Sept. 3, 1974 H. KUERTEN ET AL 3,833,719

METHOD AND APPARATUS FOR MIXING GAS AND LIQUID

Filed Feb. 16, 1972

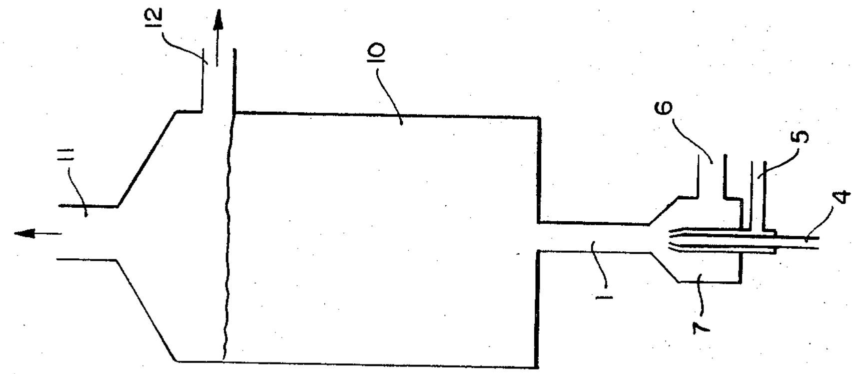


FIG. 3

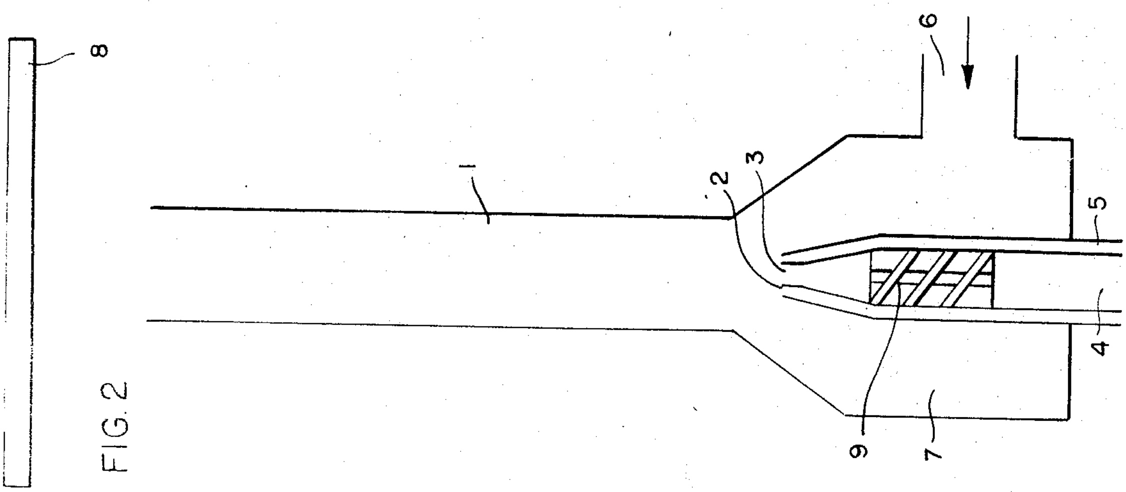


FIG. 2

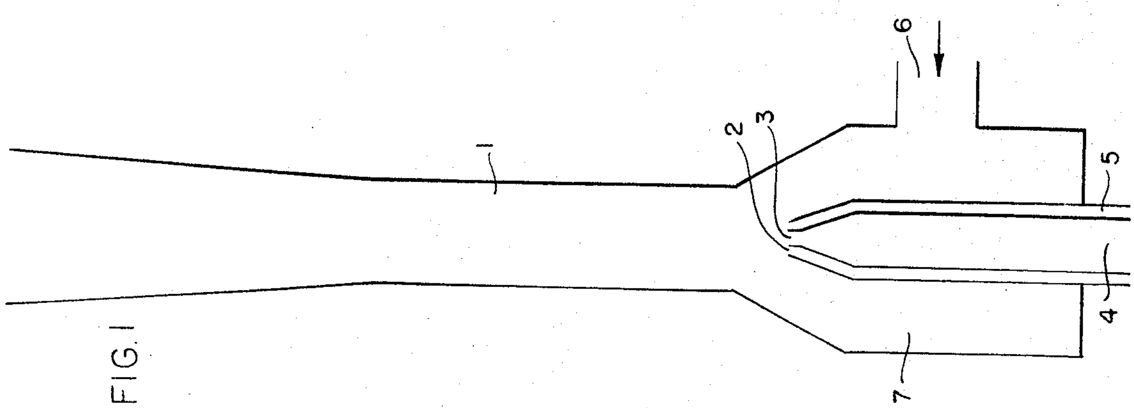


FIG. 1

# United States Patent Office

3,833,719
Patented Sept. 3, 1974





3,833,719


## METHOD AND APPARATUS FOR MIXING GAS AND LIQUID


Heribert Kuerten, Mannheim, Otto Nagel, Neustadt, Rolf Platz, Mannheim, and Richard Sinn, Ziegelhausen, Germany, assignors to Badische Anilin- & Soda-Fabrik Aktiengesellschaft, Ludwigshafen (Rhine), Germany



Filed Feb. 16, 1972, Ser. No. 226,808
Claims priority, application Germany, Feb. 19, 1971, P 21 07 960.7
Int. Cl. B01f *3/00*
U.S. Cl. 423—659 7 Claims



### ABSTRACT OF THE DISCLOSURE

The invention relates to a method of mixing a gas with liquids in a tubular reactor by feeding the gas and the liquids to a mixing zone. The invention also relates to an apparatus for carrying out this method. Rapid mixing is effected by feeding a stream of liquid to the mixing zone through one or more nozzles whose axes extend in the same direction as the axis of the mixing zone, the injected liquid having a velocity of from 5 to 100 m./s., whilst a second stream of liquid of much lower velocity is introduced into the reactor inlet zone surrounding said nozzles. The gas is fed to the mixing zone through one or more gas inlets located near the orifices of the liquid nozzles. The mean cross-sectional area of the mixing zone bears a specific ratio to the cross-sectional area of the orifices of the liquid nozzles and the length of the mixing zone bears a specific ratio to the hydraulic diameter thereof. The method and apparatus are particularly suitable for carrying out reactions in which short residence times are desirable and the reaction products must not recontact the starting materials.


The present invention relates to a method and apparatus for mixing gas and liquid in a tubular reactor.

In a number of chemical reactions involving gases and liquids, the mixing operation constitutes an important factor. In industry, mixing is generally effected by means of mechanically driven stirrers of various designs. However, leaks inevitably occur at the stirrer shaft, particularly in reactions carried out under pressure. It is thus preferred to use arrangements in which there are no moving parts. It is known for example to mix a gas with a liquid by injecting a stream of liquid coaxially into a mixing tube which is cylindrical over its entire length or which has a short cylindrical neck followed by a conically diverging tube, the jet of liquid mixing, in the mixing tube, with a stream of gas fed to the annular space between the liquid jet and the wall of the mixing tube. Such so-called ejector reactors are known, for example, as Venturi absorbers in the chemical industry. It is frequently necessary to connect the relatively small ejector reactors to a larger reaction chamber for continuation of the reaction, which reaction chamber may be in the form of a bubble column for example. In a large number of reactions between a gas and a liquid, however, the degree of gas/liquid mixing achieved in ejector reactors is inadequate for the provision of satisfactory yields and conversions.

We have now found that the mixing of gas and liquid in a tubular reactor by feeding a gas and liquid to a mixing zone may be advantageously carried out by feeding a stream of liquid to a mixing zone through one or more liquid nozzles whose axes extend in the same direction as the axis of the mixing zone, the injector liquid having a velocity of from 5 to 100 m./s., whilst a second stream of liquid of much lower velocity is introduced into the reactor inlet zone surrounding said nozzles, the gas being fed to the mixing zone through one or more gas inlets



located near the orifices of the liquid nozzles and the mean cross-sectional area of the mixing zone being from 5 to 500 times as large as the cross-sectional area of the orifice of the liquid nozzle or the sum of the cross-sectional areas of the orifices of the liquid nozzles, whilst the length of the mixing zone is from 2 to 30 times as great as its hydraulic diameter.

In an advantageous embodiment of the method, spiral motion is imparted to the liquid flowing through the liquid nozzles before it emerges therefrom and/or spiral motion is imparted to the liquid/gas mixture in the mixing zone.

Using this method, high yields and also high space-time yields may be achieved. Since our novel method makes use of smaller tubular reactors than those employed in conventional methods, for example, for reactions in bubble columns, the method of the invention involves considerably less expense than prior art methods.

Our new method is generally suitable for mixing gases and liquids in processes either for effecting exchange of matter or for inducing a reaction between a gas and a liquid. It is particularly suitable for carrying out chemical reactions between gases and liquids where rapid and thorough mixing is required. It will be appreciated that the gas and liquid need not be pure substances but may also be any desired mixtures of substances. In the method of the invention it is also possible to mix a gas with two different liquids, one of which is injected through a nozzle while the other is fed to the reactor inlet zone surrounding the nozzle. The method of the invention is used to advantage in carrying out reactions between gas and liquid where the reaction product should not recontact the starting materials. For example, the present method may be used for the absorption of chlorine in water or the reaction of propylene with aqueous chlorine solution to form propylene chlorohydride. When applying our method to said reactions, the reaction conditions generally used for such processes, for example catalyst, temperature and pressure, are not affected. However, the greater mixing rate and improved thoroughness of mixing achieved in our method increase the reaction rate and thus improve the degree of conversion. It may therefore be advantageous to determine new optimum process parameters such as average residence time, temperature, pressure and amount of catalyst used, since the optimum values hitherto found in an industrial process may now no longer apply due to the higher reaction rate obtained in our method. Using this novel method, it is frequently possible to carry out reactions between a gas and liquid at somewhat lower temperatures and in many cases higher yields of reaction product are obtained. The method is advantageously used in the oxidation of organic and inorganic compounds with oxygen or oxygen-containing gases such as air, for example the oxidation of sodium sulfite in aqueous solution with air to form sodium sulfate.

The method of the invention is also advantageous for carrying our processes for effecting the transfer of material, for example the absorption of chlorine in water or the absorption of phosgene in organic solvents such as methylene chloride.

It is an important feature of our method that a preferably relatively small stream of liquid is injected through a nozzle at a velocity of from 5 to 100 m./s. and preferably from 10 to 30 m./s. whilst a second, preferably relatively large stream of liquid, is fed to the reactor inlet zone surrounding the nozzles at a considerably lower velocity than that of the injected liquid. In general, the ratio of injected liquid to the liquid fed to the said inlet zone is from 1:1 to 1:50 and preferably from 1:1 to 1:10. Advantageously, the velocity of the liquid fed to said inlet zone is from 0.1 to 20 m./s. and preferably from 0.5 to 5 m./s.

The feed of liquid to the reactor inlet zone surrounding the nozzles may be effected through one or more feed lines and the actual number of such feed lines is not critical.

The average cross-sectional area of the mixing zone should be from 5 to 500 times and preferably from 10 to 100 times as large as the cross-sectional area of the liquid nozzle orifice or the sum of the cross-sectional areas of the liquid nozzle orifices, and the length of the mixing zone should be from 2 to 30 times as great as its hydraulic diameter. The length and the hydraulic diameter of the inlet zone may be varied within wide limits. The mixing zone generally has a constant cross-sectional or a cross-section which increases in the direction of flow, and it may vary in design. In general, a cylindrical tube or alternatively a mixing tube having a short cylindrical neck followed by a conically diverging tube is used. The inlet zone may also vary in design, although it generally takes the form of a cylindrical tube.

By the term "hydraulic diameter" of a zone we mean the diameter of a cylindrical tube which has the same length as the zone in question and shows the same pressure loss when fluid is passed therethrough at the same rate.

In the method of the invention, a single liquid nozzle or a plurality of liquid nozzles, for example from 2 to 10 such nozzles, may be used. Where a plurality of liquid nozzles is used, these may be arranged in a circle or in one or more close groups. The gas is also fed through one or more, for example from 2 to 10, gas nozzles, the number of gas nozzles and the number of liquid nozzles being the same or different. The gas is generally introduced in the proximity of the orifices of the liquid nozzles. The nozzle orifices may be in the form of, say, round holes, slots or even annular gaps. The gas generally emerges from the nozzle(s) in the same direction as the jet(s) of liquid and the gas velocity is conveniently not higher than that of the jet(s) of liquid. In general, the velocity of the injected gas is from 5 to 50 m./s. Preferably, the gas and injected liquid are introduced through a two-component nozzle, the liquid being fed through the central orifice of the nozzle whilst the gas flows through the annular gap coaxially surrounding the said central orifice.

In a preferred embodiment of the method, spiral motion is imparted to the injected liquid before it leaves the nozzle and/or spiral motion is imparted to the liquid/gas mixture in the mixing zone. Spiral motion may be imparted to the injected liquid for example by placing a twist guide in the form of a single-pitch or multiple-pitch screw in the path of the liquid upstream of the nozzle outlet or by arranging for the liquid to flow into the feed line of the nozzle tangentially. Spiral motion may be imparted to the liquid/gas mixture in the mixing zone for example by imparting a twist to the slow outer stream also, for example by providing a rifled inlet to said mixing zone. It is particularly advantageous to create a back-pressure at the end of the mixing zone. This may be achieved, for example, by connecting a sufficiently high bubble column to the outlet of the mixing zone. Alternatively, an energy-consuming system in the form of baffle plates or centrifugal separators may be placed downstream of the mixing zone. Another method of creating a back-pressure is to insert a pressure-holding valve downstream of the mixing zone. In general, the mixing zones are arranged vertically, the gas and liquids being caused to flow upwardly therethrough. Alternatively, the gas and liquid may be caused to flow downwardly through vertical mixing zones or the mixing zones may be disposed horizontally or in an inclined position, as desired.

The invention is further described with reference to the accompanying drawings.

In FIG. 1, the mixing zone is designated by the reference numeral **1**, the gas nozzle outlet by **2**, the liquid nozzle outlet by **3**, the liquid and gas nozzle inlets by **4** and **5** respectively, whilst the reference numeral **6** designates the liquid inlet to the inlet zone **7**. The transition from inlet zone to mixing zone is conveniently gradual in order to prevent the liquid flowing from the inlet zone to the mixing zone from detaching itself from the walls of these zones.

FIG. 2 illustrates a combination of the jet reactor with baffle plate **8** disposed downstream of the mixing zone and the use of a twist guide in the path of the injected liquid.

FIG. 3 illustrates a combination of the jet reactor with a conventional bubble column **10** having a gas outlet **11** and liquid outlet **12**. In this case, the pet reactor serves as the gassing device for the bubble column.

## EXAMPLE 1

The reaction was carried out using a tubular reactor having a diameter of 20 mm. (see FIG. 1). The length of the mixing zone was 150 mm. and the diameter of the liquid nozzle was 5 mm. The liquid nozzle was coaxially surrounded by an annular gas nozzle.

The liquid passed through the liquid nozzle at a velocity of 20 m./s. consisted of 1.4 $m^3$/hr. of an aqueous sodium sulfite solution containing 600 moles/$m^3$ of sodium sulfite and 0.27 mole/$m^3$ of cobalt sulfate as catalyst. The reaction temperature was 20° C. and the pH was adjusted to 9.2 $m^3$/hr. of air (STP) were passed through the annular nozzle. A futher 2.0 $m^3$/hr. of aqueous sodium sulfite solution of the above concentration were passed to the inlet zone through a separate inlet. The slow stream of liquid had a velocity of 2.2 m./s. In the oxidation of the sodium sulfite to sodium sulfate, the conversion, based on atmospheric oxygen, was 52%.

When the reaction was carried out in the above-described manner but without feeding liquid to the inlet zone through the separate inlet, the conversion was only 15% based on atmospheric oxygen.

## EXAMPLE 2

The reaction was carried out in the manner described in the first paragraph of Example 1, a screw having been placed in the path of the liquid fed to the nozzle in order to impart spiral motion to said liquid. The conversion of sodium sulfite to sodium sulfate, based on atmospheric oxygen, was 70%. (When comparing the yield of Example 2 with that of Example 1 it should be noted that the overall momentum of the twisted jet in Example 2 was 1.32 times smaller than that of the untwisted jet in Example 1.)

## EXAMPLE 3

The reaction was carried out as described in the first paragraph of Example 1, a baffle plate having been placed at a distance of 20 mm. from the outlet of the mixing zone. The conversion of sulfite to sulfate was 80%.

## EXAMPLE 4

The reaction was carried out using a tubular reactor having a diameter of 20 mm. The length of the mixing zone was 200 mm. and the diameter of the liquid nozzle was 3 mm. The liquid nozzle was coaxially surrounded by an annular gas nozzle.

The liquid passed through the liquid nozzle at a velocity of 15 m./s. consisted of 333 l./hr. of water, the resulting jet of water being concentric with the mixing zone. A further 667 l./hr. of water were fed to the inlet zone surrounding the nozzles through a separate inlet at a lower velocity. 1.5 $m^3$/hr. of chlorine (STP) were fed through the annular gas nozzle. The chlorine was completely absorbed within the mixing zone and a 70% saturation of the water was achieved.

We claim:

1. A method of mixing a gas with liquids in a tubular reactor by feeding the gas and liquids to a mixing zone, wherein a stream of liquid is fed to a mixing zone through one or more liquid nozzles whose axes extend in the same direction as the axis of the mixing zone, the injected liquid having a velocity of from 5 to 100 m./s., while a second stream of liquid is introduced at a velocity in the range of 0.1 to 20 m./s. and also substantially slower than said injected liquid's velocity into the reactor inlet zone surrounding said nozzle or nozzles, the gas being fed to the mixing zone through one or more gas inlets located near the orifices of the liquid nozzles and the mean cross-sectional area of the mixing zone being from 5 to 500 times as large as the cross-sectional areas of the orifices of said liquid nozzles, the ratio of said injected liquid to said liquid of said second stream being in the range of 1:1 to 1:50, and the length of the mixing zone is from 2 to 30 times as great as its hydraulic diameter.

2. A method as claimed in claim 1, wherein spiral motion is imparted to the liquid flowing through the liquid nozzle or nozzles before it leaves said nozzle or nozzles.

3. A method as claimed in claim 1, wherein spiral motion is imparted to the liquid/gas mixture in the mixing zone.

4. A method as claimed in claim 1, wherein back pressure is placed on the liquid/gas mixture at the downstream end of the mixing zone.

5. A method as claimed in claim 4, wherein the velocity of the gas fed to the mixing zone is from 5 to 50 m./s. and also not higher than said injected liquid's velocity.

6. A method as claimed in claim 4, said injected liquid being fed through a central orifice of the liquid nozzle and said gas being supplied as an annular gas stream about the liquid flowing from said central orifice.

7. A process as claimed in claim 4, said injected liquid having a velocity of 10 to 30 m./s., said second stream of liquid having a velocity of 0.5 to 5 m./s., and said gas fed to said mixing zone having a velocity which is not higher than said velocity of said injected liquid.

**References Cited**

UNITED STATES PATENTS

| | | | |
|---|---|---|---|
| 2,747,974 | 5/1956 | Felger | 23—252 R |
| 2,361,150 | 10/1944 | Petroe | 23—252 R |
| 2,798,794 | 7/1957 | Muench et al. | 23—252 R |
| 3,755,452 | 8/1973 | Sinn et al. | 260—586 B |

OSCAR R. VERTIZ, Primary Examiner

H. S. MILLER, Assistant Examiner

U.S. Cl. X.R.

261—18 B, 75, 76; 423—199