Aug. 6, 1935. F. A. CANON 2,010,217

CATALYTIC APPARATUS

Filed May 6, 1929

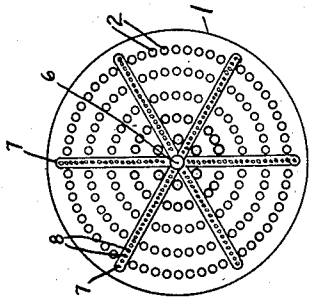


FIG 2

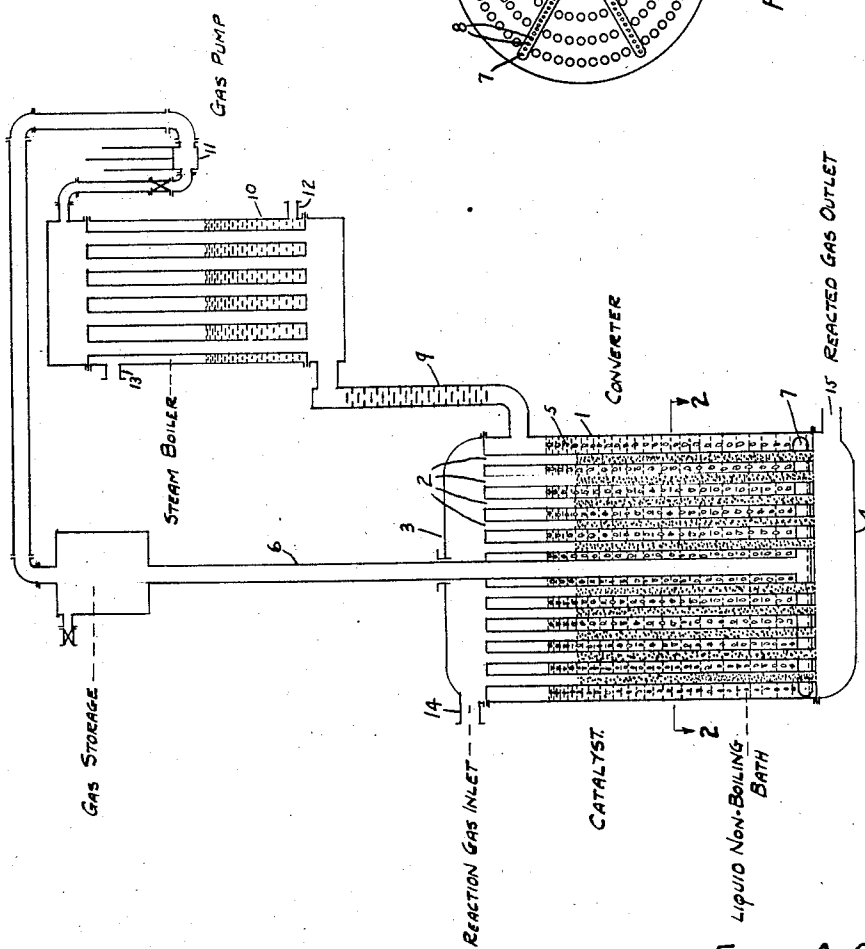


FIG. 1

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Patented Aug. 6, 1935 2,010,217

UNITED STATES PATENT OFFICE

2,010,217

CATALYTIC APPARATUS

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Application May 6, 1929, Serial No. 360,730

1 Claim. (Cl. 23—288)

This invention relates to catalytic apparatus and more particularly to tubular converters cooled by baths. Tubular converters for vapor phase catalytic reactions have been cooled by baths boiling or non-boiling, but in the case of strongly exothermic reactions, such as for example, the oxidation of organic compounds control has been difficult in large converter sizes with baths which are non-boiling, since it is difficult to remove the heat from the catalyst tubes located in the center of the converter with a non-boiling bath unless elaborate circulation means are employed. The boiling bath converters possess greater cooling capacity but are open to the disadvantages of boiling baths which, for high temperatures such as are used in the oxidation of organic compounds, practically necessitates using mercury or an alloy of mercury.

The present invention avoids all of the disadvantages of a non-boiling bath, removes the heat equally effectively from large and small converters and does not contain any moving parts. At the same time all of the advantageous features of the non-boiling baths are retained. In the present invention a tubular converter is provided with a non-boiling bath and bubbles of a gaseous cooling medium are continuously forced through the bath, absorbing the exotherm generated by the reaction and acting as effectively in the center of the converter as at the periphery, therefore, making large converters as readily controlled as small ones. The cooling medium may be any inert gas such as nitrogen, carbon dioxide, (where it does not attack the bath) steam, etc. I prefer to use steam because of its greater cooling capacity which is approximately double that of such gases as nitrogen and, therefore, a smaller amount of gas may be circulated which simplifies the operation of the apparatus.

The present invention is applicable to the oxidation of organic compounds, such as for example naphthalene to phthalic anhydride, benzol, phenol and tar phenols to maleic acid, substituted and unsubstituted side chain aromatic compounds such as toluols, xylenes, etc., to the corresponding aldehydes and acids, aromatic hydrocarbons to quinones, such as benzol to benzoquinones, naphthalene to alphanaphthaquinone, anthracene to anthraquinone, phenanthrene to phenanthraquinone, acenaphthene or acenaphthylene to acenaphthaquinone and the like, phenanthrene to diphenic acid, phthalic anhydride and maleic acid, acenaphthene and acenaphthylene to bisacenaphthylidenedione, naphthaldehydic acid, naphthalic anhydride and hemimellitic acid, fluorene to fluorenone, isoeugenol to vanillin and vanillic acid, cresol and its analogues to salicylaldehyde and salicylic acid and their analogues, heterocyclic compounds such as furfural to maleic and mesotartaric acid, aliphatic alcohols such as methyl alcohol to aldehydes such as formaldehyde, ethylene chlorinhydrine to chloracetic acid, and the like, require a very careful temperature control in order to prevent undesired side reactions and losses by total combustion, which easily take place as the reactions are in unstable equilibrium and tend to proceed too far because of great exotherm.

Other vapor phase catalytic reactions, such as the purification of organic compounds, for instance crude polynuclear aromatic compounds such as crude anthracene, crude phenanthrene, crude acenaphthene, crude anthraquinon, crude naphthalene or crude mononuclear aromatic hydrocarbons by selective oxidation of undesired impurities, such as, for example, carbazole in the case of crude anthracene, likewise require a careful temperature control and control of the exotherm evolved, as does the catalytic purification of by-product ammonia by oxidation of organic impurities or sulfur compounds. A similar careful temperature control is necessary in hydrogenations, synthesis of ammonia, oxidation of ammonia and the like.

The invention will be described in connection with the drawing which shows diagrammatically a typical converter system embodying the prinpiple of the present invention, and in which

Figure 1 is a vertical section through the whole system; and

Figure 2 is a horizontal section taken along the line 2—2 of Figure 1.

In the drawing the converter is shown of the usual tubular type with a shell 1, catalyst tubes 2, top piece 3 and bottom piece 4, and is provided with gas inlets 14, and gas outlets 15. The tubes are surrounded by a non-boiling bath as shown at 5 which may, for example, be a molten metal, such as lead or a eutectic mixture of salts, such as a mixture of sodium nitrate and sodium nitrite or any other suitable bath. The upper portion of the catalyst tubes are empty and this portion is surrounded for a considerable distance by the nonboiling bath. Reaction gases passing down through the empty upper portions of the catalyst tubes are preheated by means of the non-boiling bath surrounding the tubes. The bath does not completely fill the chamber surrounding the tubes as is shown, and the empty space above the bath level is connected to a steam boiler 10 by the pipe 9 filled with baffles. The boiler is shown diagrammatically as a fire tube boiler with feed water inlet 12, and steam outlet 13. From the top of the boiler 10, a pipe leads to a pump 11 shown diagrammatically as a plunger pump, and which forces the gas through the pipe 6 and reservoir into the perforated spider 7 which is provided with the perforations 8 and is located at the bottom of the converter. The gas emerging from the holes 8 in the form of fine bubbles passes

upwardly through the bath 5, absorbs the heat generated by the reaction in the catalyst tubes and becomes highly heated, passing on up through the pipe 9 where any liquid bath carried along as spray is caught by the baffles and returned. The gas then passes through the steam boiler generating steam and being thereby cooled.

The drawing is purely diagrammatic and details of construction are omitted, thus for example, in practical operation it is usually necessary to provide suitable insulation and temperature measuring means. The pump 11 is driven by any suitable source, and may advantageously be driven by an electric motor, the speed of which is controlled by the temperature of the bath, or the temperature of the catalyst in the tubes through suitable relays (not shown as their construction is well-known to the chemical engineer).

The gaseous medium may, as has been stated above, be an inert gas such as nitrogen in which case the steam pressure in the boiler 10 may be chosen at any desired figure. Where, however, steam is used as a circulating means, the pressure of the steam generated in the boiler 10 should be higher than the pressure under which the steam is forced through the pipe 6 in order to avoid condensation. When steam is used, of course, a connection from the pipe 6 to a source of steam should be provided in order to make up for any steam which leaks out. If the source of steam is at a higher pressure than that normally desired in the pipe 6, the connection should be provided with a suitable reducing valve.

It will be seen that the cooling medium is distributed throughout the bath and that it removes the heat in close proximity to the catalyst tubes where the heat is generated. The system is therefore equally effective with large and small converters as there is no danger of the center tubes being inadequately cooled. Normally the converter shell will be well insulated as is usual in bath converters where the shell is not relied on as a cooling means, but there is always some cooling due to the converter shell. This may be compensated by decreasing the size or the number of holes near the ends of the pipes of the spider 7, in order to effect uniform temperature regulation. In many cases however this will not be necessary as the number of tubes in the angle made by any two arms of the spider increases from the center toward the periphery and this automatically provides for a greater proportion of cooling gas bubbles per tube in the center. In fact this effect may in many cases be more than sufficient to counter balance any cooling due to the converter shell and it may be necessary, particularly where the number of spider arms is relatively small, to actually provide a larger number of holes at the ends of the spider arms. The arrangement of holes and their size will, of course, be determined for any particular installation and can be readily computed by the skilled engineer from the exotherm of the reaction and the dimensions of the converter. The invention, of course, is not limited to a uniform cooling effect, but in most cases this is desirable and may be considered as a preferred embodiment of the invention.

The distribution of the cooling gas is greatly facilitated by the fact that the gas bubbles rapidly passing through the bath produce an effective churning so that even without elaborate distributor piping a satisfactory uniformity of cooling may be achieved. The drawing which shows a distributor spider is of course merely an illustration of a typical embodiment and any other design of distributor may be used; for example, a number of concentric rings of perforated pipe may be used where extreme uniformity of gas distribution is desired, or any other suitable shape may be adopted as called for by the conditions under which the converter is to operate.

The drawing in its purely diagrammatic form illustrates the principle under which the invention operates. The precise structural details of the converter installations will vary with different converters and different reactions and while it is preferable to recover the heat of the cooling medium and make it useful as by the steam boiler as shown in the drawing, this, of course, is not essential to the invention and if desired, the gaseous medium may be cooled by ordinary air coolers, or it may be used to preheat incoming gases or for any other purpose. In cases where the nature of the non-boiling bath permits, air may be used and after passing through the boiler 10 it may, if desired, be exhausted to the atmosphere. This makes for a somewhat cheaper apparatus but it is less desirable from an operating standpoint, since the temperature of the incoming air would then vary with the temperature in the building and less uniform control would be achieved. It is also normally undesirable to blow gases into the bath at too low a temperature especially where the reaction gases pass down through the catalyst tubes, as in such a case there is the danger that the lower portions of the tubes may become too cool to continue the reaction and a closed circuit fluid recirculation of the cooling medium is normally preferable. Of course, when steam is used, it does not have to be recirculated for this reason as the inlet temperature is determined by the steam pressure and where there is a use for low pressure steam for heating purposes, it may be desirable to use fresh steam as a cooling means. This procedure however requires very clean steam as of course any solid impurities accumulate in the bath 5 or impurities such as oil are likely to be decomposed by the hot bath. The particular type of cooling gas or vapor circuit adopted therefore will necessarily be chosen in accordance with the other factors obtaining in the particular plant.

In the drawing round tubes have been shown as catalyst compartments and for many purposes this type of converter construction presents many advantages. It should be understood however that the catalyst compartments or tubes may be of any shape, such, as, square, hexagonal, oval, etc., and in some cases elongated rectangular tubes or catalyst compartments may be desirable and are of course included. The expression tubes as used in the claim being of course a generic expression and including catalyst compartments or tubes of various shapes.

What is claimed as new is:

A catalytic apparatus comprising a tubular converter, catalyst in the tubes, a non-boiling liquid bath surrounding the tubes, means for introducing the gaseous cooling medium into the lower portion of the bath and means for removing the same from the upper portion, the gas introducing means being non-uniform in a plane normal to the catalyst tubes, the non-uniformity being such as to provide for the introduction of a quantity of the cooling gas increasing from periphery toward the center of the plane.

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