Nov. 14, 1933. A. O. JAEGER 1,935,053

CATALYTIC APPARATUS

Filed June 28, 1929

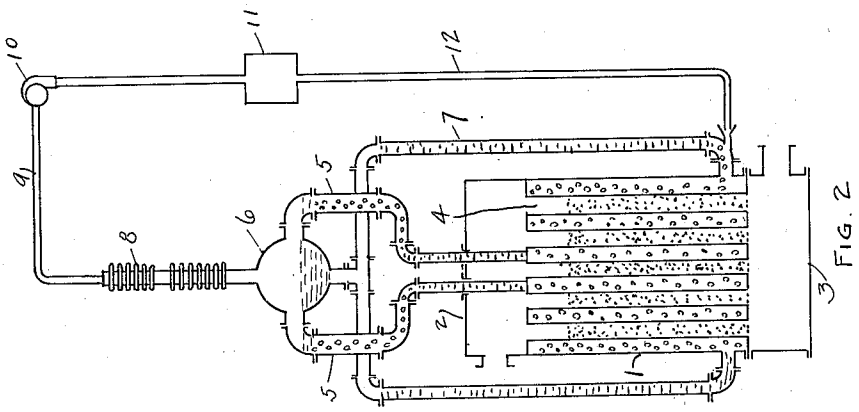


FIG. 2

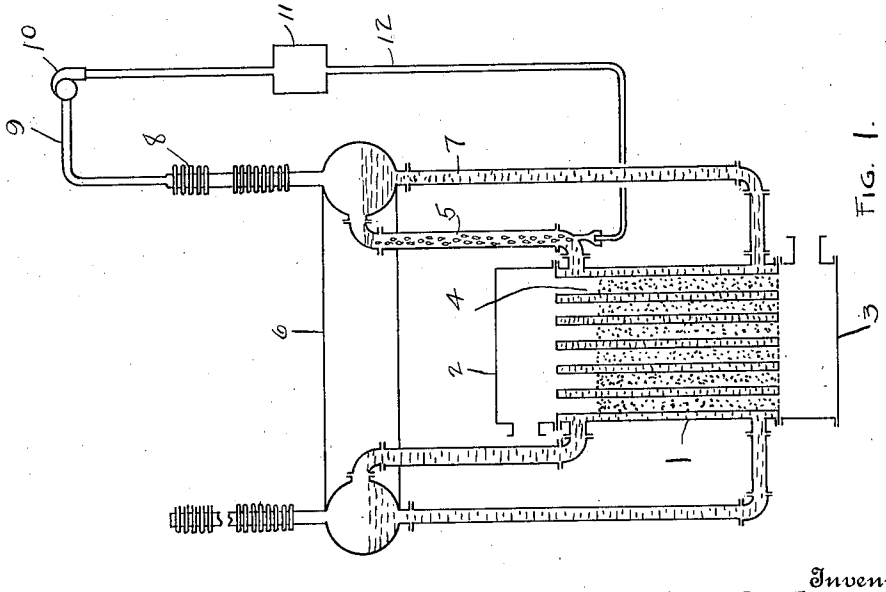


FIG. 1.

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CATALYTIC APPARATUS

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16 Claims. (Cl. 23—288)

This invention relates to catalytic apparatus and more particularly to converters cooled by baths.

In my Patent 1,900,715, I have described and claimed converters in which a bath is used having a boiling point approximately at the temperature desired in the hottest portion of the converter bath, which bath is subjected to sufficient pressure when in contact with the catalytic zones that it does not boil and is then circulated to a zone of lower pressure where it is permitted to boil, and then recirculated with or without further cooling to a lower portion of the bath in the converter. This type of converter presents many advantages fully enumerated in the application above referred to, but possesses the disadvantage that in large sizes it is sometimes difficult to provide for sufficiently rapid circulation.

The present invention is an improvement over the type of converter described in my prior patent above referred to, and consists in the acceleration of the circulation of the bath by introduction at a point below the highest level of an inert gas such as, for example, carbon dioxide, nitrogen, steam, or other suitable medium. The bubbles of the inert gas rising through the liquid reduce its specific gravity and bring about and greatly enhance the circulation. At the same time the inert gas may aid in the cooling as it can be introduced at any suitable temperature and, thus, forms a desirable additional cooling means. The enhanced circulation possible by means of the present invention permits the use of larger converters at higher loadings and provides for a more effective and more uniform cooling effect due to the greater speed of circulation.

The present invention is applicable to bath converters using any suitable type of boiling bath liquid and is particularly effective with bath converters for organic oxidations where baths containing mercury, mercury alloys, sulfur, and the like are used. It should be understood however that the present invention is of general applicability and in cases of exothermic reactions which take place at lower temperatures other boiling baths such as those containing mineral oils may be used.

The present invention may, for example be used in connection with the following reactions:—the oxidation of aromatic hydrocarbons, for example, naphthalene to alphanaphthaquinone, phthalic anhydride and maleic acid; benzol, phenol, or tar phenols to maleic acid, and compounds containing the group—$CH_2—CH=CH—CH_2—$ may be oxidized to maleic and fumaric or mesotartaric acid; anthracene to naphthalic anhydride; cresol can be oxidized to salicylaldehyde and salicylic acid; toluol and various halogen and nitro-substituted toluols or other aromatic side chain compounds such as xylenes, pseudocumene, mesitylene, paracymene, and the like, may be oxidized to the corresponding aldehydes and acids with air or other oxidizing gases such as, for example, mixtures of oxygen and nitrogen containing a smaller percentage of oxygen than air, mixtures of carbon dioxide and oxygen, and the like. Phenanthrene may be oxidized to phenanthraquinone, diphenic acid, phthalic anhydride and maleic acid; fluorene to fluorenone; eugenol and isoeugenol to vanillin and vanillic acid; methyl alcohol and methane to formaldehyde, ethyl alcohol to acetic acid; ethylene chlorhydrine to cloracetic acid, and the like.

Another important series of organic reactions which are strongly exothermic are the purification of crude compounds by selective burning out of impurities, such as, for example, the catalytic purification of crude anthracenes with various degrees of impurity with total combustion of carbazole, dead oils and in some cases phenanthrene; purification of crude naphthalenes and crude mononuclear hydrocarbons, such as benzols and the like; purification of ammonia from coal tar with the burning out of organic impurities such as phenolic bodies or sulfur compounds, both organic and inorganic.

Reactions in which mixtures of organic compounds are oxidized to intermediate compounds with removal of impurities are also effectively carried out by means of the present process, thus, for example, crude anthracenes, phenanthrenes, and the like may be oxidized to anthraquinone, phenanthraquinone, diphenic acid or phthalic anhydride with concomitant removal of carbazole and dead oils by total combustion. The oxidation of crude tar acids to maleic and fumaric acids with combustion of certain impurities is another example.

Many reductions and hydrogenations are strongly exothermic, such as, for example, the reduction of many nitro-compounds, as nitrobenzene, dinitrobenzene, nitrophenol, nitronaphthalene and their homologues to the corresponding amines or hydrogenated amines. The reduction of carbon monoxide to methane, higher alcohols, ketones or petroleum like products, the hydrogenation of aromatic compounds to alicyclic compounds, such as benzene to cyclohexane, naphthalene to tetraline or decaline, anthracene to hydrogenated anthracene, phenol to cyclo-

hexanol, acetylene to ethylene and ethane, and the like.

Composite reactions in which oxides of carbon are reduced in the presence of other organic compounds, aliphatic, alicyclic or aromatic, are also in many cases strongly exothermic and well suited for the present process.

In addition to the extremely important contact sulfuric acid process there are other inorganic catalyses which are exothermic and which can be effectively carried out by means of the present invention. Examples of such reactions are the synthesis of ammonia from its elements, the oxidation of ammonia to oxides of nitrogen, and the like.

The invention will be described in greater detail in connection with the drawing which discloses some typical embodiments and in which:

Fig. 1 is a vertical section through a converter using the circulation of the present invention only in the riser pipes; and

Fig. 2 is a vertical section through a modified converter in which the circulation of the present invention is used throughout the whole height of the bath.

The converter shown in Fig. 1, which is of course entirely diagrammatic in nature as are all the drawings in this case, consists of a shell 1, top piece 2, bottom piece 3 and catalyst tubes 4 fitting into two tube-sheets, and being surrounded by a bath which, under the pressure in the converter, boils just above the temperature desired for the upper portion of the catalyst tubes. The bath is connected to a drum 6 through riser pipes (5) while the bottom of the converter is connected to the bottom of the drum through down pipes 7. The vapor space of the drum 6 connects to reflux condensers 8, from one of which a pipe 9 leads to a blower 10, the discharge of which is connected through an equalizing reservoir 11 and pipe 12 to a nozzle in the bottom of the riser pipes 5.

In operation the reaction gases flow through the catalyst tubes, for example, from the top to the bottom and the heat evolved causes the temperature of the bath to rise whereupon the bath begins to circulate by thermosiphon action. As the heated bath rises in the riser pipes 5, its pressure diminishes and it may boil in the riser pipes, accelerating the circulation. The blower 10 blows bubbles of air into the riser pipes 5, greatly accelerating the circulation therein and in many cases provides sufficient cooling so that the bath will not boil at all in the riser pipes. The speed of circulation can be governed by the amount of inert gas injected which, in turn, if desired, may be thermostatically controlled in the ordinary way by the temperature either of the bath or of the catalyst. The electric relays are of the usual type and are not shown as their particular structure forms no part of the present invention, any suitable design being usable.

The modification shown in Fig. 1 operates very satisfactorily for converters of moderate size or where the reaction is not excessively exothermic. The bath may boil during normal operation in the riser pipes or the drum or it may possess a boiling point sufficiently high so that under normal circumstances it remains just below the boiling point, but will nevertheless boil if excessive temperatures are generated, thus acting to a certain extent as a safety valve. The particular boiling point of the bath will be determined in accordance with the requirements of the reaction in question.

Fig. 2 shows a modification which is applicable to converters of much larger size than is the modification shown in Fig. 1. The same reference numerals are used to designate similar parts in the two figures. It will be noted that the riser pipes in Fig. 2 rise from the center of the converter, whereas the return of heated liquid is at the side, thus preventing any possibility of overheating the central catalyst tubes which may occur with very large converters and highly exothermic reactions when the modification shown in Fig. 1 is used. The construction in Fig. 2 is somewhat more complicated, but for more intense cooling the additional capacity frequently makes the additional complication worth while.

In the modification shown in Fig. 2, the inert gas is discharged into the bath at the bottom of the converter so that the bubbles not only serve to accelerate the flow in the riser pipes but also speed up the flow in the converter itself. Since the amount of gas introduced is, of course, somewhat greater and its contact with the bath is for a longer period of time, the cooling effect is normally also greater, thus making this modification particularly applicable to highly exothermic reactions.

The gas circuit shown in Fig. 2 may, of course, be applied to the modification shown in Fig. 1 and vice versa, the injection of gas into the riser pipes only may be applied to the modification shown in Fig. 2. The point at which the gas is introduced will be determined by the speed of circulation required and the other factors involved.

It will be noted that it is possible to operate the system with the bath normally not boiling or only boiling a very little, most of the excess heat being taken out by the circulating gas or by cooling in the down pipes 7. The heat evolved can, of course, be utilized by passing the gases either before or after encountering the blower 10 through suitable heat exchangers, for example a steam boiler, thus making the heat of reaction economically available. Similarly, where the bath boils in the drum 6 and riser pipes 5 this heat may be used by using water jacketed reflux condensers instead of the air-cooled refluxes shown in the drawing. The operation is, of course, the same and the choice will depend on the amount of condensation of bath liquid which is required. Where the boiling is very slight, it will be normally preferable to use the cheaper design of air-cooled refluxes, whereas with a large amount of boiling it is frequently worth while to utilize this heat in raising steam or for any other desired purpose. Any mechanically entrained bath may be removed before the gases circulate through the bath, if necessary by suitable baffles in the reflux or in the pipe above it. In normal operation baffles are not necessary, but in some cases with a very rapid gas circulation they may be desirable and are, of course, included in the scope of the invention.

The drawing shows the converters of the present invention in a purely diagrammatic form, it being understood, of course, that the usual features of converter design, such as insulation and the like and suitable accessories, such as temperature measuring instruments, electric relays and the like, will be applied by the skilled engineer.

This application is in part a continuation of my co-pending application Serial No. 275,044 filed May 4, 1928, now Patent No. 1,900,715.

What is claimed as new is:

1. A method of carrying out exothermic, vapor phase, catalytic reactions, which comprises passing the reaction gases through at least one catalyst zone, absorbing the exothermic heat of reaction by a non-boiling liquid in heat exchanging relation thereto, removing the liquid from the catalytic zone, reducing its pressure to a point at which the liquid will boil at or only slightly below the temperature at which it leaves the catalyst zone, condensing any vapors of bath given off, returning the condensate and unboiled liquid to the catalytic zone at the pressure there existing and accelerating the circulation by introducing an inert gaseous medium at a portion of the circuit where the normal circulation is upward, the point being below the highest point thereof.

2. A method of carrying out exothermic, vapor phase, catalytic reactions, which comprises passing reaction gases through at least one catalyst zone, absorbing the exothermic heat of reaction by a non-boiling liquid in heat exchanging relation thereto, removing the liquid from the catalytic zone, reducing its pressure to a point at which at least a part of the liquid boils, condensing the vapors thus given off, and returning the condensate and unboiled liquid to the catalyst zone at the pressure there existing, and accelerating the circulation by introducing an inert gaseous medium at a portion of the circuit where the normal circulation is upward, the point being below the highest point thereof.

3. A method of carrying out exothermic, vapor phase, catalytic reactions, which comprises passing reacting gases through at least one catalytic zone or compartment, circulating a bath liquid over said zone in a closed circuit, the catalytic zone being sufficiently below the highest point in the system so that the liquid is under sufficient pressure while in contact with the catalytic zone to prevent its boiling, the liquid having a boiling point at atmospheric pressure not substantially above the highest temperature it normally acquires in passing over the catalyst zone and accelerating the upward circulation by introducing an inert gaseous medium at a point below the highest point of the system.

4. A method of carrying out exothermic, catalytic, vapor phase reactions, which comprises passing reaction gases through at least one catalytic compartment or zone, circulating a bath liquid over said zone in a closed circuit, the catalytic zone being sufficiently below the highest point in the system so that the liquid is under sufficient pressure while in contact with the catalytic zone to prevent its boiling, the liquid having a boiling point so that a portion at least boils on reaching the highest point in the zone, and accelerating the upward circulation of the liquid by introducing an inert gas therein at a point below the highest level thereof.

5. A method of carrying out exothermic, catalytic, vapor phase reactions, which comprises passing reaction gases downwardly through at least one catalytic compartment or zone, circulating a bath liquid over said zone in a closed circuit, the upward flow being counter-current to the reaction gases, the catalytic zone being sufficiently below the highest point in the system so that the liquid is under sufficient pressure while in contact with the catalytic zone to prevent its boiling, the liquid having a boiling point so that a portion at least boils on reaching the highest point in the zone, and accelerating the upward circulation of the liquid by introducing an inert gas therein at a point below the highest level thereof.

6. A method according to claim 1, in which the gaseous medium is introduced into the lowest portion of the circuit.

7. A method according to claim 2, in which the gaseous medium is introduced into the lowest portion of the circuit.

8. A method according to claim 1, in which the inert gas is disengaged from the bath at its highest point and is recirculated.

9. A method according to claim 2, in which the inert gas is disengaged from the bath at its highest point and is recirculated.

10. A method according to claim 1, in which the inert gas is disengaged from the bath at the point of lowest pressure and recirculated, at least a portion of the heat thereof being abstracted.

11. A method according to claim 2, in which the inert gas is disengaged from the bath at the point of lowest pressure and recirculated, at least a portion of the heat thereof being abstracted.

12. An apparatus for carrying out exothermic vapor phase catalytic reactions, which comprises at least one catalytic compartment containing a catalyst and having bath space in heat exchanging relation thereto, means for circulating a bath under pressure through said space in a closed circuit, means for maintaining at least one portion of said circuit at a pressure lower than the space in heat exchanging relation to the catalytic zone, and means for introducing a gaseous medium into a portion of said circuit where the circulation is upward.

13. An apparatus for carrying out exothermic, vapor phase, catalytic reactions, which comprises at least one catalytic compartment containing a catalyst and having bath space in heat exchanging relation thereto, means for circulating a bath under pressure through said space in a closed circuit, means for maintaining at least one portion of said circuit at a pressure lower than the space in heat exchanging relation to the catalytic zone, and means for introducing a gaseous medium into the lowest portion of said circuit.

14. An apparatus for carrying out vapor phase, exothermic, chemical reactions, which comprises in combination a converter provided with a plurality of catalyst compartments and a bath space therearound, a vapor disengaging vessel above said converter, means on said vessel for condensing vapors and returning condensate thereto, means connecting the upper portion of the bath space of the converter to said vapor disengaging vessel, means for connecting the liquid space of said vapor disengaging vessel to a low portion of the converter bath space, and means for introducing a gaseous medium into a portion of said circuit where the circulation is upward.

15. An apparatus for carrying out vapor phase, exothermic, chemical reactions, which comprises in combination a converter provided with a plurality of catalyst compartments and a bath space therearound, a vapor disengaging vessel above said converter, means on said vessel for condensing vapors and returning condensate thereto, means connecting the upper portion of the bath space of the converter to said disengaging vessel, means for connecting the liquid space of said vapor disengaging vessel to a low portion of the converter bath space and means for in-

troducing a gaseous medium into the lowest portion of said circuit.

16. An apparatus for carrying out exothermic, vapor phase, catalytic reactions, which comprises at least one catalytic compartment containing a catalyst and having bath space in heat exchanging relation thereto, means for circulating a bath under pressure through said space in a closed circuit, means for maintaining at least one portion of said circuit at a portion lower than the space in heat exchanging relation to the catalytic zone, and means for introducing a gaseous medium into a portion of said circuit where the circulation is upward, means for removing the gaseous medium at the portion of lowest pressure of said circuit, and means for recirculating the gaseous medium so removed.

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