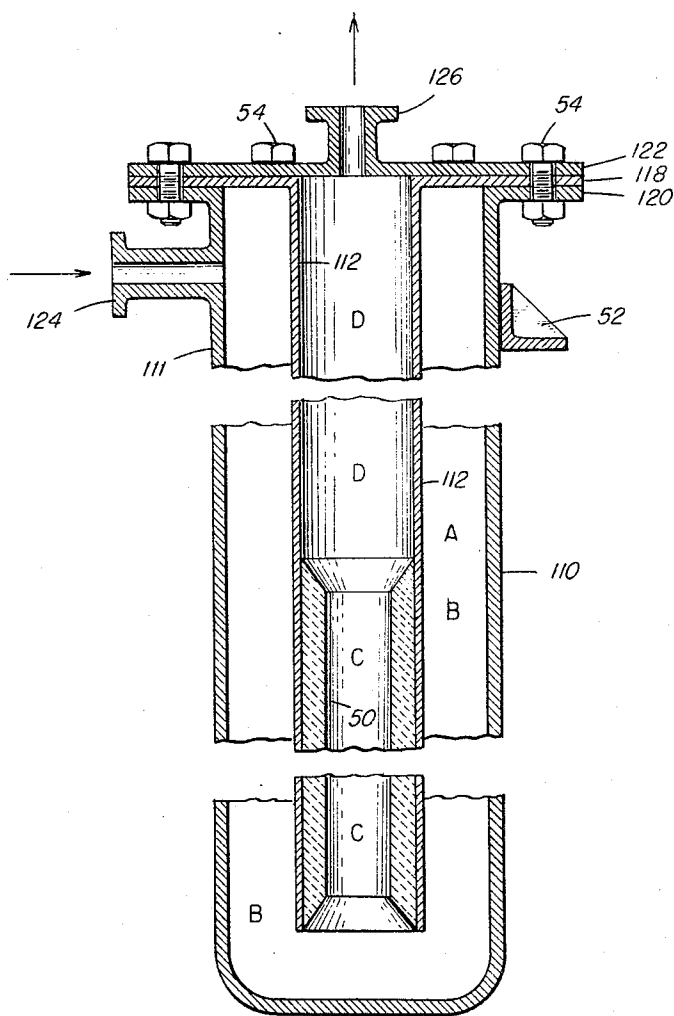


FIG. 2

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3,288,874

APPARATUS AND METHOD FOR HIGH
TEMPERATURE REACTIONS

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Filed June 27, 1963, Ser. No. 291,077
20 Claims. (Cl. 260-672)

This invention relates to controlling high temperature conversions involving fluid reactants as well as apparatus for such control. The invention has especial application to high temperature reactions of materials in the gaseous phase and particularly to the thermal hydrodealkylation of alkyl aromatic compounds.

The dealkylation of alkyl aromatic hydrocarbons with hydrogen in the presence of a catalyst is now being carried out on a commercial scale and proposals have been made for conducting such reactions without a catalyst at high temperatures and high pressures. However, a number of practical operating difficulties have hampered the development of a noncatalytic process. The reaction temperature of about 1300-1400° F. would ordinarily be expected to cause some trouble as it is relatively close to the operating limits of many materials of construction, but these difficulties are greatly compounded in dealkylation reactions due to other prevailing conditions. Moreover, the high pressures in the 600-700 p.s.i.g. range create some problems at commonly employed temperatures and these are aggravated by the high reaction temperature characteristic of the thermal dealkylation process. Such problems can sometimes be overcome at considerable expense by using high alloy steels. Furthermore, there is an abundance of hydrogen present and it is well known that this gas attacks many steels, including alloy steels. Methane and other light hydrocarbons are also present in substantial amounts as by-products of the reaction and frequently as components of the hydrogen-rich gas supplied as charge and these materials can under the conditions prevalent have a carburizing or case hardening effect on ferrous metals and alloys with a consequent loss in strength and ductility. It will be readily appreciated that carburizing and hydrogen attack add to the already substantial problem of withstanding the high thermal stresses and high pressures and particularly the large expansion and contraction forces encountered in starting up and shutting down the equipment. To complicate matters even more, the exothermic dealkylation reactions have a tendency to build up excessive reaction temperatures usually with the undesirable deposition of coke in the equipment. Finally, it has not been commercially feasible to dealkylate certain undesirable feed stocks. For example, in the production of naphthalene from its alkylated precursors, feed stocks with end points above 525° F. that contain in excess of a total of about 10 percent by weight or normally liquid paraffins, naphthenes and olefins have been avoided because satisfactorily high yields could not be obtained without excessive coking.

In the highly exothermic production of benzene from toluene and hydrogen, the overall heat release amounts to about 22,500 B.t.u. per mol of toluene converted, which is sufficient in theory to raise the reaction temperature under stoichiometric conditions about 360° F. A heat level of at least 1100° F., and preferably at least 1200° F., is necessary to provide a reaction rate fast enough for commercial purposes but temperatures above about 1400° F. are unacceptable as the benzene product begins to crack and hydrogenation of the fragments releases additional quantities of heat that result in excessive temperatures and the rapid and undesirable deposition of coke in the equipment. Normally liquid nonaromatic hydrocarbons to

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further complicate the problem when present in any substantial quantity, by cracking in the reactor at lower temperatures and then releasing a great deal of heat during the hydrogenation of their fragments. To alleviate these undesirable conditions, it has been found desirable in the operation of an adiabatic reactor to pass through the reactor a large excess of hydrogen over that required by stoichiometry and also inert diluent gases. This is readily accomplished by introducing a combination of recycle gas containing a mixture of lower hydrocarbons and hydrogen and a hydrogen-rich make-up gas. However, even with this large volume of gases to serve as a heat reservoir, it is easy for excessive temperatures to occur in certain locations in such a reactor.

An object of the invention is to provide improved control of reaction temperatures and reaction rates in gaseous phase reactions.

Another object of the invention is to prevent excessive reaction temperatures in exothermic gaseous phase reactions.

A further object of the invention is to provide more uniform temperatures and rates in gaseous phase reactions.

Still another object of the invention is to provide improved temperature control in the dealkylation of alkyl aromatic hydrocarbons.

A still further object of the invention is to provide a method for dealkylating alkyl naphthalenes without substantial formation of coke.

Yet another object of the invention is to minimize the preheating of reactants outside of the reaction vessel.

A still further object of the invention is to substantially cool conversion products prior to leaving the reaction vessel.

Another object of the invention is to minimize the need for highly heat resistant equipment in a high temperature process.

Other objects and advantages in the invention will be apparent to those skilled in the art upon consideration of the details below.

The above and other benefits of the invention are obtained by the continuous process for high temperature conversions of fluid reactants described hereinafter along with apparatus adapted to carry such conversions wherein a fluid reactant feed passes longitudinally through at least one confined continuous passage along an initial section of which the fluid reactant is simultaneously in indirect heat transfer relationship with a first heat exchange fluid through a confining wall of the passage and in indirect heat transfer relationship through a confining wall with a fluid conversion product flowing in a reverse direction through at least one confined return passage disposed longitudinally along said continuous passage, then circulating the fluid reactant through a final section of said continuous passage in which it is converted while in indirect heat transfer relationship through a confining wall with a second heat exchange fluid maintained separate from said first heat exchange fluid and acting to control the temperature of the reactant fluid to the desired conversion level, introducing the resulting fluid conversion product at the end of the continuous passage into the communicating open end of the return passage, passing the product through said return passage in reverse direction and withdrawing the product from said return passage. As used herein, the expression continuous passage is employed to designate a unitary, uninterrupted or integral passage containing a plurality of sections or regions rather than an endless passage. It might also be termed the primary passage or anterior passage since the fluid reactant passes through it before entering the return passage.

Other features of the invention include more specific control of heat transfer while carrying out exothermic reactions, performing certain dealkylation reactions by

the improved method and various structural details of the apparatus.

For a better understanding of the nature and objects of this invention, reference should be had to the accompanying drawings in which:

FIGURE 1 is a vertical section through one form of reaction vessel according to the present invention, and

FIGURE 2 is a similar view of another embodiment of the reactor.

Referring now to FIGURE 1, the elongated tubular vessel 10 contains an inner tube 12 comprising a wide tubular section 14 joined by the tapering or funnel-shaped section 15 to the narrower portion 16. This inner tube assembly is supported concentrically in tube 10 by the flange 18 resting on flange 20 of the outer tube. The upper ends of these tubes are covered by a plate 22 which is provided with a feed inlet 24 and a products outlet 26. Leakage from the various components is prevented with gaskets, sealing compounds or ground joints, and plate 22 and flanges 18 and 20 are securely fastened together with bolts as in FIGURE 2, screws, clamps, or other suitable means.

Optionally, a thermowell 28 may be concentrically mounted on the bottom of cover plate 22 to reduce the cross sectional area inside inner tube 12 and thus increase the velocity of the hot reaction products flowing upward therein for the purpose of increasing the transfer of heat through the wall of tube section 14. If desired, a solid core member of ceramic or other heat resistant material may be substituted for the hollow thermowell 28.

The above apparatus is constructed in known fashion of materials suitable for the conditions to be encountered in service. For instance, for a reaction at temperatures of 1200-1400° F. and pressures of 500-700 p.s.i.g., all of the elements named are desirably fabricated of heat resistant steel alloys, but only tube 10 and possibly thermowell 28 need to have heavy walls to withstand the pressure.

The entire vessel is suspended from its upper end by the flange 20 which rests upon the top 30 of the housing 32. Such suspension minimizes strains due to expansion and contraction which are ordinarily severe in elongated structures supported in conventional manner at high temperatures.

The housing is divided into an upper annular heating section or furnace 34 which is desirably lined at the side, top and bottom walls with an adequate thickness of fire brick 38 or other refractory material and also a cooling well 36. Any suitable means may be employed for heating the furnace to the desired temperature including electrical resistance, high frequency, induction or radiant elements or preferably a series of gas or oil burners 40 uniformly spaced around furnace 34 and each provided with a secondary air port 42 and adjustable shutters 44. The products of combustion leave through flue 46. While the burner nozzles are shown in radial alignment, it will be appreciated by those skilled in the art that these nozzles may be disposed tangentially in the combustion chamber or vertically parallel to tube 10 if desired.

In the lower section 36 of the housing the removal of excess reaction heat evolved in the reaction section in the lower part of tube 10 is accomplished by passing a cooling gas or other suitable fluid from the inlet connection 47 up the wall of tube 10 and out through the conduit 48. Circulation of this cooling medium may be effected by either natural draft or suitable fans or blowers. A wide variety of gases may be employed for cooling the lower end of tube 10 including air which is being preheated for use in furnace 34 or flue gas. The latter medium is particularly desirable from a safety standpoint in event of the rupture of tube 10 while containing flammable gases as these will not burn in a flue gas atmosphere. Heat picked

up by such flue gases can be readily recovered by passage through a waste heat boiler or a heat exchanger if this is economically justifiable.

While it is important in many exothermic reactions to dissipate the heat of reaction in order to avoid decomposing products or reactants, the amount of heat to be removed is often not very great. In some instances, there is no need to pass cooling gas through the well 36 as radiation alone will remove sufficient heat from the reaction occurring in tube 10. Where the lower or reaction end of tube 10 is relatively long in order to provide sufficient reaction volume in its interior, it will, of course, have a large area for heat transfer. In instances where only a relatively small quantity of reaction heat need be transferred through this large heat transfer area, it is contemplated that it may sometimes even be desirable to apply an insulating layer to the lower section of tube 10 to avoid cooling the reacting gases too much. An insulating cement, preferably of a refractory nature, may be used for the purpose.

The reaction vessel in FIGURE 1 is suspended or hangs vertically from its flange 20 and there is a sliding fit between tube 10 and the layer of insulation 38 at the bottom of the furnace 34; accordingly, the lower layer of insulation does not support any of the weight of the reactor. The inner tube 12 and core 28 are likewise supported from the top only. With extremely long tubes it may be desirable to provide spiders at one or more suitable locations around sections 14 and 16 of the inner tube and the core 28 in order to maintain the designed spacing of these elements within the tube 10 and thus promote uniform fluid flow and even temperatures at various levels of the inner tube 12.

The apparatus of this invention is well suited for reactions involving high temperatures and pressures. Considerable expansion of the tubular members occurs especially in starting up a cold reactor and as a result of their relatively great length. The present equipment is particularly well adapted to minimize the heavy expansion or contraction strains set up. Tube 10, the inner tube 12 and any core or thermowell 28 are fixed only at a single end; hence all other parts of these members are free to expand or contract freely in response to temperature changes. Also the strains arising from expansion or contraction are minimized by hanging such apparatus elements from a support rather than providing lateral supports at one or several points. However, it will be appreciated that the equipment of this invention may be disposed or oriented in any direction according to desire. For example, the reaction vessel 10 of FIGURE 1 may be aligned horizontally with the flanged end held by fixed supports and roller supports placed under the tube 10 at one or more locations along its length to permit movement. Such orientation is not generally recommended as it introduces bending stresses which are absent when the equipment is suspended vertically.

In operations according to the present invention, reactants which are desirably preheated to a substantially elevated temperature which is below the threshold of an exothermic reaction are admitted into annular zone A which extends substantially the length of broad section 14 of the inner tube and also usually equals the height of furnace 34. In zone A the reactants are heated under conditions providing optimum heat transfer with the gases moving at relatively high velocity through this zone of limited cross sectional area while heat is being transferred through the large cylindrical heat transfer areas of the inner tube section 14 of maximum diameter and also through the upper section of exterior tube 10. Generally the major portion of the heat required to raise the temperature of the reactants until the reaction starts is derived from heat given up by the reaction products in the manner described hereinafter, and for some reactions this

may be the only heat supply required. However, for many reactions, and particularly the thermal hydrodealkylation reactions mentioned herein, it is necessary, or at least desirable, to obtain the balance of the required heat from furnace 34. In some modifications of the present process it is contemplated that the furnace will furnish more heat than the reaction products to the entering reactants.

The temperature of the reactants increases as they proceed downward in the annular zone A and a temperature sufficient to initiate the exothermic reaction is attained at or shortly before reaching the bottom of zone A and entering the annular zone B of enlarged cross sectional area. No substantial input of heat from other regions into the reactants occurs in zone B. The linear velocity of flow decreases as a result of the increased area there, and the length of the zone is selected to provide an adequate residence time for the reaction to proceed to an economic optimum under the chosen conditions. Unless the particular reaction has no tendency to continue generating exothermic heat to the point of decomposition of the reactants or products or of damage to the apparatus, heat is dissipated from the tube 10 surrounding zone B at a rate sufficient to avoid such undesirable results.

The reaction is largely complete by the time the reaction mixture reaches the bottom of zone B and turns upward into the narrow zone C in the tube section 16 of the return conduit 12. For many exothermic reactions the temperature at this point may be only 25–40° F. higher than at the entrance of zone B. There is comparatively little transfer of heat between the reaction products in zone C and the reacting materials at slightly lower temperatures in the annular zone B and none is needed. This results from the facts that the narrow tube section 16 provides very little heat transfer surface, typically only 5 to 50 percent of that of tube section 14 facing zone A and there is relatively small temperature differential across tube 16, often as little as 10–25° F. The main function of tube 16 is to channel the reaction products to a fourth confined zone D of annular shape where they transfer a substantial amount of their heat to the reactants in the outer annular zone A. A relatively fast flow through a narrow tube section 16 is desirable to carry the reaction mixture quickly to a cooler zone D after an adequate residence time at the maximum desirable reaction temperature. The core or thermowell 28 speeds up the flow of gases through zone D by reducing the cross sectional area there. When a thermowell is employed, an optional thermocouple (not shown) is desirably located at the bottom of this element and may be used to regulate the heat input to furnace 34 in relation to the temperature of the uncooled reaction products. If desired, another thermocouple can be located at the bottom of zone A and the difference in temperature between the bottoms of zones D and A may be utilized to control the cooling in well 36 by varying the flow of cooling gases through inlet 47 directly with said temperature differential.

In FIGURE 2, a modification of the heat exchanger-reactor is shown in which the narrow elongated zone C is formed by the application of a layer of refractory insulation 50 to the interior of inner tube 112 instead of by the use of a tube section 16 of narrower exterior diameter as in FIGURE 1. Here the outer tube 110 differs in that it is provided with a side feed inlet connection 124 and the plurality of mounting brackets or angles 52 which are employed in place of the flange of the vessel for supporting it. The products here leave through the outlet connection 126 and cover plate 122 which is secured to the flanges 118 and 120 by a series of evenly spaced bolts 54 around the rim of the cover. For temperatures around 1350° F. or higher, a suitable material for the insulating layer 50 is a calcium aluminate cement containing about 40 percent each by weight of SiO₂ and Al₂O₃, about 10 percent CaO and smaller amounts of Fe₂O₃ and TiO₂.

Certain aspects of the invention relating to dealkyla-

tion reactions are further illustrated by the following detailed examples.

EXAMPLE 1

Toluene is subjected to thermal hydrodealkylation at a temperature of 1300–1350° F. and a pressure of 600 p.s.i.g. in the presence of hydrogen in a reactor of the type shown in FIGURE 1 to produce benzene. The internal diameter of tube 10 is 8 inches and its length is 39.5 feet. The inner tube sections 14 and 16 have lengths of 17.5 and 21.5 feet respectively, their exterior diameters are 6.0 and 1.5 inches respectively and both have a wall thickness of 0.25 inches.

A mixture of nitration grade toluene, recycle gas from a high pressure separator for the reaction products and a make up gas containing hydrogen with a minor proportion of methane is preheated to 850° F. whereby all hydrocarbons are vaporized and thereafter introduced into the reaction vessel through inlet elbow 24 at a rate of 847 lbs./hr. and a pressure of 605 p.s.i.g. By the time the charge reaches the bottom of zone A its temperature is up to 1325° F. as a result of the heat recovered from the product gases passing upward through zone D and to a lesser extent the combustion of a typical refinery fuel gas in furnace 34. During its residence time of 20 seconds in reaction zone B, the temperature of the reaction mixture rises slightly to about 1350° F. Flue gas at an inlet temperature of 700° F. is blown through the cooling chamber 36 to dissipate the excess heat of reaction and prevent the temperature from rising further in zone B. In their passage up the narrow tube section 16, the hot reaction products continue to react to a minor degree and their temperature rises to about 1355° F. The reaction is substantially complete upon reaching tube section 15. In the initial annular zone D the product gases are quickly cooled by transferring a substantial amount of heat through the inner tube to the incoming reactants in zone A, and the reactor effluent at the outlet elbow 26 has a temperature of about 1050° F. The reaction is continued for an extended period with conversion occurring of the composition and rate listed below and without deposition of coke within the reaction vessel.

	Pound Mols./Hour	
	Charge	Reaction Products
Toluene.....	4.09	1.28
Benzene.....	0.46	3.22
H ₂	24.53	21.20
Methane.....	28.77	26.62
C ₂ -C ₃	0.080	0.099
Other Aromatics.....	0.015	0.035

EXAMPLE 2

A feed stock of substantial alkyl naphthalene content is prepared by subjecting a light fuel oil fraction from a catalytic cracking operation to a sulfur dioxide treatment to extract a fraction rich in aromatic hydrocarbons and fractionally distilling the extract to separate a cut having a boiling range of 446–586° F. By reason of its high end point and the high content of paraffins, naphthenes, and olefins, as listed in the table below, this feed stock is one that is considered unsuitable for commercial thermal hydrodealkylation. However, upon subjecting this stock, together with hydrogen and a typical recycle gas composed predominantly of hydrogen and methane, to such noncatalytic dealkylation in a small scale unit of the type described in connection with FIGURE 1 good results are obtained for an extended period. The table which follows lists the operating conditions and the distinctly satisfactory yield which is obtained without encountering deposition of carbon.

Table 1

Operating conditions:		Charge
Feed temp. at inlet zone A, ° F. -----		875
Temp. at end of zone A, ° F. -----		1,305
Temp. at end of zone B, ° F. -----		1,340
Temp. of effluent of zone D, ° F. -----		1,075
Pressure, p.s.i.g. -----		600
Contact time, sec. -----		19.2
H ₂ /HC molar ratio -----		17.2
Total H ₂ input, s.c.f./b. -----	12,802	10
Fresh H ₂ input, s.c.f./b. -----	6,388	
Gas recycle, s.c.f./b. -----	11,610	
H ₂ purity of gas recycle, mol. percent -----	55.2	
H ₂ consumption, s.c.f./b. -----	2,873	
Product yields, percent charge:		15
Dry gas, percent wt. -----	44.3	
C4-5 -----	None	
C6-400° F., percent vol. -----	11.6	
400° F.+ fraction, percent vol.:		
Naphthalene -----	0.2	21.5
Methyl naphthalene -----	3.8	10.7
Other alkyl naphthalenes -----	31.7	1.5
Anthracenes-phenanthrenes -----	0.8	2.5
Other aromatics -----	31.7	7.5
Indanes, indenenes, tetralins, etc. --	8.5	--
Benzothiophenes -----	--	2.5
Paraffins, naphthenes and		
olefins -----	23.3	--
Total -----	100.0	45.6

The dry gas product listed in the above table consists of normally gaseous hydrocarbons containing from 1 to 3 carbon atoms per molecule.

It is apparent that the process and apparatus of this invention are particularly adaptable to the dealkylation of a wide variety of hydrocarbons and mixtures thereof. Thus, the charging stocks may contain alkylated aromatic hydrocarbons, such as toluene, xylenes, ethyl benzene, propyl benzene, methylethyl benzene, diethyl benzene and the like and likewise the indanes, such as 1-methyl indane, 2-methyl indane, 4-methyl indane, 5-methyl indane; 4,5,6-trimethyl indane, 1,1-dimethyl indane, 1,2-dimethyl indane, 1,2,3-trimethyl indane and 1,2,3,4,5,6,7-heptamethyl indane. The aromatic hydrocarbon fraction may include alkyl naphthalenes and compounds such as alkyl tetralins. Unalkylated aromatic materials such as benzene, indane, tetralin, and naphthalene may also be present. Olefins, naphthenes and paraffins may also be contained in these feed stocks.

For instance, economical feeds for benzene production are reformates boiling above 200° F. with atmospheric end boiling points up to about 400° by the A.S.T.M. procedure, these charge stocks are obtained by catalytically reforming naphthas in the C₆-250° F. boiling range. The reforming operation also produces an ample net yield of a desirable hydrogen-rich gas which may be used as a reactant in the dealkylation step. If naphthalene is also desired as a product, the reformat used as feed for that purpose should have an end point well in excess of 400° F. When it is desired to produce both benzene and naphthalene, mixtures of the two reformat charging stocks may be used. It is sometimes desirable, although not necessary, to concentrate the aromatic hydrocarbons in a feed of this type by subjecting it to conventional extraction processes and then employ the resulting aromatic hydrocarbon fraction as the feed.

For maximum yields of naphthalene, the feed stock is desirably a 400° F.+ reformat and such may be obtained by reforming kerosines boiling in the 380-650° F. range. Preferred dealkylation feeds are reformates with initial boiling points of about 400-420° and end points of about 500-550°, and these are desirably obtained by reforming 400-475° F. kerosines.

It is desirable to preheat the feed to a temperature between about 750 and 1000° F. before introduction into the apparatus described hereinbefore. In the initial section designated as zone A of the reactor, the charge should be further heated to a temperature of 1100-1350° F. for benzene production and to a temperature of at least 1200° for naphthalene production inasmuch as the desired reaction temperatures are within the over-all ranges of 1100-1400° F. for producing benzene and 1200-1400° F. for naphthalene. The preferred temperature ranges are about 1200-1400° for benzene and 1250-1400° F. for naphthalene. In the return passage, and especially the upper section of zone D thereof, the dealkylation products are cooled to a temperature of 1150° F. or lower before leaving through outlet fitting 26 and a temperature no higher than 1050° F. is preferred.

In addition to the ranges of reaction temperatures stated earlier, the other reaction conditions should be kept within certain limits for good results. The hydrogen in the gaseous portion of the charge should be sufficient to avoid depositing coke and should amount to at least about 4000 s.c.f./b. of normally liquid hydrocarbons in the feed. With relatively pure toluene or a monoalkyl naphthalene, it is preferred that the hydrogen charge does not exceed 12,500 s.c.f./b., and rates between 4,500 and 10,000 s.c.f./b. are especially recommended, which range is equivalent to 3.6 to 8 mols of hydrogen per mol of toluene. Excessive quantities of hydrogen are not only wasteful of fuel and power but they also tend to produce an undesirable cracking of aromatic rings in the material charged.

The hydrogen content of the normally gaseous portion of the charge may be at least 20 mol percent and preferably between about 40 and 85 mol percent. The total reaction pressure is also significant and should be maintained between about 250 and 800 p.s.i.g. (preferably about 350-700) with an inlet hydrogen partial pressure of about 130 to 785 p.s.i. (preferably about 250-550).

Suitable residence times for the hydrodealkylation reactions run from about 2 to 80 seconds and the range of about 10-70 seconds is preferred for commercial purposes.

As indicated earlier, it has not proven commercially feasible to produce naphthalene from certain difficult feed stocks with end points above 525° F. that contain more than 10% by weight of nonaromatic normally liquid hydrocarbons, and especially those with more than 15%, because of the excessive formation of coke. However, in concurrently filed application Serial No. 290,942 of Edward J. Moll, Jr., entitled "Thermal Hydrodealkylation of Naphthalene Precursors," it is disclosed that coking may be avoided in thermal hydrodealkylation reactions with such feed stocks by a combination of a high hydrogen charging rate and extremely close control of reaction temperatures. From about 8,400 to 21,000 s.c.f. of hydrogen (preferably about 10,500 to 17,500 s.c.f.) should be charged per barrel of normally liquid hydrocarbons in the feed. The reaction temperatures should be held within about 20° F. of the preselected figure and also not below 1,200° nor above 1,400°, and a minimum of 1250° is more desirable. Example 2 hereinbefore demonstrates the application of these teachings in conjunction with the instant invention, and in holding the reaction temperatures steady within 20°, one may ignore the brief interval in zone A when the reactants are above the reaction threshold temperature while being heated up to about the average reaction temperature.

While the apparatus of the present invention has been described in relation to individual units, it will be appreciated that these may be mounted in batteries or multiple units for large scale operations. This is especially suitable for high temperature and high pressure reactions where relatively narrow tubular reaction vessels are preferred. Such multiple units may be mounted in a single

extended housing having single or multiple furnace sections and cooling wells.

In addition to the many benefits described hereinbefore, there are a number of other advantages obtainable with the invention including the fact that the cooling of the reaction products in zone D minimizes, and in some cases eliminates, the need for quenching the reaction products. Also, it will be apparent that the heating of the feed in the initial zone A and the cooling of the reaction mixture in zone B and the products in final zone D effectively level out the temperatures encountered and thus an isothermal process is approximated. The charge may be preheated before entering the reactor to a considerably lower temperature than formerly was the case. Only the reaction vessel is subjected to the full severity of the reaction conditions so far as heat and corrosion are concerned, hence the outside piping and other equipment may be constructed with thinner walls and/or lower alloys at a considerable saving.

Although the novel process and apparatus have been described herein in connection with a few limited embodiments of the invention, it will be appreciated by those skilled in the art that this invention is applicable in many equipment manifestations and to a wide variety of processes, including hydrocracking, catalytic reforming, catalytic hydrodealkylation and hydrogenation reactions. Therefore, it is to be understood that the present invention is not restricted to specific embodiments or details mentioned hereinbefore unless such limitations are specifically included in the appended claims.

I claim:

1. A method for conducting high temperature conversions of fluid reactants which comprises passing a fluid reactant feed longitudinally through at least one confined continuous passage along an initial section of which the fluid reactant is simultaneously in indirect heat transfer relationship with a first heat exchange fluid through a confining wall of the passage having high heat transfer characteristics and in indirect heat transfer relationship through a confining wall having high heat transfer characteristics with a fluid conversion product flowing in a reverse direction through at least one confined return passage disposed longitudinally along said continuous passage, then circulating the fluid reactant through a final section of said continuous passage in which it is converted while in indirect heat transfer relationship through a confining wall with a second heat exchange fluid maintained apart both from said first heat exchange fluid and from said conversion product and acting to control the temperature of the reactant fluid at the desired conversion level, introducing the resulting fluid conversion product at the end of said continuous passage into the communicating open end of a return passage, passing the product through said return passage in reverse direction to the flow in said continuous passage and withdrawing the product from said return passage.

2. A method according to claim 1 in which said fluid reactant is heated in said initial section of the continuous passage to a temperature sufficient to initiate an exothermic reaction and the reaction occurring in said final section of the continuous passage is controlled by cooling with said second heat exchange fluid.

3. A method according to claim 1 in which a high indirect heat transfer is maintained between said product and the feed in said initial section of the continuous passage and a low indirect heat transfer is maintained between said product and the fluid within said final section of the continuous passage.

4. A method according to claim 1 in which said feed is heated sufficiently while passing through said initial section of the continuous passage at least partially by a high indirect transfer of heat from said product to initiate an exothermic reaction and a low indirect heat transfer is maintained between said product and the fluid within said

final section of the continuous passage while cooling the exothermic reaction with said second heat exchange fluid.

5. A method according to claim 1 in which said feed is heated sufficiently while passing through said initial section of the continuous passage at least partially by a high indirect transfer of heat from said product to initiate an exothermic reaction, a low indirect heat transfer is maintained between said product and the fluid within said final section of the continuous passage while cooling the exothermic reaction with said second heat exchange fluid and said product is rapidly passed through the initial region of said return passage which is in heat exchange relationship with the fluid within said final section of the continuous passage.

6. A method according to claim 1 in which said feed is heated sufficiently while passing through said initial section of the continuous passage at least partially by a high indirect transfer of heat from said product through a large heat transfer area to initiate an exothermic reaction, the exothermic reaction is cooled by said second heat exchange fluid, and said product is maintained in low indirect heat transfer relationship with said fluid reaction mixture in said final section of the continuous passage by means including a substantially smaller heat transfer area.

7. A method according to claim 1 in which said fluid reactant is preheated to an elevated temperature below the threshold of said conversion before it is introduced into said initial section of the continuous passage.

8. A method according to claim 1 in which a feed containing an alkyl aromatic hydrocarbon and a proportion of hydrogen amounting to at least 4000 standard cubic feet of hydrogen per barrel of normally liquid hydrocarbons is heated to a temperature between about 1100 and 1350° F. in said initial section of the continuous passage, reacted to form an unalkylated aromatic hydrocarbon at temperatures between about 1100 and 1400° F. under a pressure between about 250 and 800 p.s.i.g. in said final section of the continuous passage, and said product is cooled to a temperature between 900 and 1150° F. in said return passage.

9. A method according to claim 1 in which a feed containing an alkyl naphthalene and a proportion of hydrogen amounting to at least about 4000 standard cubic feet of hydrogen per barrel of normally liquid hydrocarbons is preheated to a temperature between about 700 and 1000° F., further heated in said initial section of the continuous passage to a temperature between 1200 and 1350° F., reacted to form naphthalene under a pressure between about 250 and 800 p.s.i.g. and temperatures throughout said final section of the continuous passage maintained between about 1200 and 1400° F., and said product is cooled in said return passage to a temperature between 900 and 1150° F. by transferring heat indirectly at a high rate to the fluid within said initial section of the continuous passage.

10. A method according to claim 1 in which a feed containing an alkyl benzene and a proportion of hydrogen between about 4500 and 10,000 standard cubic feet of hydrogen per barrel of normally liquid hydrocarbons is preheated to a temperature between 700 and 1000° F., further heated in said initial section of the continuous passage to a temperature between 1100 and 1350° F., reacted to form benzene under a pressure between about 250 and 800 p.s.i.g. at temperatures between about 1200° F. and 1400° F., and said product is cooled in said return passage to a temperature between 900 and 1150° F. by transferring heat indirectly to the fluid reactant in said initial section of the continuous passage.

11. Apparatus for conducting high temperature conversions of fluid reactants which comprises a vessel having at least one confined continuous passage with an initial section in indirect heat transfer relationship with a first heat exchange fluid in an adjacent confined channel through a confining wall of the passage having high heat transfer characteristics and in indirect heat transfer

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relationship through a confining wall having high heat transfer characteristics with at least one confined return passage disposed longitudinally along said continuous passage and a final section of said continuous passage in indirect heat transfer relationship through a confining wall with a second heat exchange fluid in an adjacent confined channel, return conduit means for each said return passage disposed for the flow of a fluid conversion product in a reverse direction relative to the flow of a fluid reactant through said continuous passage and provided with an open end communicating with the end of said final section of the continuous passage, means for introducing a fluid reactant into said vessel, means for separately introducing each of said heat exchange fluids at a predetermined temperature into contact with each of said confining walls respectively associated therewith in heat transfer relationship, and means for withdrawing a fluid conversion product from said vessel.

12. Apparatus according to claim 11 in which the cross sectional area of said final section of the continuous passage is substantially greater than the cross sectional area of the initial region of said return passage in indirect heat transfer relationship therewith.

13. Apparatus according to claim 11 in which said initial and final sections of the continuous passage are in indirect heat transfer relationship with final and initial regions respectively of said return passage and the cross sectional area of said final section of the continuous passage is greater than the several cross sectional areas of said initial region of the return passage and of said initial section of the continuous passage.

14. Apparatus according to claim 11 in which said initial and final sections of the continuous passage are in indirect heat transfer relationship with final and initial regions respectively of said return passage and the ratios of the cross sectional areas of said final section of the continuous passage, said initial region of the return passage and said initial section of the continuous passage are within the ranges of about 100:1-10:15-75 respectively.

15. Apparatus according to claim 11 in which said vessel is elongated and a single end thereof is attached to a fixed support.

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16. Apparatus according to claim 11 in which said vessel is elongated and suspended vertically by a single end thereof from a fixed support.

17. Apparatus according to claim 11 in which heat is transferred to said initial section of the continuous passage from said first heat exchange fluid flowing at a higher temperature through a confined channel disposed longitudinally along said initial section, and said final section of the continuous passage is cooled by flowing said second heat exchange fluid at a lower temperature through a confined channel disposed longitudinally along said final section.

18. Apparatus according to claim 11 in which heat is transferred to said initial section of the continuous passage from said first heat exchange fluid flowing at a higher temperature through a confined channel disposed longitudinally along said initial section, said final section of the continuous passage is cooled by flowing said second heat exchange fluid at a lower temperature through a confined channel disposed longitudinally along said final section and both of said channels are bounded by at least one wall located outside said vessel.

19. Apparatus according to claim 11 in which said return passage is surrounded for at least a substantial proportion of its length by said continuous passage.

20. Apparatus according to claim 11 in which said vessel comprises an elongated tube of substantially uniform diameter with a narrower tube of varying cross section mounted coaxially therein, said continuous passage is located in the annular space between the tubes, said return passage is located within the inner tube and the diameter of the portion of the inner tube within said initial section of the continuous passage is substantially greater than the diameter of the inner tube within said final section of the continuous passage.

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