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(54) PROCESS OF AND APPARATUS FOR PREPARING DINITROGEN PENTOXIDE.

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Description

This invention relates to the preparation of dinitrogen pentoxide (N_2O_5) by the reaction between dinitrogen tetroxide (N_2O_4) and ozone.

N_2O_5 is a powerful and selective nitrating agent, especially when dissolved in inert organic solvents. Hitherto, its use as a nitrating agent has been limited because of its relatively high cost and low thermal stability.

One process of N_2O_5 preparation which is capable of producing a high purity product with a relatively long shelf life is the reaction of gaseous N_2O_4 with ozone. Ozone is produced at a maximum concentration of about 4 wt% in oxygen within a silent electrical discharge ozoniser, and is immediately reacted with a stoichiometric amount of N_2O_4 vapour in an inert gas carrier to give N_2O_5. The inert gas stream containing the N_2O_5 product is then brought into contact with a cold surface maintained at a low temperature of typically -70°C to condense out the product as a solid, whilst at the same time minimising losses.

There are several disadvantages with this process. These are:

1. Since the concentration of ozone in the carrier gas stream is very low, the volume of carrier gas is high in comparison with the volume of gaseous N_2O_5 product. The potential for product losses in the carrier gas is therefore high, which is one reason for employing a very low recovery temperature.
2. The reaction between N_2O_4 and ozone is exothermic, which has a detrimental effect on the thermally unstable N_2O_5 product.
3. The recovered product is a solid which is difficult to transport, store, and use.
4. A very low product recovery temperature of typically -70°C must be used, which leads to high operating costs especially since the high volume of carrier gas must also be cooled to the same temperature.
5. The product is condensed onto a cold surface whose thermal efficiency falls as the thickness of product builds up.
6. The maintenance of a stoichiometric ratio of N_2O_4 and ozone is difficult to achieve, especially when these reagents are present in such low concentrations. A slight excess of ozone is lost in the carrier gas which adds to operating costs, whereas a slight excess of N_2O_4 leads to N_2O_4 contamination in the condensed product.
7. Any water vapour present in the ozone/gas stream from the ozoniser produces nitric acid contamination in the product by reaction with N_2O_5.

It is an object of the present invention in a first aspect to provide a process for generating N_2O_5 by reaction between ozone and N_2O_4 whereby the aforementioned disadvantages are overcome or at least mitigated in part.

Accordingly, a process for preparing N_2O_5 comprises the steps of

(a) providing a solution of N_2O_4 in a first body of a volatile inert organic solvent,

(b) contacting the solution with a carrier gas containing ozone at a temperature sufficient to promote formation of N_2O_5 in, and evaporation of solvent into, the carrier gas,

(c) contacting the N_2O_5 -laden carrier gas with a second body of inert organic solvent at a temperature below that of step (b) to condense the N_2O_5 therein, and

(d) recovering the condensed N_2O_5 within the solvent from step (c).

The principal advantage of the gas/liquid interaction in step (b) is that the heat generated during the exothermic reaction between ozone and N_2O_4 in step (b) is both absorbed by intimate contact with liquid solvent of relatively high specific heat capacity and counteracted by the latent heat of vaporisation of the solvent and N_2O_4. In this way, large temperature increases are suppressed and thermal decomposition of the N_2O_5 avoided. Although this means that the carrier gas becomes contaminated with solvent vapour, step (c) provides an effective means of recovering this solvent by low temperature condensation into the second body of solvent whilst at the same time directly converting the N_2O_5 into a readily storable, transportable and useable form without an intermediate solids handling stage.

The chemical identity of the first and second bodies of solvent are preferably the same to avoid the problem of one solvent contaminating the other. Preferably, this solvent comprises C_1 or C_2 chloroalkane, especially dichloromethane, or a chlorofluorocarbon, especially a C_1 or C_2 chlorofluoroalkane.

A further advantage of producing N_2O_5 in step (b) is that any moisture introduced within the carrier gas (which preferably comprises ozonised oxygen or ozonised air) hydrates some of the N_2O_5 and becomes trapped as relatively involatile nitric acid condensate within the liquid solvent product of step (b). This obviates the need to provide a very dry source of carrier gas. If however the carrier gas is essentially moisture free then the product of step (b) will not be contaminated with nitric acid, and since it too will contain some dissolved N_2O_5 this solvent product may be blended with the N_2O_5 -containing solvent of step (d) to form the principal product of the present method.

The carrier gas used in step (b) preferably contains a stoichiometric excess of ozone of preferably up to 5%, more preferably from 0.5 to 2%, over that required to generate the N_2O_5. Any excess ozone present in the carrier gas is carried over to and at least partly absorbed by the second body of inert solvent, with the result that it prolongs the shelf life of the N_2O_5 in the product solvent by reconverting its principal product of thermal decomposition (N_2O_4) back into N_2O_5.

Step (b) is preferably performed within the temperature range -20 °C to +30 °C, more preferably -15 °C to +20 °C, and most preferably at a temperature at which N_2O_4 remains strongly coloured in solution so that its complete removal from solution can be detected by the absence of colour using, for example, colourimetric detection means. Strong solution colouration by N_2O_4 is observed at temperatures at or above -15 °C, especially at temperatures at or above -10 °C. At or above these temperatures N_2O_5 in solution is essentially colourless and so its presence in solution does not interfere with the colourimetric detection of N_2O_4. However, at temperatures above +20 °C the N_2O_5 becomes increasingly thermally unstable and so a maximum temperature of +10 °C is preferred in order to promote a reasonable rate of solvent and nitrogen oxide evaporation.

The carrier gas and first body of solvent are preferably contacted in co-current flow to promote continuous and efficient formation of N_2O_5 in the gas phase. Co-current flow ensures that the concentrations of ozone and N_2O_4 in the carrier gas and solvent respectively are at a maximum at the beginning of co-current flow contact and at a minimum at the end of that contact. The first body of solvent is preferably continuously recirculated in a closed loop to facilitate its replenishment with low concentrations of fresh N_2O_4 and to facilitate the transfer of N_2O_5 into the gas phase. In order to provide a large surface area of intimate contact between the first body of solvent and the carrier gas and to promote rapid vaporisation of N_2O_4 and solvent, step (b) is conveniently conducted in a first packed column.

Step (c) is preferably conducted at a temperature of -15 °C or less, preferably -20 °C or less, in order to ensure substantially complete recovery of N_2O_5 from the carrier gas. At temperatures below -50 °C no significant improvement in N_2O_5 and ozone removal from the carrier gas occurs, but since the vapour pressure of the solvent steadily declines with decreasing temperature so the amount of solvent recovered from the gas into the second body of inert solvent increases and this in turn obviates the need for a subsequent solvent recovery step. For this reason, step (c) may be performed at temperatures typically as low as -70 °C though at the expense of increased cooling costs and increased amounts of crystalline N_2O_5 formed in solution. In order to promote efficient recovery of N_2O_5 from the carrier gas, the carrier gas and the second body of inert organic solvent are preferably contacted in continuous countercurrent flow, most preferably within a second packed column. This has the added advantage of minimising the temperature of, and hence the solvent vapour concentration in, the spent carrier gas. As with the first body of solvent, the second body of solvent is preferably continuously recirculated in a closed loop to facilitate its reuse in step (c) and to promote the build-up of a high concentration of N_2O_5 therein.

The carrier gas is preferably provided as a single stream which contacts the first and second bodies of solvent in sequence.

The concentration of ozone in the carrier gas is preferably at least 0.1 wt% and will not normally exceed 4 wt%. In order to promote complete reaction between the ozone and N_2O_4, it is preferable that the concentration of N_2O_4 in the solvent is carefully controlled to ensure that the vapour pressure of N_2O_4 is approximately equal to the vapour pressure of ozone at the commencement of step (b). This requires that the average concentration of N_2O_5 in the solvent is preferably between 0.005 and 0.05 wt% and is more preferably between 0.01 and 0.02 wt%. Close control over N_2O_4 concentration can be maintained by continuously or intermittently adding N_2O_4 at a known rate to the first body of solvent recirculating in a closed loop. The N_2O_4 is preferably added as a concentrated solution of typically between 25 and 60 wt% in the same organic solvent as that of the first body so as to make up for solvent losses by evaporation into the carrier gas.

In a second aspect of the present invention, there is provided an apparatus for performing the process of the first aspect comprising a reaction vessel having an ozone generation means a solvent supply means connected thereto, an absorber vessel having a cooling means connected thereto and having a gas vent and a solvent product outlet extending therefrom, and a carrier gas transfer line connecting between the reaction and absorber vessels.

The apparatus preferably further includes a first solvent recirculation means connecting between a solvent inlet and a solvent outlet of the reaction vessel, and a second solvent recirculation means connecting between a solvent inlet and a solvent outlet of the absorber vessel.

The ozone generation and solvent supply means are preferably connected in co-current flow to the reaction vessel whereas the solvent inlet and gas transfer lines to the absorber vessel and preferably connected in countercurrent flow. Each vessel preferably comprises a packed column.

Examples of the present invention will now be described with reference to the following apparatus and process descriptions and to the accompanying drawing

The apparatus illustrated in Figure 1 comprises a feedstock reservoir 2, a metering pump 6 with a motor 8, a reactor column 10 containing a packed section 12, a reactor column recirculation pump 14, an absorber column 16 containing a packed section 18, and an absorber column recirculation pump 20.

A reactor solvent recirculation line 22 extends between the bottom and top of the reactor column 10 through the reactor column recirculation pump 14. Similarly, an absorber solvent recirculation line 24 extends between the bottom and top of the absorber column 16 through the absorber recirculation pump 20. Each of the reactor and absorber recirculation lines has a product offtake line 26 and 28 respectively fitted with a valve 30 and 32 respectively. A feedstock transfer line 34 connects between the reservoir 2 and the reactor recirculation line 22 through the metering pump 6. A solvent makeup line 36 connects with the absorber recirculation line 24.

A gas inlet line 38 extends from an oxygen supply 40, through an ozone generator 42 and into the top of the reactor column 10. At the top of the absorber column 16 is a gas outlet line 44. The two columns are connected below their respective packed sections by a gas transfer line 46, and a liquid return line 48 having a valve 50. The absorber column 16 is mounted slightly higher than the reactor column 10 to allow excess liquid collected in the bottom of the absorber column to overflow into the reactor column through the return line 48 when the valve 50 is open.

Cooling coils 52, 54 and 56 are fitted, respectively, in the reservoir 2, in the bottom of the reactor column 10 and in the bottom of the absorber column 16 for controlling the temperature of the solvent at various parts of the apparatus.

A. BATCH PROCESS (SEMI-CONTINUOUS)

Process Description

1. With the equipment dry and purged with nitrogen to remove all traces of moisture, a quantity of moisture-free inert organic solvent is charged to the reactor column 10 and the absorber column 16.
2. The absorber column recirculation pump 20 is switched on and the temperature of the solvent in the absorber column 16 brought down to below -15 °C using the cooling coil 56, the actual temperature selected being set by the partial pressures of the components in the gas stream to minimise process and solvent losses. The reactor column recirculation pump 14 is also switched on. Valves 30 and 32 are kept closed and valve 50 is kept open.
3. The oxygen stream is then switched on from its source 40 to reduce the temperature of the solvent in the reaction column by evaporation, and the ozone generator 42 brought into operation and adjusted for optimum conditions. The ozone-containing oxygen stream passes down the packed section 12 of the reactor column 10 in co-current flow with reactor column solvent, through the gas transfer line 46, up the packed section 18 of the absorber column 16 in countercurrent flow with absorber solvent, and out through the gas outlet line 44.
4. Using the metering pump 6, the N_2O_4 solution in the reservoir 2 is continuously metered into the circulating solvent of the reactor colum 10 and thereby flows down the packed column co-currently with the ozonised gas stream. The relative flow rate of N_2O_4 and ozone through the reactor column are selected to ensure that ozone is always present in stoichiometric excess. Contact between solvent and gas streams causes a proportion of the ozone to dissolve into the liquid phase and react exothermically with the N_2O_4 to give N_2O_5 in solution. At the same time some of the N_2O_4 and solvent vaporises, the vaporised N_2O_4 then immediately reacting exothermally with the ozone in the gas stream to produce more N_2O_5. The effect of metering the N_2O_4 solvent into solvent recirculating within a closed circuit is to keep the N_2O_4 concentration in the solvent entering the reaction column very low (typically between 0.01 and 0.02 wt%) to maintain a low N_2O_4 partial pressure so that its concentration released into the gas stream by vaporisation is approximately equal to the low concentration of ozone present in the gas stream. The gas and liquid flow in the reactor column 10 is co-current. This ensures that the concentrations of the reactive components (ozone and N_2O_4) in these two streams are at a maximum at their point of contact and at a minimum when the streams separate and permits maximum contact time for the reaction to take place. In this way, complete reaction between the ozone and N_2O_4 is promoted, losses are minimised, and the liberated heat of reaction between the N_2O_4 and ozone can be effectively and progressively counteracted by the cooling effect produced by the evaporation of the solvent and the N_2O_4 so as to suppress thermal decomposition of the N_2O_5 once formed. Any water vapour present in the gas stream reacts with N_2O_5 to form nitric acid, which being relatively involatile condenses out in the reactor column solvent.

5. By appropriate use of the cooling coils 52 and 54, the temperature within the packed section 12 of the reactor column 10 is maintained at a temperature above that which N_2O_4 and N_2O_5 are effectively stripped out of solution (typically above -10°C) and yet within a temperature range in which N_2O_4 in solution is strongly coloured brown. Thus, colour indicator 58 is used to adjust the speed of the pump 6 to ensure the flow of N_2O_4 in solution to the reactor column 10 is lower than that which causes a brown colouration in the solvent at the base of the column. This in turn means that the required excess of ozone is present in the gas stream since all the N_2O_4 is being converted to N_2O_5. The required concentration of the N_2O_4 solution in the reservoir is calculated with the assistance of the level indicator 60 to ensure that the flow rate of solvent pumped through the pump 6 matches the rate of solvent evaporation into the gas stream which is equivalent to a constant solvent level indication in the reactor column 10.

6. In the packed section 18 of the absorber column 16, the gas stream, now laden with N_2O_5 and saturated with solvent, contacts the recirculating low temperature absorber solvent in countercurrent flow. This causes the N_2O_5, solvent, and excess ozone components within the gas stream to condense out into the solvent. Countercurrent flow ensures that the temperature of the gas, and thus the concentration of each of these components, is at a minimum as the gas leaves the column. As a safety measure, any excess solvent carried over from the reactor column 10 to the absorber column 16 is allowed to return under gravity through the valve 50. This prevents accidental build-up of excess solvent within the absorber column 16.

7. When the required quantity of N_2O_5 has been prepared, as judged by a mass balance to give a final solution concentration at a desirable level for subsequent usage, the ozone generator 42 is turned off together with the oxygen source 40 and the metering pump 6. The equipment is then allowed to warm up (typically to between -20°C and 0°C) to ensure that any crystals of N_2O_5 formed in the absorber column 16 redissolve in the solvent. The valve 32 is then opened and the N_2O_5 solution in the absorber column 16 drawn off through the offtake line 28.

8. The solution in the reactor column 6, if it is not contaminated with nitric acid, can be drawn off through the offtake line 26 with valve 30 open and blended with the solution from the absorber column.

9. The equipment can then be recharged with fresh solvent and the process repeated.

Example 1 - Batch (Semi-Continuous)

N_2O_4 (5kg) was dissolved in dichloromethane (5kg), to give a 50 wt% feedstock which was metered in to the absorber column at 0.38kg per hour.

The reactor and absorber columns were charged with dichloromethane (41kg).

Solvent was recirculated through each column in a closed loop at a rate, typically 1200kg per hour, calculated to be above the minimum wetting conditions and below the flooding conditions for the packed column used.

Oxygen was fed through the ozone generator to give 2 wt% ozone at an ozone flowrate of 100g/hr.

With the reactor column at 0°C and absorber column at -50°C, the reaction was complete after approximately 26 hours.

The product output from the absorber column was a 12 wt% N_2O_5 solution (40kg) containing 0.5 wt% nitric acid.

The solution output from the reactor column was a 6 wt% N_2O_5 solution (8kg) containing 2 wt% nitric acid.

Typical vent losses were N_2O_5 (0.1kg) and dichloromethane (1.5kg).

Typical system losses (ie remaining within system) were N_2O_5 (0.3kg) and dichloromethane (2kg).

B. CONTINUOUS WITH VALVE 50 CLOSED

Process Description

1. Process steps 1 to 6 of the batch (semi-continuous) process are performed as described above, except that product is taken off continuously through line 28 with valve 32 open, and fresh dry solvent is continuously added through line 36 as required in order to maintain the concentration of N_2O_5 in the product as necessary.

2. The continuous process may be shut down intermittently if and when an unacceptably high concentration of nitric acid builds up in the solvent within the reactor column, to be replaced by fresh acid-free solvent. The pump 6 is first switched off and reaction and evaporation within the reactor column allowed to continue for a short while to remove all trace of N_2O_4 and N_2O_5 from the solvent. Thereafter, the oxygen source 40, ozone generator 42 and recirculation pump 20 are switched off and the nitric acid-laden solvent in the reactor column 10 is drained out through the offtake line (with valve 30 open) for subsequent solvent recovery. Fresh dry solvent is then charged to the reactor column 10, and the continuous process restarted.

Example 2 - Continuous with Valve 50 Closed

Feedstock consisting of 50 wt% N_2O_4 in dichloromethane was metered into the reaction column at 0.38kg/hr.

The reactor and absorber columns were charged with dichloromethane (41kg) which was recirculated at 1200 kg/hr through each column.

Oxygen was fed through the ozone generator to give 2 wt% ozone at an ozone flowrate of 100g/hr.

Solvent within the reservoir, reactor column and absorber column was controlled at, respectively, 15 °C, 0 °C, and -25 °C.

As soon as equilibrium conditions were obtained, product consisting of 6 wt% N_2O_5 and less than 0.5% nitric acid in dichloromethane was taken continuously from the absorber column at a rate of 1.8kg/hr.

The temperature of the absorber column was controlled at -25 °C necessitating additional solvent vapour recovery from the spent gas stream. The higher absorber temperature had the additional advantage that it suppressed the formation of an N_2O_5 slurry in the absorber solvent which could otherwise block the pump 20 and line 24.

C. CONTINUOUS WITH VALVE 50 OPEN

Process Description

1. Process steps 1 to 6 of the batch (semi-continuous) process are performed as described above, except that valve 28 remains shut, valve 50 is kept open, and product is taken off continuously through line 26 with valve 30 open.
2. The absorber column may be operated at a low temperature (typically -50 °C) at which an N_2O_5 slurry will form, but since the slurry flows into the warmer absorber column it dissolves and so the product taken from the reactor column contains only dissolved N_2O_5. Since any nitric acid formed in the process can only be removed with the product, it is highly desirable that the oxygen source supplies dry, essentially moisture-free oxygen to the ozone generator.

Example 3 - Continuous with Valve 50 Open

Process conditions were identical to those of Example 2, except that the flow of make-up solvent through line 36 was reduced to increase the concentration of N_2O_5 in the product (now taken from the reaction column) from 6 wt% to 12 wt% now made possible by the higher temperature of, hence higher N_2O_5 solubility within, the product solvent.

Claims

1. Process for preparing N_2O_5 comprising the steps of:
   (a) providing a solution of N_2O_4 in a first body of a volatile inert organic solvent,
   (b) contacting the solution with a carrier gas containing ozone at a temperature sufficient to promote formation of N_2O_5 in, and evaporation of solvent into, the carrier gas,
   (c) contacting the N_2O_5 -laden carrier gas with a second body of inert organic solvent at a temperature below that of step (b) to condense the N_2O_5 therein, and
   (d) recovering the condensed N_2O_5 within the solvent from step (c).

2. Process according to claim 1 characterised in that the chemical identity of the first and second bodies of inert organic solvent are the same.

3. Process according to claim 1 or claim 2 characterised in that the solvent of the first and/or second bodies of solvent comprises a C_1 or C_2 chloroalkane or a chlorofluorocarbon.

4. Process according to any one of the preceding claims characterised in that the carrier gas employed in step (b) contains a stoichiometric excess of ozone over that required to react with the N_2O_4.

5. Process according to claim 4 characterised in that the carrier gas contains a stoichiometric excess of ozone of up to 5%.

6. Process according to any one of the preceding claims characterised in that step (b) is performed within the temperature range -20 °C to + 30 °C, preferably -15 °C to +20 °C.

7. Process according to any one the preceding claims characterised in that step (c) is performed within the temperature range -15 °C to -70 °C.

8. Process according to claim 7 characterised in that step (c) is performed within the temperature range -20 °C to -50 °C.

9. Process according to any one of the preceding claims characterised in that the solvent product of step (d) is blended with at least part of the solvent product of step (b).

10. Process according to any one of the preceding claims characterised in that each of the first

body of solvent and carrier gas is provided as a separate stream.

11. Process according to claim 10 characterised in that the carrier gas and first body of solvent are contacted in step (b) in co-current flow.

12. Process according to claim 10 or claim 11 characterised in that the first body of solvent is recirculated.

13. Process according to any one of claims 10 to 12 characterised in that the second body of solvent is provided as a separate stream.

14. Process according to claim 13 characterised in that the carrier gas and second body of solvent are contacted in step (c) in countercurrent flow.

15. Process according to claim 13 or claim 14 characterised in that the second body of solvent is recirculated.

16. Process according to any one of the preceding claims characterised in that the concentration of N_2O_4 in the first body of solvent employed in step (b) is from 0.005 to 0.05 wt%.

17. Apparatus for performing the process according to claim 1 comprising:
a generally vertically disposed reaction vessel (10) an upper portion of which is packed, said vessel having a solvent product outlet (26) towards the bottom thereof and associated means for continuously recirculating solvent downwards through the packed portion thereof;
ozone generating means (42) arranged to supply ozone into the upper portion of the said reaction vessel (10);
means (2,6) for supplying solvent and N_2O_4 into the top of said reaction vessel (10);
a generally vertically disposed absorber vessel (16) an upper portion of which is packed, said vessel having at the top thereof a gas vent (44) and towards the bottom thereof a solvent product outlet (28);
means for continuously recirculating solvent downwards through the absorber vessel (16) through the packed portion thereof; and
a carrier gas transfer line (46) connecting between the reaction and absorber vessels in each case at a point below the packed portions of the respective vessels.

Patentansprüche

1. Verfahren zur Herstellung von N_2O_5, das die folgenden Stufen umfaßt:
(a) Bereitstellen einer Lösung von N_2O_4 in einer ersten Substanz in Form eines flüchtigen, inerten, organischen Lösungsmittels,
(b) In-Berührung-Bringen der Lösung mit einem Ozon enthaltenden Trägergas bei einer zur Förderung einer N_2O_5-Bildung in dem und zur Verflüchtigung des Lösungsmittels in das Trägergas ausreichenden Temperatur,
(c) In-Berührung-Bringen des mit N_2O_5 beladenen Trägergases mit einer zweiten Substanz in Form eines inerten, organischen Lösungsmittels bei einer unter der Temperatur von Stufe (b) liegenden Temperatur zur Kondensation des N_2O_5 darin und
(d) Gewinnen des kondensierten N_2O_5's in dem Lösungsmittel aus Stufe (c).

2. Verfahren nach Anspruch 1, dadurch gekennzeichnet, daß die erste und zweite Substanz in Form eines inerten, organischen Lösungsmittels chemisch identisch sind.

3. Verfahren nach Anspruch 1 oder Anspruch 2, dadurch gekennzeichnet, daß das Lösungsmittel der ersten und/oder zweiten Substanz in Form eines Lösungsmittels ein C_1- oder C_2-Chloralkan oder einen Chlorfluorkohlenstoff umfaßt.

4. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß das in Stufe (b) verwendete Trägergas, bezogen auf die zur Reaktion mit dem N_2O_4 erforderliche Menge, einen stöchiometrischen Überschuß an Ozon enthält.

5. Verfahren nach Anspruch 4, dadurch gekennzeichnet, daß das Trägergas einen stöchiometrischen Überschuß an Ozon von bis zu 5 % enthält.

6. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß Stufe (b) in einem Temperaturbereich von - 20 °C bis +30 °C, vorzugsweise -15 °C bis +20 °C durchgeführt wird.

7. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß Stufe (c) in einem Temperaturbereich von - 15 °C bis -70 °C durchgeführt wird.

8. Verfahren nach Anspruch 7, dadurch gekennzeichnet, daß Stufe (c) in einem Temperaturbereich von -20 °C bis -50 °C durchgeführt wird.

9. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß das Lösungsmittelprodukt der Stufe (d) mit mindestens einem Teil des Lösungsmittelprodukts der Stufe (b) vermischt wird.

10. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß sowohl die erste Substanz in Form eines Lösungsmittels als auch das Trägergas als getrennter Strom bereitgestellt werden.

11. Verfahren nach Anspruch 10, dadurch gekennzeichnet, daß das Trägergas und die erste Substanz in Form eines Lösungsmittels in Stufe (b) im Gleichstrom in Berührung gebracht werden.

12. Verfahren nach Anspruch 10 oder Anspruch 11, dadurch gekennzeichnet, daß die erste Substanz in Form eines Lösungsmittels in den Kreislauf zurückgeführt wird.

13. Verfahren nach einem der Ansprüche 10 bis 12, dadurch gekennzeichnet, daß die zweite Substanz in Form eines Lösungsmittels als ein getrennter Strom bereitgestellt wird.

14. Verfahren nach Anspruch 13, dadurch gekennzeichnet, daß das Trägergas und die zweite Substanz in Form eines Lösungsmittels in Stufe (c) im Gleichstrom in Berührung gebracht werden.

15. Verfahren nach Anspruch 13 oder Anspruch 14, dadurch gekennzeichnet, daß die zweite Substanz in Form eines Lösungsmittels in den Kreislauf zurückgeführt wird.

16. Verfahren nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß die Konzentration an N_2O_4 in der in Stufe (b) verwendeten, ersten Substanz in Form eines Lösungsmittels 0,005 bis 0,05 Gew.-% beträgt.

17. Anlage zur Durchführung des Verfahrens nach Anspruch 1 mit einem im allgemeinen senkrecht angeordneten Reaktionsgefäß (10), dessen oberer Teil gepackt ist, wobei das Gefäß einen Lösungsmittelproduktauslaß (26) in Richtung auf seinen Boden und eine damit verbundene Vorrichtung zur kontinuierlichen Rückführung von Lösungsmittel nach unten durch seinen gepackten Teil aufweist,
einer Ozon erzeugenden Vorrichtung (42), die so angeordnet ist, daß Ozon in den oberen Teil des Reaktionsgefäßes (10) eingeführt wird,
einer Vorrichtung (2, 6) zur Einführung von Lösungsmittel und N_2O_4 in die Oberseite des Reaktionsgefäßes (10),
einem im allgemeinen senkrecht angeordneten Ansorbergefäß (16), dessen oberer Teil gepackt ist, wobei das Gefäß eine Gasablaßöffnung (44) an seiner Oberseite und einen Lösungsmittelproduktauslaß (28) in Richtung auf seinen Boden aufweist,
einer Vorrichtung zur kontinuierlichen Rückführung von Lösungsmittel nach unten durch das Absorbergefäß (16) durch seinen gepackten Teil hindurch und
einer Trägergasüberführungsleitung (46), die zwischen dem Reaktions- und Absorbergefäß jeweils an einem Punkt unter den gepackten Teilen der jeweiligen Gefäße eine Verbindung bildet.

Revendications

1. Procédé pour préparer N_2O_5, comprenant les étapes consistant à :
   (a) fournir une solution de N_2O_4 dans un premier corps d'un solvant organique inerte volatil ;
   (b) mettre la solution en contact avec un véhicule gazeux porteur, contenant de l'ozone, à une température suffisante pour favoriser la formation de N_2O_5 dans le véhicule gazeux et l'évaporation du solvant et son passage dans le véhicule gazeux ;
   (c) mettre le véhicule gazeux, chargé de N_2O_5, en contact avec un second corps de solvant organique inerte à une température inférieure à celle de l'étape (b) pour y provoquer la condensation du N_2O_5, et
   (d) récupérer le N_2O_5 condensé au sein du solvant provenant de l'étape (c).

2. Procédé selon la revendication 1, caractérisé en ce que l'identité chimique des premier et second corps de solvant organique inerte est la même.

3. Procédé selon la revendication 1 ou la revendication 2, caractérisé en ce que le solvant des premier et/ou second corps de solvant comprend un chloroalcane en C_1 ou C_2 ou un chlorofluorocarbone.

4. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que le véhicule gazeux utilisé dans l'étape (b) contient un excès stoechiométrique d'ozone par rapport à la quantité nécessaire pour réagir avec le N_2O_4.

5. Procédé selon la revendication 4, caractérisé en ce que le véhicule gazeux contient un excès stoechiométrique d'ozone allant jusqu'à 5 %.

6. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que l'étape (b) est réalisée dans l'intervalle de la température compris entre -20 °C et +30 °C, de préférence entre -15 °C et +20 °C.

7. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que l'étape (c) est réalisée dans l'intervalle de la température allant de -15 °C à -70 °C.

8. Procédé selon la revendication 7, caractérisé en ce que l'étape (c) est réalisée dans l'intervalle de la température allant de -20 °C à -50 °C.

9. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que le produit de l'étape (d) dans du solvant est mélangé avec au moins une partie du produit dans du solvant de l'étape (b).

10. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que le premier corps de solvant et le véhicule gazeux sont fournis chacun sous forme d'un courant séparé.

11. Procédé selon la revendication 10, caractérisé en ce que le véhicule gazeux et le premier corps de solvant sont mis en contact, dans l'étape (b) en circulant dans le même sens.

12. Procédé selon la revendication 10 ou la revendication 11, caractérisé en ce que le premier corps de solvant est remis en circulation.

13. Procédé selon l'une quelconque des revendications 10 à 12, caractérisé en ce que le second corps de solvant est fourni sous la forme d'un courant séparé.

14. Procédé selon la revendication 13, caractérisé en ce que le véhicule gazeux et le second corps de solvant sont mis en contact, dans l'étape (c) en circulant à contre-courant.

15. Procédé selon la revendication 13 ou la revendication 14, caractérisé en ce que le second corps de solvant est remis en circulation.

16. Procédé selon l'une quelconque des revendications précédentes, caractérisé en ce que la concentration du N_2O_5 dans le premier corps de solvant servant dans l'étape (b) est de 0,005 à 0,05 % en poids.

17. Appareil pour mettre en oeuvre le procédé selon la revendication 1, cet appareil comprenant :

un récipient de réaction ou réacteur (10) disposé de façon générale verticalement et dont une partie supérieure est garnie, ledit récipient comportant vers sa partie inférieure une sortie (26) de produit dans le solvant et un moyen associé pour faire recirculer en continu le solvant en un courant descendant traversant la partie garnie de ce récipient ;

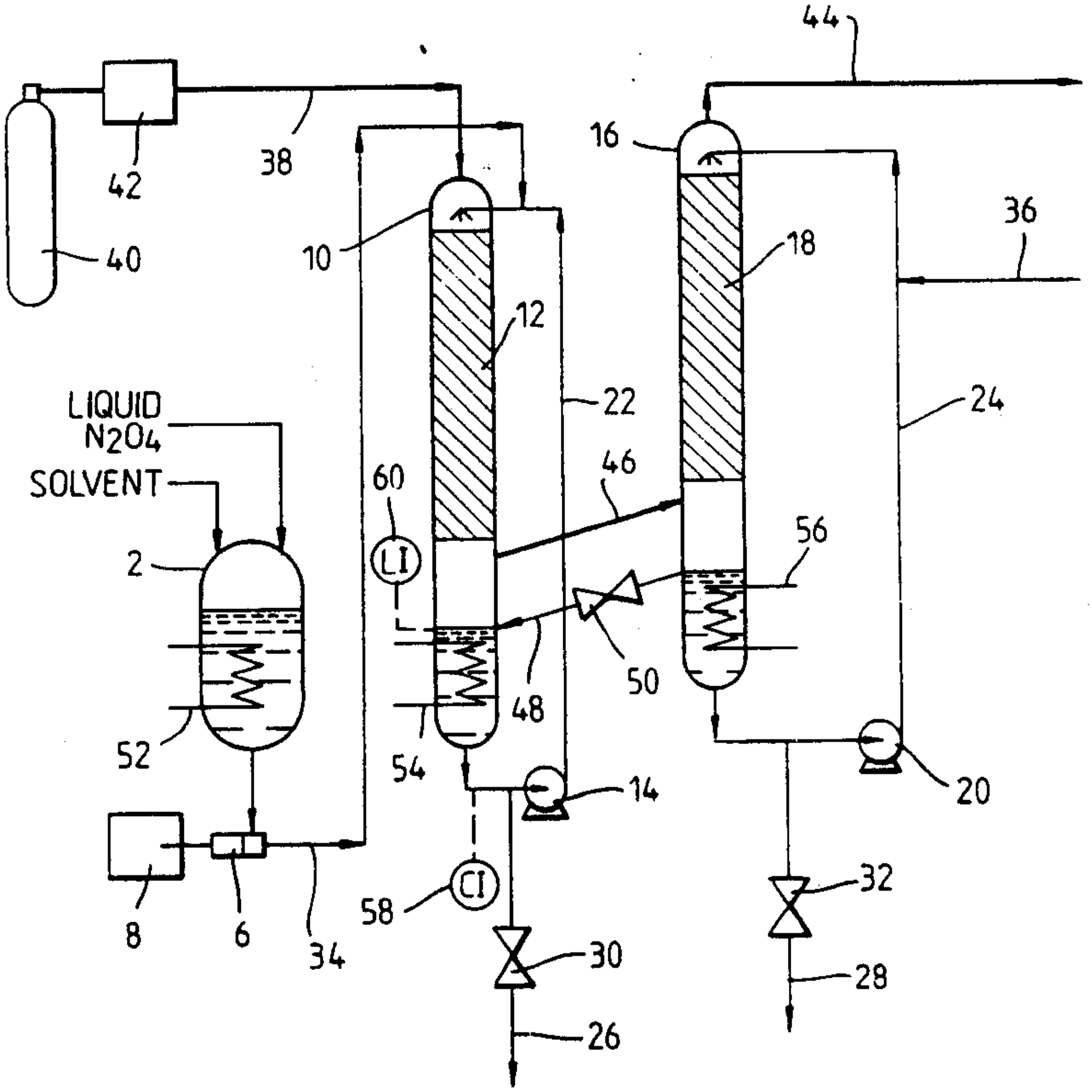
un moyen (42) engendrant de l'ozone et disposé de façon à fournir l'ozone introduit dans la partie supérieure dudit récipient (10) de réaction ou réacteur ;

un moyen (2, 6) pour fournir du solvant et du N_2O_4 introduits dans la partie supérieure dudit réacteur (10) ;

un récipient d'absorption (16), disposé de façon généralement verticale, dont une partie supérieure est garnie, ledit récipient ayant à sa partie supérieure un évent (44) de départ de gaz et vers sa partie supérieure une sortie (28) de produit dans du solvant ;

un moyen pour faire recirculer continuellement le solvant à travers la partie garnie de l'absorbeur (16), et vers le bas du récipient d'absorption (16) ; et

un conduit (46) de transfert de véhicule gazeux établissant la liaison entre les récipients de réaction et d'absorption, dans chaque cas en un point situé au-dessous de la partie garnie des récipients respectifs.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