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CHEMICAL PROCESS CONTROL

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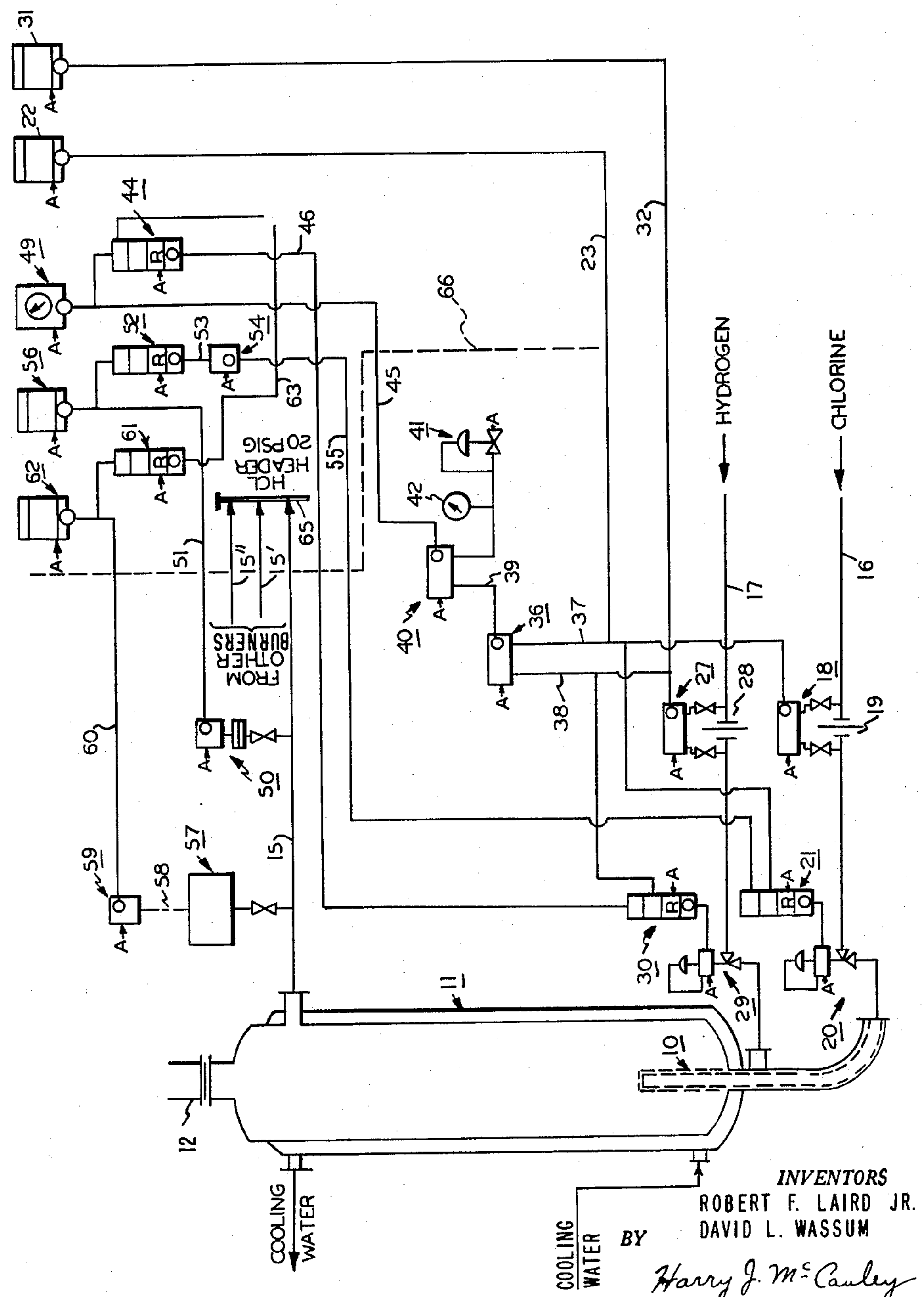

INVENTORS
ROBERT F. LAIRD JR.
DAVID L. WASSUM

BY

Harry J. McCauley
ATTORNEY

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CHEMICAL PROCESS CONTROL

Robert F. Laird, Jr., Bridgeton, N.J., and David L. Wassum, Wilmington, Del., assignors to E. I. du Pont de Nemours and Company, Wilmington, Del., a corporation of Delaware

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This invention relates to a method and apparatus for chemical process control, and particularly to a method and apparatus for the control of a chemical process operating on gaseous reactants and producing a gaseous form product.

Chemical processes, such as the manufacture of HCl by the combustion of H_2 with Cl_2, and the manufacture of HBr by the combustion of H_2 with Br_2, are typical of processes which utilize gaseous reactants and manufacture gaseous phase products. For considerations of safety as well as product quality, it is usually imperative that one of the reactants be supplied in a precisely regulated fixed ratio with respect to the other, independent of the existing rate of product manufacture, which latter usually varies from zero to 100% based on the demand of next-following downstream facilities. Moreover, it is desirable to obtain products at a steady, appreciably high, superatmospheric pressure, thus dispensing with downstream pumps and separate pressurized storage facilities. We have found, in addition, from experience with an HCl manufacturing system, that it is advantageous to conduct the combustion of gaseous phase reactants at substantial gage presures, because a more stable flame is thereby obtained, thus eliminating flame-out difficulties as well as obtaining improved product quality.

The effectuation of semi-automatic control as the resultant of a sensed product condition coupled with a type of proportioned ratio supply of reactants is taught in U.S.P. 2,909,413; however, operation is at near-atmospheric presure and, additionally, requires human intervention, particularly as regards the critical ratio control of reactants.

We have now found that it is possible to base control on product pressure solely or, if desired, on product pressure and product quality independently, which gives completely automatic control of a highly reliable nature. At the same time there is preserved freedom as regards human intervention, and there is obtained safe operation at steady appreciable gage pressures.

An object of this invention is to provide an improved method and apparatus for chemical process control which is, within reasonable limits, substantially independent of the pressure level of operation. Another object of this invention is to provide a method and apparatus for automatic chemical process control which has an extremely high reliability, quick response characteristics and a wide range of operational capability. The manner in which these and other objects of this invention are accomplished will become clear from the detailed description and the drawing, which is a schematic representation of a preferred control arrangement for the combustion manufacture of HCl according to this invention.

Generally, this invention consists of a method and apparatus for controlling the flow of a pair of gases introduced into a reactor, which gases interact one with the other to form a gaseous phase product within the reactor, comprising, in order of operations effected in the direction running from the sources of supply of said pair of gases towards said reactor, sensing the flow of a first gas of said pair of gases and applying a signal representative of the rate of flow of said first gas to one side of an instantaneous gas flow rate ratio determination balance, sensing the flow of a second gas of said pair of gases and applying a signal representative of the rate of flow of said second gas to the other side of said gas flow rate ratio determination balance, removing gaseous product from the reactor, sensing the existing pressure of the gaseous product and controlling the rate of supply of said first gas to preserve substantially a predetermined value of existing pressure of the gaseous product, and independently deriving from said gas flow rate ratio determination balance a signal representative of the existing ratio of flow of said second gas with respect to said first gas in control relationship of the rate of supply of said second gas by adjustment of the flow of said second gas of said pair of gases to preserve substantially a predetermined ratio of supply of said second gas with respect to said first gas.

Referring to the drawing, the combustion manufacture of HCl according to this invention is effected by burning hydrogen gas, supplied through the outer annular portion of conventional burner nozzle **10**, in vaporous chlorine, supplied through the central pipe portion of the burner nozzle. Burner nozzle **10** is mounted vertically and centrally of a water-cooled upright reactor **11**, provided at the top with an explosive pressure rupture disc **12**, gaseous HCl being withdrawn as product through line **15** leading to downstream facilities not shown.

In the following description, the chlorine, supplied to the process through line **16**, is arbitrarily considered to be the "first fluid," whereas the hydrogen, supplied to the process through line **17**, is thus the "second fluid," as these terms are employed in the appended claims. Typically, the chlorine flow rate varies from 0–2000 lbs./hr. of superheated (to about 35° C.) vaporous chlorine supplied at 40 p.s.i.g., the flow rate of which is controlled responsive to the commercial pneumatic flow rate transmitter indicated generally at **18**, which operates in accordance with a sensed pressure drop across orifice **19** disposed in chlorine line **16**. It is convenient to use pneumatic control equipment throughout, and this is indicated in block representation with control air inputs denoted by the letter A in all instances. The chlorine flow control valve denoted generally at **20** is located downstream from transmitter **18** and is actuated by commercial type controller **21** as hereinafter described. Controller **21**, as well as all of the controllers employed in the system detailed, is of the reverse-acting type, denoted by the letter "R" drawn in each individual controller block. Manual control of chlorine feed as well as the recording of the supply of this gas is effected by the flow recording control station **22** connected to transmitter **18** by a multiplicity of pneumatic lines, in this instance four in number, represented, however, as a single line **23** to simplify the showing.

The control of supply of hydrogen (typically furnished at a rate of 0–65 lbs./hr. at 40 p.s.i.g. and ambient temperature) parallels that of the chlorine, the hydrogen flow rate transmitter **27** being of the pneumatic type, obtaining its signal from orifice **28** mounted in hydrogen line **17**. The hydrogen flow control valve is of conventional type, indicated generally at **29** located downstream from orifice **28**, which is actuated by controller **30**. Manual control of hydrogen feed as well as the recording of the supply of this gas is effected by flow recording control station **31**, connected to transmitter **27** by a multiplicity of pneumatic lines, in this instance four in number, represented in single line convention at **32**.

A rather precisely controlled excess of hydrogen must be maintained in the off gas from reactor **11**, from the standpoint of both product quality and operating safety, and this is typically 1–2 mol percent. The volumetric gas flow rate ratio determination bridge employed in the system described herein was a Sorteberg Type "D" pneumatic force bridge **36** arranged for division such that the numerator is always smaller than the denominator. This, in conjunction with computing relay **40**, hereinafter described, provides a signal proportional to instantaneous flow ratio of extremely high sensitivity, i.e., the full scale ratio range for this application is typically 1.00:1 to 1.10:1.

As seen in the drawing, flow rate ratio bridge **36** receives its signal representative of chlorine flow through pneumatic line **37** running to flow rate transmitter **18**, and its signal representative of hydrogen flow through pneumatic line **38** running to flow rate transmitter **27**. Ratio bridge **36** supplies the input through line **39** to a conventional type of computing relay **40**, the reference pneumatic source for which is conventional pressure regulator **41**, which is provided with its own pressure gage **42** on the output side. Hydrogen-to-chlorine flow ratio control is effected through commercial controller **44**, receiving its input from computing relay **40** via pneumatic line **45** and transmitting its output via pneumatic line **46** to hydrogen controller **30**. The hydrogen-to-chlorine flow ratio indication control station is indicated at **49**, the controller for which has a pneumatic set for cascade control and a ratio range of 1.0:1 to 1.1:1.

The HCl product gas pressure transmitter **50** is tapped into product discharge line **15** and, typically, has a range of 0–60 p.s.i.g. This provides the pneumatic signal input, via line **51**, to HCl gas pressure controller **52**. The output of the latter is a pneumatic input to high and low limit pneumatic relay **54** via line **53**, which, in turn, applies the control reference input to chlorine controller **21** via line **55**. The HCl pressure recording station is indicated generally at **56**.

The apparatus as hereinbefore described is completely operational and can be utilized without further equipment; however, due to variations in purity and density of the gaseous reactants and the small inherent error of the volumetric flow measuring devices hereinbefore described, measurement inaccuracies can result. It is, therefore, preferred to superimpose an independent control in the form of a product purity check, which can adjust the ratio of the reactants supplied to maintain a preselected product purity. Thus, there is provided a thermal conductivity analyzer **57**, e.g., a Leeds & Northrup Co. Model 7802–D–A9–W17, adapted to measure the content of hydrogen (0–5% by volume range) in the HCl gas and generate an electrical control signal (0–20 mv. D.-C.) which is applied through the electrical conductor **58**, shown in broken line convention in the drawing, to the electric-to-pneumatic potentiometer **59**. The latter, in turn, supplies the pneumatic input via line **60** to the hydrogen-in-HCl analysis controller **61**. The hydrogen in HCl recording station is denoted generally at **62** and the connection impressing independent control as a function of gas analysis is via line **63**, furnishing a control reference input to the hydrogen-to-chlorine flow ratio controller **44**.

In large scale operation it is necessary to utilize more than one burner-reactor, and this is accomplished by providing duplicate arrangements each identical with that described, which are individually provided with output lines **15**, **15′** and **15″**, all discharging into a common header **65**.

In one installation provided with three 2000 lbs./hr. each maximum capacity burners, it was necessary to produce high quality HCl for use in an organic chlorination process which operated at superatmospheric pressure and which, therefore, required a steady HCl product supply at 20 p.s.i.g. regardless of the magnitude of demand for product. This was directly accomplished with the apparatus of this invention without resort to pumps or intermediate storage of any kind and, during sustained operation, it was ascertained that burner operation was at the same time substantially improved as regards both flame stability and also the combustion per se by the operation at higher pressure level. In this specific installation it was convenient to locate all instruments shown to the right of broken line **66** remote from the process on a common control room panel, and this was readily accomplished.

The installation hereinbefore described was provided with three HCl reactors discharging to a common header **65**; however, in the testing operation detailed, one reactor was maintained in continuous standby and only two were kept in service at any one time. In order to safeguard against operation of the chlorine flow controller **21** at possibly unstable extremes of its range, the pneumatic relay **54** for each reactor was set at a chlorine low limit flow rate control of 500 lbs./hr. and a high limit flow rate control of 1800 lbs./hr., and which proved entirely practicable.

The chlorine flow rate is immediately measured by orifice **19** and the flow rate information obtained by transmitter **18**, which delivers an output signal representative of the instantaneous rate of chlorine supply to one side of ratio determination bridge **36** and simultaneously to chlorine flow recording control station **22** via line **23**.

The hydrogen flow rate is measured by its own orifice **28** and the corresponding instantaneous flow rate signal sent on by hydrogen flow transmitter **27** via line **38** to the other side of flow ratio determination bridge **36**, and also to hydrogen flow recording control station **31** via line **32**.

Ratio determination bridge **36**, operating through computing relay **40**, delivers a continuous signal to the hydrogen-to-chlorine flow ratio control station **49** which, together with controller **44** connected in the same control loop therewith, resets the control point of hydrogen flow rate controller **30** via line **46**, and thus adjusts valve **29** appropriately, so as to maintain at all times the 1–2 mol percent excess of hydrogen in the HCl product gas.

The product HCl delivery pressure is continuously measured by HCl pressure transmitter **50**, which indicates the instantaneous existing pressure at recording control station **56** and adjusts the set point of chlorine controller **21**, adjusting valve **20** appropriately, via the control loop inclusive of controller **52**.

As previously stated, the hydrogen-in-HCl analyzer **57** is an optional auxiliary functioning entirely independent of the rest of the control apparatus. When used, this analyzer transmits a signal constituting a continual check on excess hydrogen content in the product via the control loop inclusive of hydrogen-in-chlorine recording station **62** and the associated controller **61** which, together, independently resets the control point of the hydrogen-to-chlorine flow ratio controller **44** to compensate for product composition variations actually determined.

The control system of this invention displays sufficiently high inherent stability to make it possible to bring the manufacturing process to a steady level of operation within a relatively short period of time. Operational records obtained at recording stations **22**, **31** and **56** revealed a surprisingly effective capability to adjust automatically to very large, instantaneous product demand changes. At the same time, experience obtained over a relatively long period of sustained operation has demonstrated a very high reliability, which indicates that the control is a marked improvement over other methods now available to the art.

It will be understood that this invention is not limited to combustion type reactions but, of course, is generally applicable to other types, including, specifically, catalyst-promoted gas phase reactions. Moreover, while control is effected on the basis of interrelationship between a given pair of gases, it will be clear that other reactants ingaseous liquid or solid phase can additionally be present, so long as the plan of control based on the two chosen gases is practicable for the over-all reaction involved.

From the foregoing, it is apparent that this invention can be modified in numerous respects without departure from its essential spirit, and it is intended to be limited only within the scope of the appended claims.

We claim:

1. A method for controlling the flow of a pair of gases introduced into a reactor, which gases interact one with the other to form a gaseous product within said reactor, comprising, in order of operations effected in the direction running from the sources of supply of said pair of gases towards said reactor, sensing the flow of a first gas of said pair of gases and applying a signal representative of the rate of flow of said first gas to one side of a gas flow rate ratio determination balance, sensing the flow of a second gas of said pair of gases and applying a signal representative of the rate of flow of said second gas to the other side of said gas flow rate ratio determination balance, removing gaseous product from said reactor, sensing the existing pressure of said gaseous product and controlling the rate of supply of said first gas to preserve substantially a predetermined value of said existing pressure, and independently deriving from said gas flow rate ratio determination balance a signal representative of the existing ratio of flow rate of said second gas with respect to said first gas in control relationship of the rate of supply of said second gas thereby adjusting the flow rate of said second gas of said pair of gases in response to said signal of said flow rate determination balance to preserve substantially a predetermined ratio of supply of said second gas with respect to said first gas.

2. A method for controlling the flow of a pair of gases introduced into a reactor according to claim 1 wherein said gaseous product is subjected to composition analysis, a signal is derived as a function of the existing composition of said gaseous product, and said signal is independently and additionally applied in control relationship of the set point of said gas flow rate ratio determination balance to bring said composition of said gaseous product to a predetermined level.

3. Apparatus for controlling the rate of flow of a pair of gases introduced into a reactor, which gases interact one with the other to form a gaseous product within said reactor, comprising in combination a gas flow rate ratio determination balance, means sensing the flow rate of a first gas of said pair of gases and transmitting a signal representative of the instantaneous rate of flow of said first gas as one input to said gas flow rate ratio determination balance, means sensing the flow of a second gas of said pair of gases and transmitting a signal representative of the instantaneous rate of flow of said second gas as the other input to said gas flow rate ratio determination balance, means responsive to the existing pressure of the gaseous product delivered from said reactor inclusive of a flow-regulating valve disposed downstream from said means sensing the flow of said first gas of said pair of gases controlling the rate of supply of said first gas to preserve substantially a predetermined value of said existing pressure, and means responsive to said gas flow rate ratio determination balance inclusive of a flow-regulating valve disposed downstream from said means sensing the flow of said second gas of said pair of gases independently controlling the rate of supply of said second gas to preserve substantially a predetermined ratio of supply of said second gas with respect to said first gas.

4. Apparatus according to claim 3 for controlling the rate of flow of a pair of gases introduced into a reactor, which gases interact one with the other to form a gaseous product within said reactor, provided with analyzer means connected with sample intake in open communication with the product delivery outlet of said reactor subjecting said gaseous product to composition analysis, and means responsive to said analyzer means adjusting the set point of said gas flow rate ratio determination balance to restore said gaseous product to a predetermined composition level.

References Cited in the file of this patent

UNITED STATES PATENTS

1,414,762	Townsend	May 2 ,1922
2,697,652	Ribble et al.	Dec. 21, 1954
2,909,413	Hildyard	Oct. 20, 1959