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ABSTRACTO

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l.
                The present invention relates to the catalytic
      synthesis of materials through high pressure catelytic
  24
  3.
      reaction. More specifically, the invention comprises a
      novel process and apparatus employed in the manufacture
  Б.
      of Valuable products by direct synthesis from their com-
      ponent gases which are passed at high temperatures under
      pressure over a catalyst mass, as for example, the
 7.
     manufacture of methyl alcohol or methanol from carbon
 9.
     monoxide and hydrogen, or the production of synthetic
10.
     ammonia from a gaseous mixture of nitrogen and hydrogen
     in their combining proportions. In any instance the
11.
12.
     elements should be combined in their respective propor-
     tions in the gaseous mixture and then compressed to
13.
     relatively high pressures and passed under such pressure
.14.
15.
     over a suitable catalytic agent capable of stimulating
     the combination of the two gases whereby sufficient heat
17. is produced to maintain the temperatures required for
18.
    satisfactory yields.
19.
               The art of synthesizing methanol has now become
£0.
     well established in the literature. Among the United
     States patents upon the subject that may be ments oned
21.
22.
     are patents 1,558,559; 1,608,645; 1,609,593; 1,624,924;
     1,624,925; 1,624,926; 1,624,987; 1,624,928, and 1,624,929.
23.
24.
               When a mixture of hydrogen with carbon monoxide
     and carbon dioxide, or a mixture of the two oxides, is
25.
26.
     passed over a catalytic mass comprising a mixture of
     metals or their oxides at a pressure in excess of 100
27.
28.
     atmospheres and at a temperature of about 350-450° C.,
Ž9.
    methanol is produced. In case pure carbon monoride is
     employed, the product obtained will be practically pure
30.
```

- 1. methanol, but in case of the dioxide, a molecule of water
- 2. is produced for each molecule of methanol formed. In
- 3. practice, all of the gaseous mixture does not react on
- 4. the first contact, and the residual, unreacted gases are
- 5. circulated again and again over the catalyst, the reaction
- 6. product being cooled each time to separate out the
- methanol (or methanol and water) in liquid form.
- 8. Best results are obtained when the proportion
- 9. of hydrogen present is in excess of the amount theoret-
- 10. ically required to react with the carbon oxides present,
- 11. but a strict proportioning of the ingredients present is
- 13. not necessary to the success of the process. The emount
- 13. of gases converted to methanol on each passage through
- 14. the catalyst will depend upon the catalyst activity, the
- 15. temperature of the reaction, the space velocity and a
- 16. number of minor factors.
- 1?. The present invention relates to a methanol
- is. process and apparatus which is not limited to the use of
- 19. any specific estalyst or gas proportion. It relates to
- 20. a method and apparatus by which the methanol catalyst is
- 21. kept at a uniform temperature, and by which the necessary
- 22. heat is supplied by the exothermic reaction itself. The
- 23. heat given off by the reaction is acceserved within the
- 24. reaction vessel where it is used to heat the incoming
- 25. raw materials and also to meintain the catalyst temperature.
- 26. This type of process is known as an "autothermal process".
- 27. Another advantage of the invention lies in the
- 26. close control of the catalyst temperature which is made
- 29. possible by the process and apparatus herein set forth.

Jet 3 18691

- I. The optimum reaction temperature is in the neighborhood
- 2. of 400° C. and when any part of the catalyst reaches a
- 3. temperature much in excess of 400° C., the reaction
- 4. taking place at that point no longer produces pure
- 5. methonol, various undesirable by-products being obtained.
- 6. In addition to this bad effect, a worse one is likely to
- 7. occur, namely, that a temperature much in excess of the
- 8. optimum reaction temperature may also destroy the activity
- 9. of the catelyst itself.
- 10. On the other hand, if the catalyst, or any part
- 11. of it, is cooled from the optimum temperature, the
- 12. methanol process will operate inefficiently, and the
- 13. degree of conversion will be less than the maximum
- 14. possible. For the reasons outlined, it is important
- 15. that every portion of the catelyst be maintained at
- 16. exactly the optimum reaction temperature. The invention.
- 17. provides a means and process for doing this.
- 18. As previously stated, the reaction of hydrogen
- 19. with carbon oxides to produce methenol is an exothermic
- 20. one. The reaction of pure carbon monurate with hydrogen
- 21. is more highly exothermic than the reaction of carbon
- 22. dioxide with hydrogen. Various figures have been given
- 25. In the literature which purport to give the heat
- 24. generated by the two reactions:
- 25. CO + SE2 = CH3OH, and
- 86. 602 + 3H₂ = CH30H + H₂0
- 27. and while the specific figures given may be open to
- gs. question the basic fact that more heat is given off in
- 29. the case of the reaction of carbon monoxide is not open
- 80. to dispute.

- 1. Hence it might appear at first glance that 2. autothermal operation could be much more easily achieved 3. in the case of the first instance than in the second. However, the industrial operation of the methanol process 4. 5. is not based on the use of pure gases. No matter whether pure carbon monoxide or dioxide is introduced into the б. 7. process, in the circulating system itself both gases 8. will be present owing to various side reactions. Further, the reacting gases are not introduced in precise 10. molecular proportions, and consequently there is slways some extra hydrogen present which absorbs heat from the 11. 12. catalyst or each passage through it and gives up that 13. heat when the reaction product is cooled to condense out 14. liquid methanol. For the reasons mentioned, the practical difficulties of achieving autothermal operation and close 15. 16. control of catalyst temperature are of the same order 17. no matter whether earbon monoxide or carbon dioxide is 18. used as the principal carbon exide entering the reaction though autothermal operation is rendered easier in the 19. 20. case of carbon monoxide by the somewhat greater heat of 21. reaction. 22. The present invention provides means whereby 23. the temperature of the catalyst is controlled so that 24. the optimum reaction temperature is maintained while
- 26. The nature of this present process and apparatus 27. may best be understood in connection with the drawings 28. forming a part of this specification.

overheating of the catalyst is prevented.

29. In the arewings,

25.

30. Fig. 1, represents a cross-sectional elevation 31. of the converter taken through its center;

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- 1. Fig. 2, is a sectional view of the converter
- 2. taken on the line 2--2 of Fig. 1, and
- 5. Fig. 5, is a similar view taken on the line
- 4. 3--3 of Fig. 1.
- 5. Identical reference numerals are used through-
- 6. cut the several views.
- 7. In Fig. 1, the reference numeral 1 represents
- 8. the pressure resistant and corrosion resistant well of
- 9. the converter. In practice, this may be composed of
- 10. chrome-vanadium steel or some similar alloy, and may
- 11. be internally lined or plated with copper and chromium.
- 12. The member 1 is actually an elongated tube, both ends of
- 13. which are closed by similar structures. The top of the
- 14. member 1 is closed by a plug 2, which rests on small
- 15. shoulders on the internal well of 1. A pressure tight
- 18. joint is obtained by means of pressure exerted on the
- 17. plug Shoulders by means of a ring 3, which is thread
- 18. connected to the tube 1. The ring 5 receives the pressure
- 19. screws 3a, which bear against the shoulders on the plug :
- 20. 2.
- 21. The bottom of the tube 1 is similarly closed
- 22. by the plug, or lower head, 4, which also engages smell
- 25. shoulders on the internal wall of 1, similarly as de-
- 34. scribed above, and again a pressure tight joint is
- 25. obtained by means of pressure exerted upon the plug
- 26. shoulders by means of the ring 5, which receives the pres-
- 27. sure of the screws 5a, bearing against the shoulders on
- 28. the plug 4. The ring 5 is thread connected to the tube
- 29. 1; all similar to the construction already described.
- 30. The lower and upper plugs or heads are thus held in
- 31. engagement with the gaskets 6 and 7, respectively, as

- 1. shown in Fig. 1, the action of these gaskets contributing
- 2. to form pressure tight joints under the action of the
- 3. pressure applied to the respective heads.
- 4. While the tube 1 is actually a long integral
- 5. part, the drawing has been shortened as will be seen
- 6. from an inspection thereof.
- 7. The plug has firmly secured to it in any con-
- 8. venient manner, the member 8, serving as a supporting
- 9. flange for the structure to be hereinafter described.
- 10. Inside the tube I, and supported by the flange
- II. 8, there is positioned a structure in the form of another [4]
- 12. tube and indicated generally by 8a. This structure,
- 13. hereinafter referred to as a "bomb", may consist of
- .14. copper, or of alloy steel plated with copper or chromium.
- 15. Between the tube I and the bomb Sa, there is an annular
- 16. space 9a through which gases may travel as will be later
- [17. described. The bomb 8a is closed at both top and bottom.
- 18. but its interior is in communication with the space 2a
- 19. at the top of the bomb Sa, the communication being through
- 20. the ports 10a, these ports passing through a tube spacer
- 21. header functioning as hereafter described and forming the
- 22. top closure for the bomb Sa.
- 23. Within the bemb 8a, there is positioned a
- 24. catalyst banket, catalyst, and a heat exchanger. The
- 25. component parts of these may be best understood in their.
- 28. relation to the chemical process which takes place within
- 27. the converter. For this reason, in further describing
- 28. the apparatus and process it will be assumed that the
- 29. catalyst is at the optimum reaction temperature and that
- 30. a mixture of hydrogen and carbon oxides is passing
- 31. through this converter and contacting with the datalyst
- 52. where a portion of the gas reacts to form methanol, and
- 33. that the cooled residuel hydrogen and cerbon oxide
- 34. mixture is being added to the raw gases passing into the
- 35. bomb. 6

- 1. As will be seen from Fig. 1, the bomb 3a is
- 2. conveniently formed of two sections, one of which contains
- 3. the catalyst and basket and is therefore the reaction
- 4. chamber, while the other section serves as a preheater
- 5. for the reactants.
- 8. Referring to the preheating or heat exchanging
- '7. chamber, it will be seen that this is defined by a shell
- 8. 9 provided with an annular flange 10, and containing a
- 9. plurality or nest of heat interchanger tubes 11 and a
- 10. plurality of baffles 12 and 13, the baffles 12 having
- 11. center openings while baffles 13 are side opening. It
- 12. will be seen from Fig. 1 that the lower end of shall 8
- 13. is closed by the supporting flange 8, and the shell and
- 14. flange are firmly secured to each other, as for instance,
- :15. by welding, and the flange and shell are conveniently
- 16. formed of the same materials or of materials having
- 17. approximately the same coefficient of thermal expansion
- .18. so that the union between the shell and flange will be
- 19. maintained under all conditions. The tubes il are main-
- 20. tained in proper spaced apart relation by the headers 14
- 21. and 15, through which the tubes pass.
- 22. It will be seen that the header 14 is spaced
- 23. from the flange 8, and that the flange and plug 4 are
- 34. provided with registering passageways extending through
- 25. each of these members, thus forming a continuous passage-
- 26. way or opening communicating with the interior of the
- 27. heat exchanger. The gas inlet pipe 16 passes through
- 28. this opening, and also through the header 14, the inlet
- 29. pipe 16 discharging incoming gas into the heat exchanger
- 30. around the tubes Il, there being a continuous empular
- 51. passage 17 between the inlet pipe and the flange 8 and
- 32. plug 4, this passage serving as a conduit to lead the
- 33. reaction products from the heat exchanger to the outlet

- l. It will therefore be seen that the incoming reactant
- 2. mixture discharges from the pipe 16 in close proximity to
- 3. the heat exchanger tubes 11 and envelops these tubes as
- it passes to the reaction chamber, and is warmed as it
- passes because of heat exchange with the hot reaction
- б, products passing interiorly of the tubes 11 from the
- reaction chamber to the discharge, the baffles 12 and 13
- 8. causing the reactant mixture to follow a sinuous course.
- passing through the center of baffles 12 and outside 9.
- 10. around the circumference of baffles 13, thus being
- brought into intimate and efficient thermal contact with
- 12. the tubes 11. The baffles 12 and 13 are secured in
- 15. position by any convenient way, as for example, by brazing
- them upon the tubes 11. 14.
- 15. The heat exchanger communicates with the re-
- 16. action chamber, which is defined by the shell 19 having a
- flange 19a corresponding to the flange 10 of the shell 9; 17.
- 18. When the bomb is assembled, the heat interchanger header:
- 19. 15 is secured between the flanges 10 and 19a. It will
- 20. be seen that the header 15 is provided with an opening
- 21 20, conveniently centrally located, and a flange 21,
- adapted to receive a flange 22 defining a passage 35 .22.
- ₽3. through the header 24, which acts as a header for the
- catalyst tubes as will be hereafter mentioned, the flenges 24.
- 21 and 22 being secured together in any desired manner :25.
- as by bolting or riveting, the passages 20 and 23 28.
- 27. registering with each other to form a continuous conduit
- .85 for the passage of the reactants from the heat exchanger
- 29. to the reaction chamber. The reaction chamber contains
- a plurality of tubes 25 to hold the catalyst 26, the 80.
- 31. catalyst being of such a character as will favor the
- desired reaction. The tubes 25 are held in the desired 52.
- position by the top spacer header 27 and the bottom header

- 1. 24, the catalyst 26 being retained in the tubes by the
- 2. grid 28. The reaction chamber is also provided with
- 3. baffies 29 and 30, entirely analogous to the baffles 12
- 4. and 13 in the heat exchanger.
- 5. In view of the fact that the reaction is an
- 6. exothermic reaction, it is found desirable to control
- 7. the temperature of the catalyst to prevent overheating
- 8. of the same. For this purpose, means are provided for
- 9. introducing cold resetant mixture into the gas passages
- 10. outside the catalyst tubes. This is accomplished by the
- 11. provision of a plurality of by pass inlet tubes such as .
- .12. Indicated at 31 and 32, the discharge ends of these
- 13. tubes being so positioned with respect to the cetalyst
- 14. tubes that overheating throughout the catalyst mass is
- 15: effectively prevented. For example, some of the inlet
- 16: tubes may discharge cold gas near the top of the catalyst
- 17. tubes, others at the bottom of the catalyst tubes, and
- 18. still others in intermediate positions, and it will be
- 19. obvious, of course, that the number of these inlet tubes
- 20. may be multiplied as many times as needed. There is also
- 21. provided a by pass inlet for cold reactants indicated at
- 25. 35, permitting the introduction of cold reactants into
- 23. the space 9a between the shell 1 and the bomb 8a, this
- 24. also exerting a cooling effect upon the contents of the
- 25. bomb and on the converter wall. All gases introduced
- 26. Within the converter shell and bomb pasa through the
- 27. catalyst so that both the cold gases introduced for
- 28. cooling and the preheated gas entering the reaction
- 29. chamber by way of the heat exchanger pass through the
- 30. catalyst and are converted into the desired products.
- 31. The temperature is determined by means of pyrometers
- 32. located in tubes placed in desired positions in the re-
- 33. action chamber. Such tubes are indicated at 34, 35 and 36.

- If desired, core rods 37 may be inserted in
- 2. the heat exchanger tubes 11, so as to serve to regulate
- 5. the heat exchange, or this may be accomplished in other
- 4. ways as by regulating the velocity of the gases passing
- 5. through the converter.
- 6. It may here be noted that the inlet tube 16
- 7. passes through a stuffing box 38 secured in pressure-
- 8. tight relation with the plug or head 4 through the
- 9. agencies of the pressure screws 59 and gasket 40. The
- 10. gas outlet and each of the by pass inlets are provided
- 11. with members such as shown at 41 which serve as connection
- 12. members to connect the outlet and by pass inlets with
- 13. lines leading to storage and supply, respectively. Zach
- 14. of the connection members is maintained in gas tight
- 15. relationship with the converter shell by the action of
- 16. pressure screws, such as 42, and gaskets, such as illus-
- 17. trated at 43. These members are screw threaded to
- 18. receive their respective lines.
- 19. The course of the gases will be appearent from
- 20. the drawings, the solid arrows indicating reactants, while
- 21. the broken arrows indicate the reaction products.
- 22. To summarize, therefore, it will be seen that
- 25. the converter shell I is made pressure tight by the
- 24. upper head 2 and lower head 4 held in place against the
- 25. gaskets 6 and 7 by the pressure of the retaining rings
- \$6. 3 and 5. The retaining rings have buttress threads as
- 27. indicated on the drawing.
- 38. In the heat interchanger, the tubes 11 are
- 29. expanded into the headers 14 and 15. If desired, core rods
- 30. 37 may be placed in these tubes to increase the gas
- 31. velocity through them, or this velocity may be regulated

- 1. externally of the converter. The velocity of the gas pass
- 2. ing on the outside of these tubes is accelerated by means
- 5. of the baffles 12 and 13. The gas passes through the
- 4. center of the baffles 12, and outside around the circum-
- 5. ference of buffles 13.
- The oatalyst basket contained in the shell 19
- 7. consists of a number of tubes four to eight inches in
- 8. diameter which are expanded into the tube header 24. At
- 9. the bottom of each of these tubes 25 there is placed a
- 10. catalyst supporting grid 28. On the outside of the tubes
- 11. there are gas baffles as in the case of the heat inter-
- 12. changer itself. Pyrometer tubes 34, 35 and 36 are
- 13. located at convenient points on the outside of these
- 14. catalyst tubes. In addition to those shown at least
- 15. one pyrometer should be inserted in one of these catalyst
- 18. tubes.
- 17. The novel features of this design include the
- 18. method and means of controlling the temperature within
- 19. the catalyst. This converter is intended in particular
- 20. for use with cerbon monoxide-hydrogen mixtures where the
- 21. problem is that of removal of heat from the catalyst
- 22. rather than that of preheating the entering gases. Most
- 25. of the entering gases will enter through the main gas
- 24. inlet 16 and will be heated up during its passage through
- 25. the heat interchanger by thermal contact with the reacted
- 26. gases passing counter-ourrently inside the tubes. These
- 27. warmed entering gases are then passed on the outside of
- 28. the catalyst tubes 25, being further heated up. They may
- 29. be combined with by passed cold gases entaring through 31.
- 50. The temperature on the outside of the catalyst tubes at
- 31. the bottom will be controlled at some predetermined point
- 52. as indicated by the pyroneter 35. Again these gases may

- 1. be further diluted with cold gas entering through the
- 2. port 32 which terminates at a point about half way up the
- 3. catalyst bed. The temperature at this point is again
- levelled off to decrease the gradient within the catalyst.
- 5. There is a third by passed gas inlet 35 provided at the
- 6. bottom of the converter. The cold gas passes through the
- 7. annular space described by the shells 9 and 19 and the
- 8. shell 1. Heat is picked up from the well which would
 - 9. otherwise be lost by radiation and the temperature of the
- 10, combined gases entering the catalyst controlled as desired
- 11. by regulation of the amount of gas entering at 35.
- 12. The reacted gases pass down through the tubes
- 13. 11 and cut of the converter through the outlet 18.
- 14. Although the converter herein set forth is
- 15. illustrated and described as being installed in a vertical
- 16. position, it will be understood that the angle and plane
- 17. of installation is not material to the invention, and
- 18. it will elso be apparent that many details of the con-
- 19. struction may be medified without departing from the
- 20. inventive concept.
- 21. The direction of the flow of the gases through
- 22. the converter is indicated by the arrows on the drewings.
- 23. The course of the reaction which permits autothermal
- 24. operation and accurate control of the catalyst tempera-
- 25. ture consists in heating the cool incoming gases by
- 26. thermal contact with hot gaseous reaction products. This
- 27. heating increases the temperature of the incoming gases
- 28. very substantially, but does not heat them to the optimum
- 29. reaction temperature. The heated gases are then passed
- 30. in thermal contact with the catalyst itself (where the
- 51. exothermic reaction is taking place), and this thermal

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- 1. contact not only raises the temperature of the incoming
- 2. gases substantially to the optimum reaction temperature,
- 3. but also serves to control the temperature of the catalyst
- 4. itself in two ways:
- I. To abstract heat from the catalyst so that
- 8. its temperature will not rise above the desired reaction
- 7. optimum;
- B. 2. To heat up any portions of the catalyst
- 9. which may tend to be decreased in temperature much
- 10. below the reaction optimum.
- 11. In practice, sufficient heat is generated by
- 12. the excthermic reaction taking place within the catelyst
- 13. to provide more than enough heat to warm the incoming
- 14. gases and to maintain the catalyst temperature when the
- 15. process and apparatus are operated at practical space
- 18. velocities, i.e., about 10,000 20,000. In fact, an
- 17. excess of heat is usually generated. To control the
- 18. temperature of the catalyst it is necessary to dilute
- 119. the hot gases from the heat interchanger with cold gas,
- 20. so that the mean temperature of the catalyst will not be
- 21. raised too high. The cooling means have previously
- 22. been described, but it may be here pointed out that the
- 23. present installation is distinctive in providing of such
- 24. cooling means outside of the catalyst, and circulating
- 25. the entire cooled gas mixture through the entire mass of
- 26. catalyst. To accurately observe the temperature within
- 27. the converter, pyrometers are located at convenient
- 28. points, as above explained, and the supply of cold gas
- 29. may be regulated manually or by suitable electrical con-
- 30. nection to the indicating pyrometers.
- 31. In the foregoing description of the invention, it
 - 32. has been assumed that the process and apparatus was in

- 1. operation and that the catalyst was at the proper reaction
- 2. temperature. Actually, in starting up the process and
- 3. apparatus, it is of course necessary to provide some out-
- 4. side source of heet. The process and apparatus may be
- 5. put into operation by supplying previously heated gases
- 6. to inlet 16. The simplest procedure is merely to pass
- 7. the mixture of hydrogen and carbon oxides through some
- 8. suitable gas heater and to convey them hot into the
- 9. converter. On account of the massiveness of the apparatus,
- 10. the heated gases must be passed into the converter for a
- 11. considerable period before it is raised to reaction tem-
- 12. perature, and as soon as the catalyst has been raised in
- 13. temperature to a point where the methanol reaction starts,
- 14. the heat of the reaction also assists in reising the
- 15. converter temperature provided that no cold gas is
- 16. passed into the converter.
- 17. In place of supplying hot gases to the converter.
- 18. it is of course possible to heat the catalyst sufficiently
- :19. to start the methenol reaction by means of an electric
- 20. heating element which may be positioned within the con-
- 21. verter and in contact with the catalyst.
- 22. The improved process and apparatus, as above
- 23. described, may be employed for the production of synthetic.
- 24. methanol with various catalysts and gas mixtures already
- 25. known in the art. For example, when a gas mixture com-
- 26. prising 10% of carbon dioxide and 90% hydrogen is passed
- 27. through the apparatus at a space velocity of 12,000 and
- 28. at a reaction temperature of 400° C. in contact with a
- 29. catalyst of the type described in Woodruff and Rloomfield's
- 20. U. S. Patent 1,625,929, i.e., a mixture of zinc oxide,
- 31. chromium oxide, iron oxide, and zinc chloride -- there is

- 1. produced an hourly yield of methanol amounting to about
- 2. 6.5 gallons per cubic foot of catalyst together with an
- 5. equivalent amount of water. If the gas mixture supplied
- 4. to the process and apparatus consists of 10% carbon
- 5. monoxide and 90% hydrogen, the other conditions remaining
- 6. the same, the hourly yield of the methanol is about 10
- 7. gallous per cubic foot of catalyst.
- In the place of the catalyst mentioned, other
- 9. catalysts such as those described in United States
- 10. patents 1,625,924; 1,625,925; 1,625,926; 1,625,927;
- 11. 1,625,928, and 1,825,929, may be employed.
- 12. While the apparatus has been described particu-
- 15. Larly for use in the synthesis of methanol from carbon
- 14. oxides and hydrogen, it may obviously be employed for the
- 15. catalytic manufacture of other materials such, for
- 18. instance, as ammonia, higher alcohols, synthol, etc.,
- 17. and it will be apparent that many modifications of the
- 18. details of the construction of the converter herein
- 19. illustrated may be made without substantially affecting
- 20. the essentials of the construction threof, and it will
- 21. be understood that It is therefore desired to comprehend
- 22. within the scope of the invention such modifications as
- 23. may be necessary to adapt it to varying conditions and
- 34. uses.

25. What is claimed is:

- 26. In an apparatus for carrying out exothermic
 - 27. chemical reactions, the combination within a converter
 - 25. shell of adjacent inter-communicating heat exchanger and
 - 29. reaction chembers, an inlet opening into the heat ex-
 - 50. changer for introduction of cold reactants therein, a

communicating passage for passing said heated reactants. 2. from the heat exchanger to the reaction chambers, means з. for passing heated reactants in thermal contact with all the catalyst contained in the reaction chamber, a plurality of inlets for introduction of cold reactant 5. 6. into the stream of said hot reactants at various points 7. exteriorally of the catalyst, means for passing the mix-8. ture of said reactants through all of the catalyst, and 9. means for passing the reaction products through the heat 10. interchanger in thermal contact with the reactants. ll. In an apparatus for carrying out exothermic chamical reactions the combination with a ponverter shall 12. 15. of adjacent intercommunicating heat exchanger and reaction 14. chambers within the shell and spaced apart therefrom, **15.** thereby defining an annular space between themselves and 18. shell, the reaction chamber being adapted to contain a 17. catalyst, an inlet opening into the heat exchanger for the introduction of reactions therein means for passing the heated reactant from the heat interchanger throughout the **120** . . reaction chamber in thermal contact with the catalyst. a 21, plurality of inlets for admitting portions of cold re-22. actants into thermal contact with the catalyst and into the stream of said heated reactants, means for passing all of said reactants in direct contact with the catalyst. means for passing the reaction products through the heat 86. interchanger in thermal contact with the reactants, means for introducing cold reactants into the said annular space between the shell and heat interchanger and into

contact with the catalyst.

```
1.
                    In an apparatus for carrying out exothermic
     oficial reactions the combination with a converter shell,
 2.
 3.
      of adjacent intercommunicating heat exchanger and reaction
      chambers within the shell, means in the reaction chamber
 4.
      adapted to contain a catalyst, an inlet opening into the
 5.
 в.
     heat exchanger for introduction of reactants therein,
 7.
     means for passing the said reactants from the heat ex-
      changer throughout the reaction chamber in thermal contact
 8.
     with the catalyst and then in direct contact therewith,
 9.
×10.
     means for passing reaction products through the heat
      exchanger in thermal contact with and counter-currently
11.
12.
      to the incoming reactants, a plurality of by-pass reactant
13.
     inlets opening into the reaction chamber externally of
14.
      the catalyst and distributed for efficient cooling of the
15.
      said catalyst, the said inlets being adapted to introduce
      cold reactants into the reactants coming from the heat
18.
17.
      exchanger and means for passing the entire resulting re-
18.
     actant mixture through the entire mass of satalyst.
                4. In an apparatus for carrying out exothermie
19.
     shemical reactions the combination with a converter shell
20.
21.
     of a reaction chamber and means to preheat reactant with-
     in the shell, means within the reaction chamber for con-
22.
      taining a cetalyst, means for passing heated reactants in
     thermal contect with the catelyst and then in direct con-
24.
25.
     tact therewith and a plurality of inlets opening externally
      of the catalyst containing means but adjacent thereto
26.
     and adapted to discharge cold reactant into the heated
27
     reactant to threeby prevent overheating of the catalyst
      and means for passing the entire resulting reactant
     mixture through the entire mass of catalyst.
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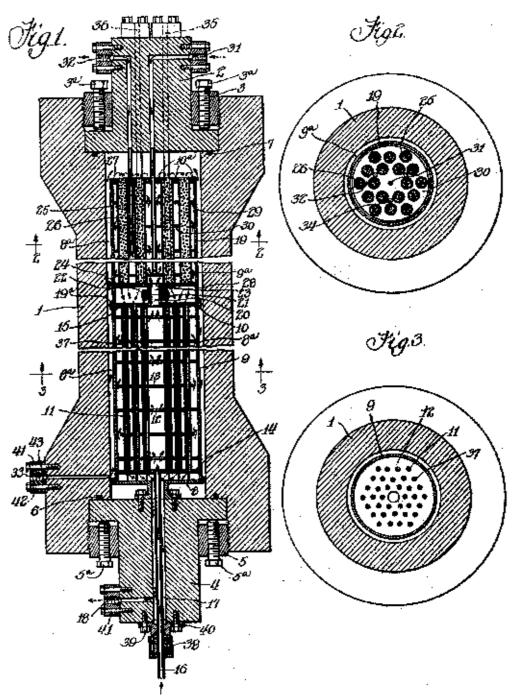
```
In an apparatus for producing methanol from
  Į.
      carbon oxides and hydrogen, the combination with a con-
  г.
      erter shell, of a reaction chamber and heat exchanger in
      communication with each other and spaced apart from the
  4.
  ₽.
      shell, and forming an annular chamber therewith, a
      plurality of catalyst containing tubes in the reaction
  6.
      chamber, the said tubes communicating with the said annu-
7.
  8.
      lar space and being also in communication with the heat
  9.
      exchanger, means for introducing reactant gaseous mixture
 10.
      into the heat exchanger in thermal contact with outgoing
 11.
      reaction products, means for passing the heated mixture
 12.
      into the reaction chamber, a plurelity of inlets opening
      into the reaction chamber externally or, but adjacent to.
 13.
 14.
      the catalyst tubes and adapted to discharge cold reactant
 15.
      gaseous mixture into the heated mixture coming from the
 16.
      heat exchanger, an inlet for introducing cold reactant
 117.
      gaseous mixture into the annular space between the shell!
      and the reaction chamber and heat exchanger and means
 18.
      permitting the passage of the entire gaseous reactant
, as.
 · 0$
      mixture in through the entire mass of catalyst.
 21.
                6. In an apparatus for synthesizing methanol
 es.
      through the catalytic reaction between hydrogen and carbon 🥎
 25.
      oxides, the combination with a converter shell of preheat-
 24.
      ing and reaction chembers within the shell and defining an
 25.
      annuar space therewith, means for passing heated reactant
 28.
      gases from the preheater into the reaction chamber, catalys ?
 27.
      containing means within the reaction chamber, inlets open-
 ž8.
      ing into the reaction chamber externally of, but adjacent
      to the catalyst containing means and adapted to discharge.
      cold gaseous reactants into the heated reactants and
 21. against the catalyst containing means, an inlet opening
```

- 1. into the annular space between the shell and reaction and
- 2. preheating chamber and adapted to discharge cold gaseous
- 3. reactants therein and means permitting the passage of all
- 4. gaseous reactant through the entire mass of catalyst.
- 5. 7. In an apparatus for synthesizing methanol
- 6. through the catalytic reaction between hydrogen and
- 7. carbon exides, the combination with a converter shell of
- 8. preheating and reaction chambers within the shell and de-
- 9. fining an annular space therewith, means for passing
- 10. heated reactant gases from the preheater into the reaction
- 11. chamber, catalyst containing tubes within the reaction
- 12. chember communicating with the said annular space and
- 13. adepted to discharge hot reaction products into the pre-
- 14. heater, inlets opening into the reaction chamber at
- 15. staggered depths therein externally of but adjacent to
- 16. the catalyst tubes and adapted to discharge cold gaseous
- 17. reactants into the heated reactants from the preheater,
- 18. means for introducing cold resotent gas into the annular
- 19. space between the shell and reaction and preheating
- 80. chambers and means permitting the passage of all the
- 21. reactant gases through the entire catalytic mass.
- 22. 8. The process of carrying out exothermal
- 25. reactions which comprises subjecting a preheated reactant
- 34. mixture to the action of a catalyst, cooling the said
- 25. mixture prior to its contact with the catelyst by
- 26. introducing into the said mixture quantities of cold
- 27. reactant mixture sufficient to prevent overheating of
- 28. the catalyst and then passing the whole reactant mixture
- 29, through the entire mass of catelyst.

- 9. The process of parrying out exothermal
 reactions which comprises subjecting a preheated reactant
 nixture to the action of a catalyst, maintaining the
- 4. catalyst at its optimum reaction temperature by the ex-
- 5. ternal application of sold resutant mixture and passing
- 6. the whole of the reactants through the entire mass of
- 7. the catalyst.
- 8. 10. The process of carrying out exothermal
- 9. reactions which comprises subjecting a hot gaseous reactant
- 10. mixture to the action of a catalyst while introducing
- 11. into said mixture prior to its passage through the cata-
- 12. lyst at points externally of but adjacent to the catalyst
- 13. in quantities sufficient to prevent overheating of the
- 14. catalyst, cold reactant mixtures and passing all of the
- 15. reactant mixture through the entire mass of the oatalyst.
- 16. Il. The process of producing methanol which
- 17. comprises subjecting a heated reactant mixture of carbon
- 18. oxides and hydrogen to the action of a methanol catalyst,
- 19. introducing into the heated mixture prior to its contact
- 20. with the catalyst, quantities of cold mixture sufficient
- 21. to prevent overheating of the catalyst and passing the
- 22. whole mixture through the entire mass of the catalyst.
- 23. 12. The process of producing methanol which
- 24. comprises subjecting a heated reactant mixture of carbon
- 25. oxides and hydrogen to the action of a methanol catalyst,
- 26. introducing into the heated mixture prior to its contact
- 27. with the catalyst and at points externally of but adjacent
- 28. to the catalyst, quantities of cold mixture sufficient to
- 29. prevent overheating of the catalyst and passing the whole
- 30. mixture through the entire mass of the catalyst.

- 1. 13. The process of producing methanol which
- 2. comprises subjecting a heated reactant mixture of carbon
- 3. oxides and hydrogen to the action of a mathanol catalyst.
- 4. cooling the catalyst through substantially its entire
- 5. extent by the introduction of cold reactant mixture into
- 6. the hot mixture at selected points external of but adjacent
- 7. to the catalyst and passing the whole reactant mixture
- 8. through substantially the entire mass of catalyst.
- 9. 14. The process of carrying out exothermal
- IC. reactions which comprises subjecting a prehéated reaction
- 11. mixture to the action of a catalyst while controlling
- 12. the cetalyst temperature by admission of cold gaseous
- 13. reactants into thermal contact with it end into the stream
- 14. of preheated gases prior to their passage through the
- 15. oatelyst.

Catalytic Apparatus 1. 30. 11. 35. 35. 35. 35. 35. 35. 35. 35.



Certified to be the drawing referred to in the specification hereunto armexed. It illian J. Colomb.

Jan 1929
Jerse Marie, Indiana, U.S.A.

134 January M.S.A.

ATTORNEY