(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
19 February 2004 (19.02.2004)

PCT

(10) International Publication Number
WO 2004/014542 A1

(51) International Patent Classification[7]: B01J 19/00, B01L 3/00

(21) International Application Number: PCT/NL2002/000537

(22) International Filing Date: 9 August 2002 (09.08.2002)

(25) Filing Language: English

(26) Publication Language: English

(71) Applicant (*for all designated States except US*): **AVANTIUM INTERNATIONAL B.V.** [NL/NL]; Zekeringstraat 29, NL-1014 BV Amsterdam (NL).

(72) Inventor; and

(75) Inventor/Applicant (*for US only*): **NAGY, Anton, John** [US/NL]; Lijnbaansgracht 165-II, NL-1016 VX Amsterdam (NL).

(74) Agent: **BROOKHUIS, H., J., A.**; Exter Polak & Charlouis B.V., P.O. Box 3241, NL-2280 GE Rijswijk (NL).

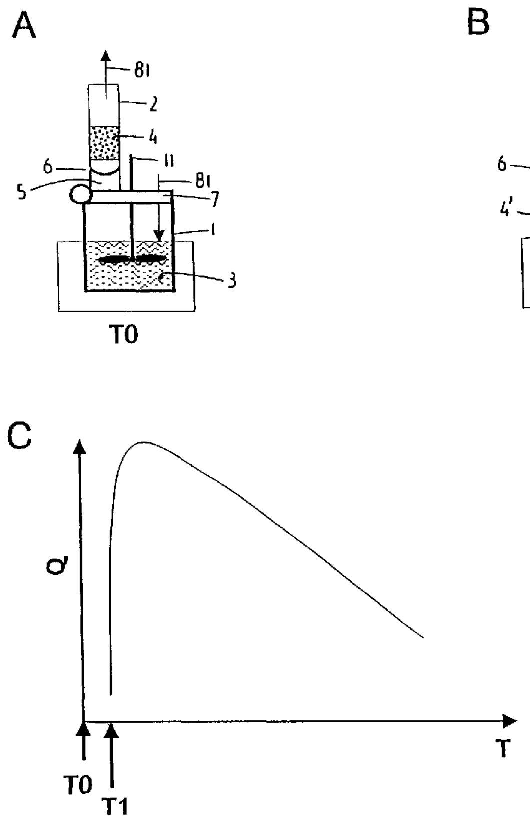
(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, OM, PH, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:
— *with international search report*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: CONDUCTING CHEMICAL EXPERIMENTS USING TWO INTERCONNECTED VESSELS



(57) Abstract: A method and device for conducting laboratory chemical experiments involving a first reactant and a second reactant, in particular a catalyst, comprising the following steps: introducing the first reactant into a reactor vessel and isolating it from the atmosphere; introducing the second reactant, in particular a catalyst, into a pre-treatment reactor and isolating it from the atmosphere, wherein the reactor vessel and the pre-treatment reactor are interconnected via a transfer passage provided with a closure that can be opened on command; pre-treating the second reactant in the pre-treatment reactor; and opening the closure so that the pre-treated second reactant, is introduced into the reactor vessel.

WO 2004/014542 A1

CONDUCTING CHEMICAL EXPERIMENTS USING TWO INTERCONNECTED VESSELS

The invention relates to a method and a device for conducting laboratory chemical experiments, involving a first reactant and a second reactant, in particular a catalyst.

In laboratory chemical experiments, at least two chemical substances or mixtures (so-called reactants) are brought together in order to react under predetermined process conditions. Often, these desired process conditions differ significantly from the conditions in the laboratory itself, as they may include for example an elevated process temperature, an elevated process pressure or the absence of oxygen and/or moisture. Moreover, these desired process conditions often have to be controlled accurately.

In current practice, the substances or mixtures participating in the reaction are first brought together in a reactor vessel. Then, the reactor vessel is closed and thus isolated from the atmosphere and brought to the desired process conditions. A significant disadvantage of this method is that the actual reaction is likely to start before the desired process conditions are reached. This affects the accuracy of the test results, which is especially adverse in (combinatorial) research experiments.

Another problem of the state of the art arises when at least one of the reactants requires a pre-treatment prior to the bringing it into contact with the other reactant(s). This pre-treatment often makes the pre-treated reactant sensitive to atmospheric influences, such as the presence of oxygen or humidity. In methods according to the state of the art, special precautions have to be taken in these cases, such as passivating the reactant by an air impermeable coating, such as a wax coating.

These disadvantages are encountered especially in combinatorial research experiments involving catalysts. These experiments often require a high process pressure and a high process temperature. Also, introducing the catalyst into the reactor vessel is complicated, especially since most catalysts are solids, often in a powdery form. Moreover, solid catalysts that are reduced as a pre-treatment, often oxidise rapidly upon contact with air or humidity, which severely complicates their introduction into a reactor vessel.

In a commonly used method for combinatorial research experiments involving a solid or powdered catalyst, the catalyst is first introduced into a pre-treatment reactor, in which it is pre-treated, e.g. reduced. In order to avoid re-oxidation by the oxygen in the atmosphere, the pre-treated catalyst is removed from the pre-treatment reactor inside a glove box in an environment of inert gas. In this glove box, the catalyst is passivated, e.g. by coating it with a layer of wax. The thus passivated catalyst is then transported through the atmosphere to a reactor vessel. After introducing the still coated catalyst into the reactor vessel, the reactor vessel is heated and pressurised to predetermined process conditions.

This method is very time-consuming and labour-intensive. Moreover, it requires complex inert-gas equipment. Due to the presence of the wax coating and due to that the catalyst is already at least partly in contact with the first reactant in the reactor vessel before the predetermined process conditions have been reached, the actual starting point of the reaction cannot always be accurately determined from the measured data that is collected during the experiment and the data from the first stage of the reaction is often not reliable.

In general, the current method is not suitable for efficiently and reliably conducting combinatorial research experiments.

It is the aim of the present invention to overcome one or more of the drawbacks mentioned above.

The present invention provides a method for conducting laboratory chemical experiments involving a first reactant and a second reactant, in particular a catalyst, comprising the following steps:

- introducing the first reactant into a reactor vessel and isolating it from the atmosphere,
- introducing the second reactant, in particular a catalyst into a pre-treatment reactor and isolating it from the atmosphere,
- wherein the reactor vessel and the pre-treatment reactor are interconnected via a transfer passage provided with a closure that can be opened on command,
- pre-treating the second reactant,
- opening the closure so that the pre-treated second reactant is introduced into the reactor vessel.

The present invention further provides a device for conducting laboratory chemical experiments, involving a first reactant and a second reactant, in particular a catalyst, in a reactor vessel, in particular a batch reactor vessel, which is isolated from the atmosphere. The device comprises a pre-treatment reactor, which is also isolated from the atmosphere, for storing a predetermined quantity of a second reactant, which quantity is adapted to the amount of second reactant that is required for an experiment. In the device according to the invention, the reactor vessel and the pre-treatment reactor are interconnected via a transfer passage provided with a closure that can be opened on command, so that the second reactant is introduced into the reactor vessel. The device according to the invention is characterised in that the device comprises pre-treatment means for pre-treating the second reactant, such as first flow means and second flow means.

The pre-treatment reactor can be designed as e.g. a fixed-bed reactor, an ebulating bed reactor, a fluidised bed reactor or a stirred tank reactor. The reactor vessel can be designed as e.g. a stirred tank reactor, a continuously stirred tank reactor, a fixed-bed reactor, an ebulating bed reactor or a fluidised bed reactor.

When conducting a laboratory chemical experiment using the method according to the invention, a first reactant is introduced in the reactor vessel and the contents of the reactor vessel is isolated from the outside atmosphere. A predetermined quantity of a second reactant is introduced in the pre-treatment reactor and the contents of the pre-treatment reactor is isolated from the outside atmosphere. The quantity of the second reactant introduced in the reactor vessel is adapted to the quantity of the second reactant that the experiment to be conducted requires.

The reactor vessel and the pre-treatment reactor are interconnected by a transfer passage provided with a closure. While the closure is shut, the reaction chambers of the reactor vessel and the pre-treatment reactor are separated from each other, so in each chamber a different process can take place.

In experiments involving catalysts, pre-treatment of the catalyst often is required. This pre-treatment has to take place before the catalyst is brought into contact with the first reactant. The pre-treatment often makes the pre-treated catalyst sensitive to atmospheric influences, such as the presence of oxygen or humidity, so measures have to be taken to avoid this influence.

In the method according to the invention, the pre-treatment takes place in the pre-treatment reactor, after the contents of the pre-treatment reactor is isolated from the atmosphere and while the closure in the transfer passage between the pre-treatment reactor and the reactor vessel is still shut.

After completion of the pre-treatment of the second reactant, the closure in the transfer passage is opened. Opening the closure establishes an open connection between the pre-treatment reactor and the reactor vessel via the transfer passage, allowing the catalyst to be displaced from the pre-treatment reactor via the transfer passage into the reactor vessel. Still, the interconnected chambers of the reactor vessel and the pre-treatment reactor and the transfer passage remain isolated from the atmosphere, so the pre-treated catalyst is not subject to atmospheric influences.

Advantageously, prior to adding the second reactant to the first reactant in the reactor vessel, the predetermined process conditions are substantially established in both the reactor vessel and the pre-treatment reactor. The advantage of this is that when the second reactant, in particular a catalyst, comes into contact with the first reactant, the predetermined process conditions are already present, so the start of the reaction takes place under the desired process conditions. This improves the quality of the measured data in the first stage of the reaction significantly.

More advantageously, the pressure in the pre-treatment reactor is somewhat higher than the pressure in the reactor vessel prior to opening the closure. This facilitates the introduction of the second reactant into the batch reactor, even when a high absolute pressure is present in the batch reactor.

Pre-treatment of a second reactant, in particular a catalyst, can involve processes such as reduction, calcination, steam treatment or coke formation. Other processes are also possible. Such pre-treatments often require a fluid (such as a gas, a vapour or a liquid) flowing into or through the pre-treatment reactor. Therefore, preferably a fluid is supplied to the pre-treatment reactor for the pre-treatment of the second reactant.

For example, pre-reducing the second reactant can be performed by using an H_2-gas flow through the pre-treatment reactor.

As already mentioned, it is very advantageous to bring at least the reactor vessel to essentially the desired process conditions before starting the reaction by bringing the second reactant into contact with the first reactant. The desired process conditions usually can be at least partially established by means of a fluid flowing into or through the reactor vessel. For example a gas flow can be used to pressurise, heat, cool or purge the reactor vessel.

More advantageously, the pre-treatment reactor is also brought to essentially the desired process conditions before allowing the second reactant to leave the pre-treatment reactor. The desired process conditions usually can be at least partially established by means of a fluid flowing into or through the pre-treatment reactor. Advantageously, the same flow means are used for pre-treating the second reactant and for conditioning the pre-treatment reactor.

Sometimes it can be required to pre-treat the first reactant prior to the experiment. In those cases, preferably the flow means used for conditioning the reactor vessel are used for the pre-treatment of the first reactant as well.

As a variant, it is conceivable to integrate the flow means of the reactor system in such a way that pre-treatment in and the conditioning of both the reactor vessel and the pre-treatment vessel can be carried out by a single flow means system.

Preferably, the closure in the transfer passage between the pre-treatment reactor and the reactor vessel is opened when a predetermined pressure difference is applied between the pre-treatment reactor and the reactor vessel. Such a closure can be for example a valve, a plug seated in a corresponding seat or, most preferably, a frangible seal. Opening the closure can be realised by using dedicated pressure means to establish the required pressure difference. Advantageously, the flow means used for pre-treatment and/or conditioning are used to establish the required pressure difference.

In an alternative embodiment, a screw feed is used to transfer the second reactant from the pre-treatment reactor to the reactor vessel. The use of a screw feed has the advantage of allowing a controlled transfer of material over time from the pre-treatment reactor to the reactor vessel.

In a preferred embodiment, displacement of the second reactant from the pre-treatment reactor to the reactor vessel is realised by establishing a pressure difference between the pre-treatment reactor and the reactor vessel. In that case, the pressure in the pre-treatment reactor is set higher than the pressure in the reactor vessel at the time of introducing the second reactant, in particular a catalyst into the reactor vessel.

If the second reactant is a solid or a liquid, the pre-treatment reactor is preferably positioned at a level above the reactor vessel, so that the displacement of the second reactant from the pre-treatment reactor to the reactor vessel is realised or at least assisted by gravity.

Alternatively, said displacement is mechanically induced by use of a plunger, which is sealed against the atmosphere. This plunger is for example operated magnetically, allowing the internal plunger to be effectively sealed from the outside atmosphere.

Preferably, the quantity of second reactant, in particular a catalyst, introduced in the pre-treatment reactor is the quantity the single experiment to be conducted requires. Upon opening of the closure in the transfer passage, generally all the second reactant, in particular a catalyst will be introduced in the reactor vessel.

In an alternative embodiment, the second reactant, in particular a catalyst is dosed to the reactor vessel with a particular feeding rate profile during the experiment. This includes, but is not limited to, ramps and steps in the feeding rate. This is particularly important for cases where the second reactant is gradually consumed in the reactor vessel and must be continuously or intermittently replaced.

It is envisaged that the pre-treatment reactor and/or the reactor vessel are additionally equipped with for example heating means, cooling means, means for measuring and/or monitoring reaction parameters, and/or thermal insulation. The reactor vessel and/or the pre-treatment reactor are envisaged to be suitably equipped for calorimetry.

Possibly, multiple pre-treatment reactors are associated with a reactor vessel. Moreover, it is envisaged the method according to the invention is applied to an array of reactors to conduct combinatorial research experiments.

Advantageously, the pre-treatment reactor is detachably secured to the reactor vessel, for example by means of a screw thread connection. For example, the reactor vessel has a lid, and the pre-treatment reactor is screwed onto that lid.

It is envisaged that the connection between the pre-treatment reactor and the reactor vessel by means of a transfer passage with a closure will be established after pre-treatment and/or conditioning. In that case, advantageously quick-fit connectors are applied, the first part of the connector being attached to the reactor vessel, the second part of the connector being attached to the pre-treatment reactor. Most advantageously connectors are used of the type that open the vessel or reactor they are attached to when the connection between the first part of the connector and the second part of the connector is established and close that vessel or reactor when the connection between the first part of the connector and the second part of the connector is broken.

The method and exemplary embodiments of the device according to the invention will be described in detail below, with reference to the drawing.

The drawing shows in:

Fig. 1: the method for performing laboratory experiments involving a second reactant, in particular a catalyst according to the prior art,

Fig. 2: the problem of unreliable process data at the start of the reaction, as encountered when using the prior art method,

Fig. 3: the method according to the invention,

Fig. 4: diagrammatical representation of a device according to the invention, suitable for use with the method according to the invention.

Fig. 5: a general layout (diagrammatically) of a device according to the invention

Fig. 6: shows a different exemplary embodiment of a device according to the invention.

Figure 1 shows the method according to the prior art for pre-treating a solid second reactant, in particular a catalyst 4, in this example in powdered form, and using it in a batch reaction.

Figure 1A shows the first step in which a catalyst powder 4 is introduced in a pre-treatment reactor 2. Then, the catalyst 4 is reduced in flowing H_2 at an elevated temperature and pressure, as figure 1B illustrates.

As the pre-treated catalyst 4′ rapidly re-oxidizes in air, the pre-treated catalyst 4′ is removed from the pre-treatment reactor 2 under inert-gas atmosphere, such as in a glove box 80 (figure 1C). There, the catalyst 4′ is then passivated by coating it with molten wax (figure 1D).

After the wax is solidified, the pre-treated, passivated catalyst 4′′ is removed from the glove box 80, and transported in the laboratory environment to the reactor vessel 1 (figure 1E). The first reactant 3 is introduced in the reactor vessel 1.

The pre-treated, passivated catalyst 4′′ is then introduced into the reactor vessel 1. The reactor vessel 1 is brought to the desired process conditions, which involves heating and pressurizing. Meanwhile the wax coating 82 starts to melt. Where contact between the pre-treated catalyst 4′ and the first reactant 3 occurs, the reaction starts.

This method is very time-consuming and labour-intensive. Moreover, it requires complex and expensive inert-gas equipment.

Figure 2 illustrates the problem of the unreliability of measurement data obtained around the start of the reaction, when the method according to the prior art is used.

Figure 2A shows the pre-treated, passivated catalyst 4′′ is added to the reactor vessel 1, in which the first reactant is already present. Then, the reactor vessel 1 is closed, and pressurised and heated, for example by means of a fluid 81.

During the heating of the reactor vessel 1, the wax coating 82 of the pre-treated, passivated catalyst 4′′ starts to melt. So, bit by bit, the pre-treated catalyst 4″ comes into contact with the first reactant 3. Wherever this contact is established, the reaction starts. At locations where the coating 82 is still present, no reaction takes place. This causes a gradual start of the reaction, taking place during a starting period (as opposed to a starting point). During this starting period, it is not known how much of the second reactant, in particular a catalyst is already taking part in the reaction.

Moreover, it takes some time to obtain the desired process conditions (regarding e.g. temperature, pressure) inside the reactor vessel, starting at the situation of figure 2A. As during heating and pressurising, the wax coating 82 of the pre-treated catalyst 4′ already starts to melt, the reaction also starts before the desired process conditions are reached.

Therefore, the measurement data collected during heating and pressurising are not reliable since the process conditions, including the quantity of the reactants that actually take part in the reaction differ significantly from the desired process conditions.

Figure 2C illustrates typical measurement results obtained from an experiment conducted in a way according to the prior art. On the horizontal axis of the graph, time is plotted, while a reaction parameter, such as conversion rate, for example, is plotted on the vertical axis. During time period X the heating and pressurising is on the way, while during period Y, the reaction takes place under constant temperature and pressure, and with all of the catalyst 4′ taking part in the reaction. The graph represented in figure 2C clearly shows the influence of the different process conditions during period X and period Y.

For these reasons, the method according to the prior art is not suitable conducting combinatorial research experiments.

Figure 3 illustrates in brief the method according to the invention.

Figure 3A shows that a first reactant 3 is introduced into a reactor vessel 1 and isolated from the atmosphere. A catalyst 4 is introduced pre-treatment reactor 2 and isolated from the atmosphere. The reactor vessel 1 and the pre-treatment reactor 2 are interconnected via a transfer passage 5 provided with a closure 6 that can be opened on command.

The catalyst 4 is reduced by means of a H_2-gas flow through the pre-treatment reactor 2. The same H2-gas flow is used here to pressurise and heat the reactor vessel 1 and the content of said reactor vessel 1.

Figure 3B shows that the closure 6 in the transfer passage is opened so that the pre-treated catalyst 4′ is introduced directly into the conditioned reactor vessel 1. Since contact with the outside atmosphere is avoided, passivation is not necessary.

Figure 3C shows typical measurement data obtained from a reaction when the method according to the invention is used. Comparing figure 3C to figure 2C, that shows typical measurement data obtained from a reaction conducted by using a method according to the prior art, it is clear that when the method according to the invention is applied, all measured data is relevant for the evaluation of the experiment.

As an example illustrating the method according to the invention, in a hydrogenation reaction, a pre-reduced solid catalyst may be heated to the reaction temperature and pressurised with hydrogen to the reaction pressure in the pre-treatment reactor. The reactor vessel containing an organic reactant in liquid form to be hydrogenated, is simultaneously heated and pressurised to the desired temperature and pressure for reaction. When both the pre-treatment reactor and the reactor vessel containing the organic reactant are equilibrated at the desired conditions, the catalyst may be injected into the reactor. This provides a very clear starting point for reaction. This is particularly important when performing large numbers of experiments in parallel where as much of the hydrogenation curve is needed as possible to quantitatively compare catalyst performance in different reactors.

In another embodiment, the process conditions in the reactor vessel are set to compensate a process response that is expected in the reactor vessel, occurring upon injection of the second reactant from the pre-treatment reactor into the reactor vessel. For example, the temperature of the first reactant in the reactor vessel may kept at a value lower than the target set point reaction in order to compensate for the exothermal reaction (that produces energy), which occurs upon injection of a second reactant into the first reactant.

Fig. 4 shows an overview of a device according to the invention. The device comprises a reactor vessel 1 for receiving a first reactant 3 and a pre-treatment reactor 2 for receiving a second reactant, in particular a catalyst. The reactor vessel 1 and the pre-treatment reactor 2 are interconnected by a transfer passage 5, which is provided with a closure. In the embodiment shown in Fig. 4, the closure is a frangible seal 6, and the transfer passage 5 and the frangible seal 6 are integrated in the pre-treatment reactor. The frangible seal 6 could be a bursting or rupture disc, e.g. a metal member, a graphite member, a glass member, a plastic member or a quartz member. Also, other materials that are suitable for the specific process conditions can be used.

The reaction chamber of the reactor vessel 1 can be isolated from the atmosphere by closing lid 7, which is provided with sealing means 15 such as an O-ring. Pre-treatment reactor 2 is screwed onto the lid 7 by means of screw thread 9, that provides an airtight closure. The pre-treatment chamber of the pre-treatment reactor 2 is isolated from the atmosphere by means of lid 8.

In an advantageous application of the embodiment shown in Fig.4, the second reactant is a catalyst 4, e.g. in a powdery form. In the pre-treatment reactor 2, the catalyst 4 is held at a distance from the frangible seal 6 by support 10, which is made of quartz wool here.

Fig. 5 shows diagrammatically a general layout of the device according to the invention, comprising pre-treatment means for the pre-treatment of the a catalyst 4 in the pre-treatment reactor 2, comprising second flow means 20.

The second flow means 20 here comprise a reservoir 21 for a fluid, a flow controller 25 (such as a mass-flow controller, a volumetric flow controller or any kind of pump), a supply line 22, a discharge line 23, a pressure control valve 24, an inlet 27 and an outlet 28.

The second flow means 20 is adapted to create a flow of a fluid such as a gas, a vapour or a liquid, through the pre-treatment reactor 2. This flow can be required for a chemical pre-treatment of a batch of catalyst 4 in the pre-treatment reactor 2, such as reduction, calcination, stream treatment or coke formation, but it can also be used to bring the pre-treatment reactor 2 to substantially the predetermined process conditions for the reaction.

Reduction of the catalyst is often a desired pre-treatment. In the shown embodiment, this pre-treatment can for instance be carried out by an H_2-gas flow through the pre-treatment reactor 2. Bringing pre-treatment reactor 2 to the predetermined process conditions may include heating, cooling, pressurising, or purging in order to obtain a specific environment (e.g. oxygen-free, or an inert gas atmosphere). This could be achieved by the same H_2-gas flow as used for pre-treatment.

The device shown in Fig. 5 also comprises first flow means 30. The first flow means 30 here comprise a reservoir 31 for a fluid, a flow controller 35 (such as a mass-flow controller, a volumetric flow controller or any kind of pump), a supply line 32, a discharge line 33, a pressure control valve 34, an inlet 37 and an outlet 38.

The first flow means 30 is used to create essentially the predetermined process conditions in the batch reactor 1, by heating, cooling, pressurising or purging the reactor vessel 1 by means of a flow of a fluid such as a gas, a vapour or a liquid.

In the embodiment of the device shown in fig. 4, the closure 6 in the transfer passage 5 is a frangible seal. Such a seal is adapted to break when a predetermined pressure difference is present between two sides of the seal 6. In cases such as this, wherein the closure 6 is opened when a predetermined pressure difference is applied between the reactor vessel and the pre-treatment reactor, the first flow means 30 and the second flow means 20 advantageously cooperate in applying this predetermined pressure difference.

In a different embodiment that is not shown in the drawing, the closure is a valve, e.g. a valve which is normally closed under spring action. When a pressure difference between the reactor vessel and the pre-treatment reactor is created that is sufficiently large, the valve will open against the spring action. This embodiment is suitable in those cases where contamination of the closure by the second reactant will not hinder the operation of the moving parts of the closure, such as when the second reactant is a gas or a liquid.

In another exemplary embodiment, that is not shown in the drawing, the closure is a plug, seated in a corresponding seat. This plug is preferably displaced from its seat by applying a pressure difference between the pre-treatment reactor and the reactor vessel. In that case, the pressure difference between the pre-treatment reactor and the reactor vessel has to be large enough to overcome the retaining force between the plug and its seat.

As is shown in Fig. 4, the reactor vessel 1 can be equipped with a stirrer 11 driven by a motor 14, and the pre-treatment reactor

2 can be provided with heating means 12 and thermal insulation 13. Also, measuring and/or monitoring devices may be present in the reactor vessel and/or the pre-treatment reactor.

Fig. 6 shows a different possible embodiment of device according to the invention.

Initially, a pre-treatment gas flows from a reservoir 41 via a flow controller 45 to the supply lines 42 and 43. Valve 46 is open, so the batch reactor vessel 1 acquires essentially the same pressure as the supply line 42. Meanwhile, gas flows via supply line 43, capillary 48 through batch storage vessel 2, where the catalyst 4 is pre-treated, for example reduced. Pressure control valve 47 controls the pressure in the system.

When the pre-treatment of the catalyst 4 has been completed, valve 49 is closed. The system is then pressurised to essentially the predetermined process pressure. Then, valve 46 is closed, and the pressure in the batch storage vessel is increased to break the frangible seal 6, in order to introduce the catalyst 4 into the batch reactor vessel.

In a different embodiment that is not shown in the drawing, a reactor vessel may be associated with multiple pre-treatment reactors.

In a different embodiment that is not shown in the drawing, an array of devices according to the invention may be used for conducting combinatorial research experiments.

CLAIMS

1. Method for conducting laboratory chemical experiments involving a first reactant and a second reactant, in particular a catalyst, comprising the following steps:
- introducing the first reactant into a reactor vessel and isolating the first reactant from the atmosphere,
- introducing the second reactant, in particular a catalyst, into a pre-treatment reactor and isolating the second reactant from the atmosphere, wherein the reactor vessel and the pre-treatment reactor are interconnected via a transfer passage provided with a closure that can be opened on command,
- pre-treating the second reactant in the pre-treatment reactor, and
- opening the closure so that the pre-treated second reactant is introduced into the reactor vessel.

2. Method according to claim 1, characterised in that, prior to opening the closure in the transfer passage, the first reactant is brought to desired process conditions.

3. Method according to claim 2, characterised in that, pre-treatment of the second reactant involves bringing the second reactant to essentially identical process conditions as the first reactant.

4. Method according to claim 2 or 3, characterised in that, the desired process conditions comprise an elevated process temperature.

5. Method according to claim 2, 3 or 4, characterised in that,
the desired process conditions comprise an elevated process pressure.

6. Method according to any of the preceding claims, characterised in that, a fluid, such as a gas, a vapour or a liquid is supplied to the pre-treatment reactor for the pre-treatment of the second reactant.

7. Method according to claim 6, characterised in that, the pre-treatment fluid is also used for pressurising the pre-treatment reactor and/or the reactor vessel.

8. Method according to claim 6 or 7, characterised in that, the pre-treatment fluid is also used for heating the pre-treatment reactor and/or the reactor vessel.

9. Method according to any of the preceding claims, characterised in that, the pre-treatment comprises reducing the second reactant, in particular a catalyst.

10. Method according to claim 9, characterised in that, the reducing pre-treatment of the second reactant is carried out by means of a reducing gas, supplied to the pre-treatment reactor.

11. Method according claim 10, characterised in that, the reducing gas is H_2.

12. Method according to any of the preceding claims, characterised in that, the pre-treatment of the second reactant, in particular a catalyst, involves calcination.

13. Method according to any of the preceding claims, characterised in that, the pre-treatment of the second reactant, in particular a catalyst, involves steam treatment.

14. Method according to any of the preceding claims, characterised in that, the pre-treatment of the second reactant, in particular a catalyst, involves coke formation.

15. Method according to any of the preceding claims, characterised in that, the second reactant is a solid, e.g. in a powdered form.

16. Method according any of the preceding claims, characterised in that, the pre-treatment reactor is of the fixed-bed type.

17. Method according to any of the preceding claims, characterised in that, multiple experiments are carried out in an array of reactor vessels, each having one or more associated pre-treatment reactors.

18. Method according to any of the preceding claims, wherein the closure in the transfer passage is of the type which opens when a predetermined pressure difference is applied between the pre-treatment reactor and the reactor vessel, and in which said predetermined pressure difference is applied in order to open the closure in the transfer passage on command.

19. Method according to claims 6 and 18, characterised in that, the fluid used for pre-treatment is also used to apply the predetermined pressure difference between the pre-treatment reactor and the reactor vessel in order to open the closure in the transfer passage.

20. Method according to claim 2, characterised in that, a fluid is used to bring the first reactant to the desired process conditions.

21. Method according to claims 18 and 20, characterised in that, the fluid that is used to bring the first reactant to the desired process conditions, is also used to apply the predetermined pressure difference between the pre-treatment reactor and the reactor vessel in order to open the closure in the transfer passage.

22. Method according to claims 18, 19 and 21, characterised in that, the fluid used for pre-treatment and the fluid used to bring the reactor vessel to the desired process conditions cooperate to apply the predetermined pressure difference between the pre-treatment reactor and the reactor vessel in order to open the closure in the transfer passage.

23. Method according any of the preceding claims, characterised in that, the pre-treatment reactor and the reactor vessel are interconnected after introducing the first reactant into a reactor vessel and isolating it from the atmosphere and introducing the second reactant into the pre-treatment reactor and isolating it from the atmosphere.

24. Method according any of the preceding claims, characterised in that, the pre-treatment reactor and the reactor vessel are interconnected after introducing the first reactant into a reactor vessel and isolating it from the atmosphere and introducing the second reactant into the pre-treatment reactor and isolating it from the atmosphere.

25. Method according any of the preceding claims, characterised in that, the pre-treatment reactor and the reactor vessel are interconnected after pre-treatment of the second reactant.

26. Device for conducting laboratory chemical experiments, involving a first reactant and a second reactant, in particular a catalyst, in a reactor vessel, in particular a batch reactor vessel, which is isolated from the atmosphere,
which device further comprises a pre-treatment reactor, which is also isolated from the atmosphere, for storing a predetermined quantity of a second reactant in particular a catalyst,
and in which the reactor vessel and the pre-treatment reactor are interconnectable via a transfer passage provided with a

closure that can be opened on command, so that the second reactant is introduced into the reactor vessel, **characterised in that**, the device comprises pre-treatment means for pre-treating the second reactant in the pre-treatment reactor while isolated from the atmosphere.

27. Device according to claim 26, characterised in that, the device comprises pressure means for applying a pressure difference between the batch storage vessel and the batch reactor vessel in order to open the closure.

28. Device according to claim 26 or 27, characterised in that, a screw feed means is provided for the transfer of the second reactant from the pre-treatment reactor to the reactor vessel.

29. Device according to any of the claims 26-28, characterised in that, the pre-treatment reactor is preferably positioned at a level above the reactor vessel, so that the displacement of the second reactant from the pre-treatment reactor to the reactor vessel is realised or at least assisted by gravity.

30. Device according to any of the claims 26-29, characterised in that, a plunger is provided in the pre-treatment reactor for the displacement of the second reactant from the pre-treatment reactor to the reactor.

31. Device according to claim 30, characterised in that, the plunger is operated magnetically.

32. Device according to any of the claims 26-31, characterised in that, upon opening of the closure in the transfer passage, substantially all the second reactant that is present in the pre-treatment reactor will be introduced in the reactor vessel.

33. Method according to any of the preceding claims, characterised in that, the second reactant is supplied to the reactor vessel with a particular feeding rate profile during the experiment.

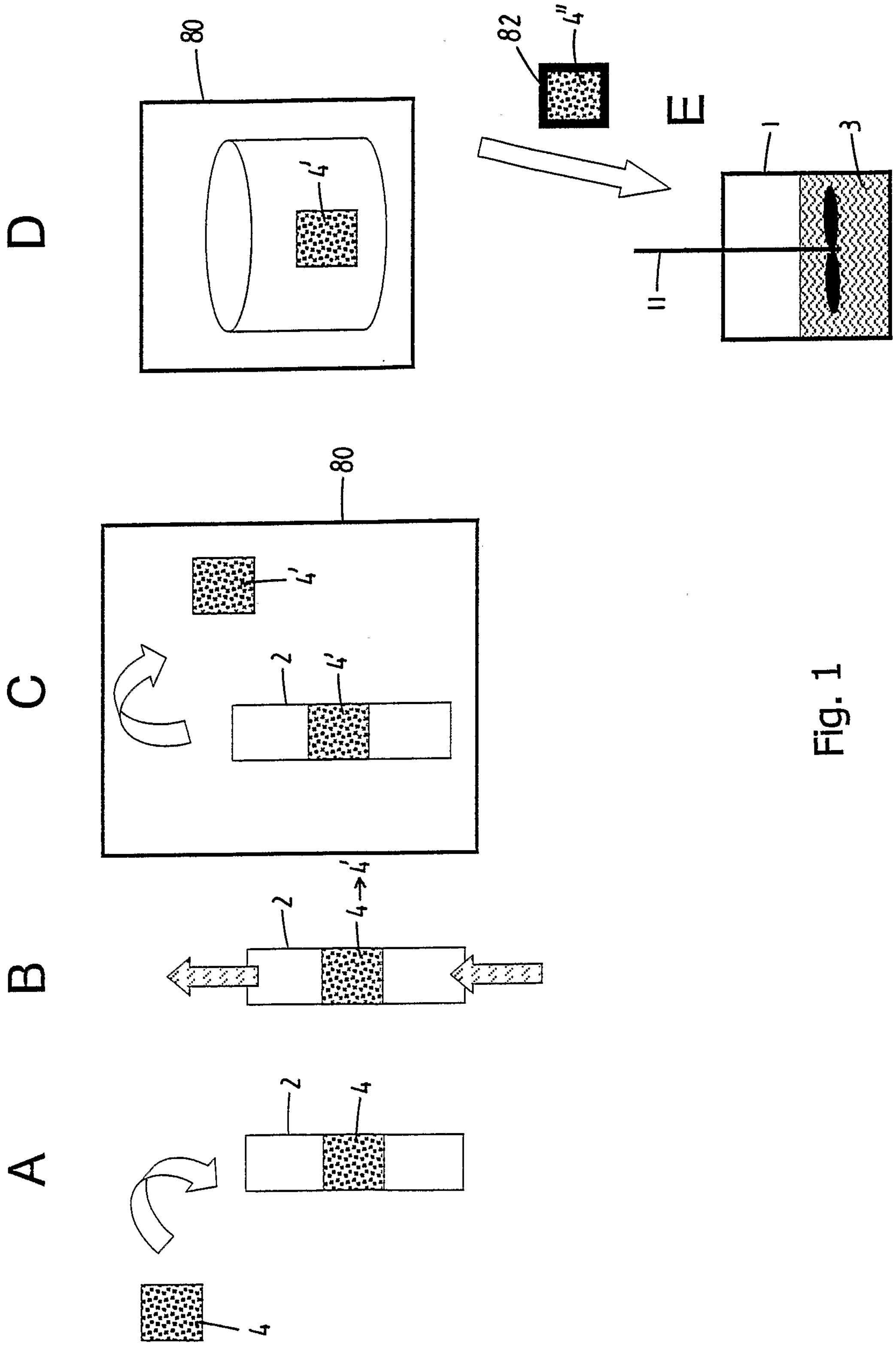


Fig. 1

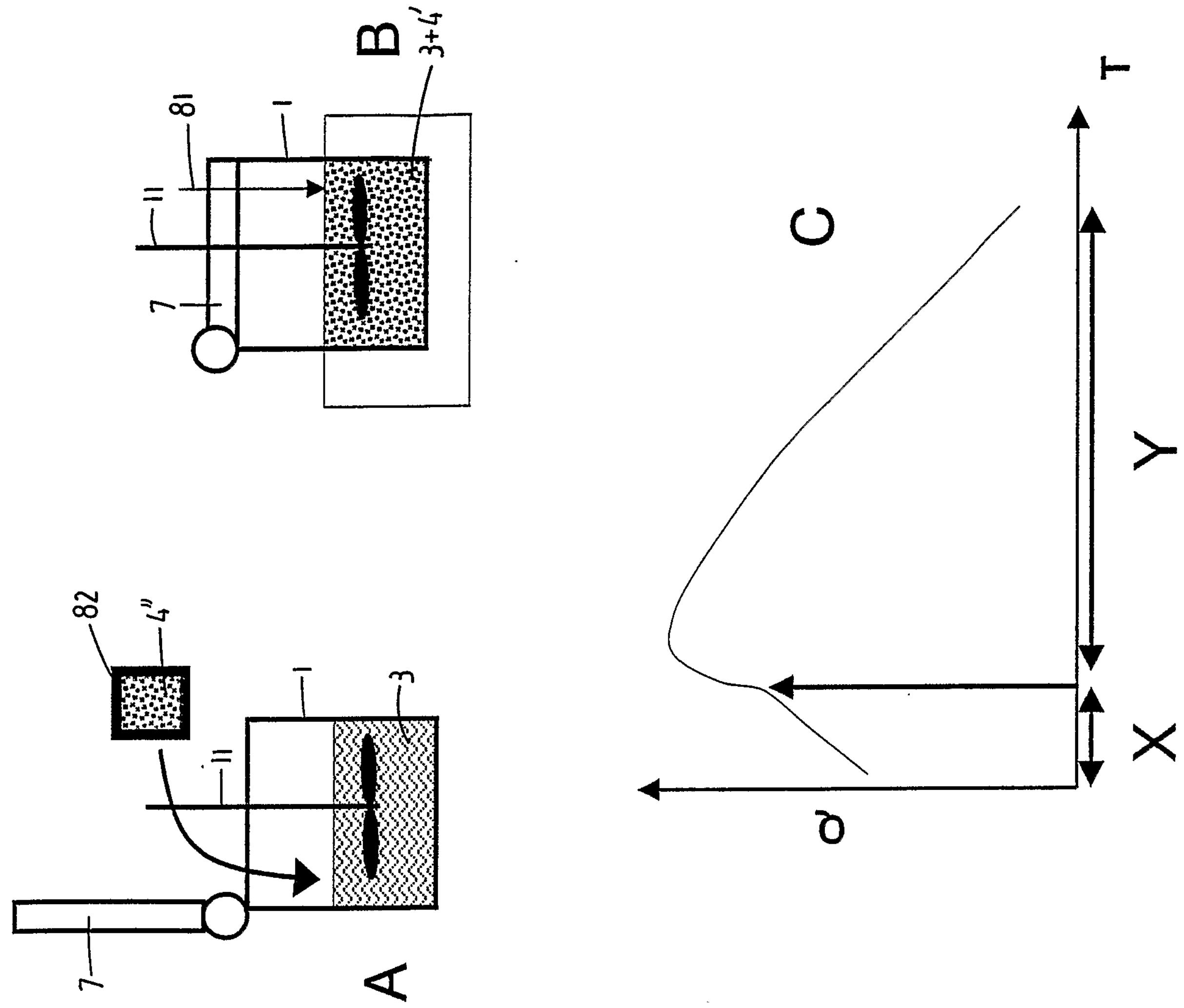


Fig. 2

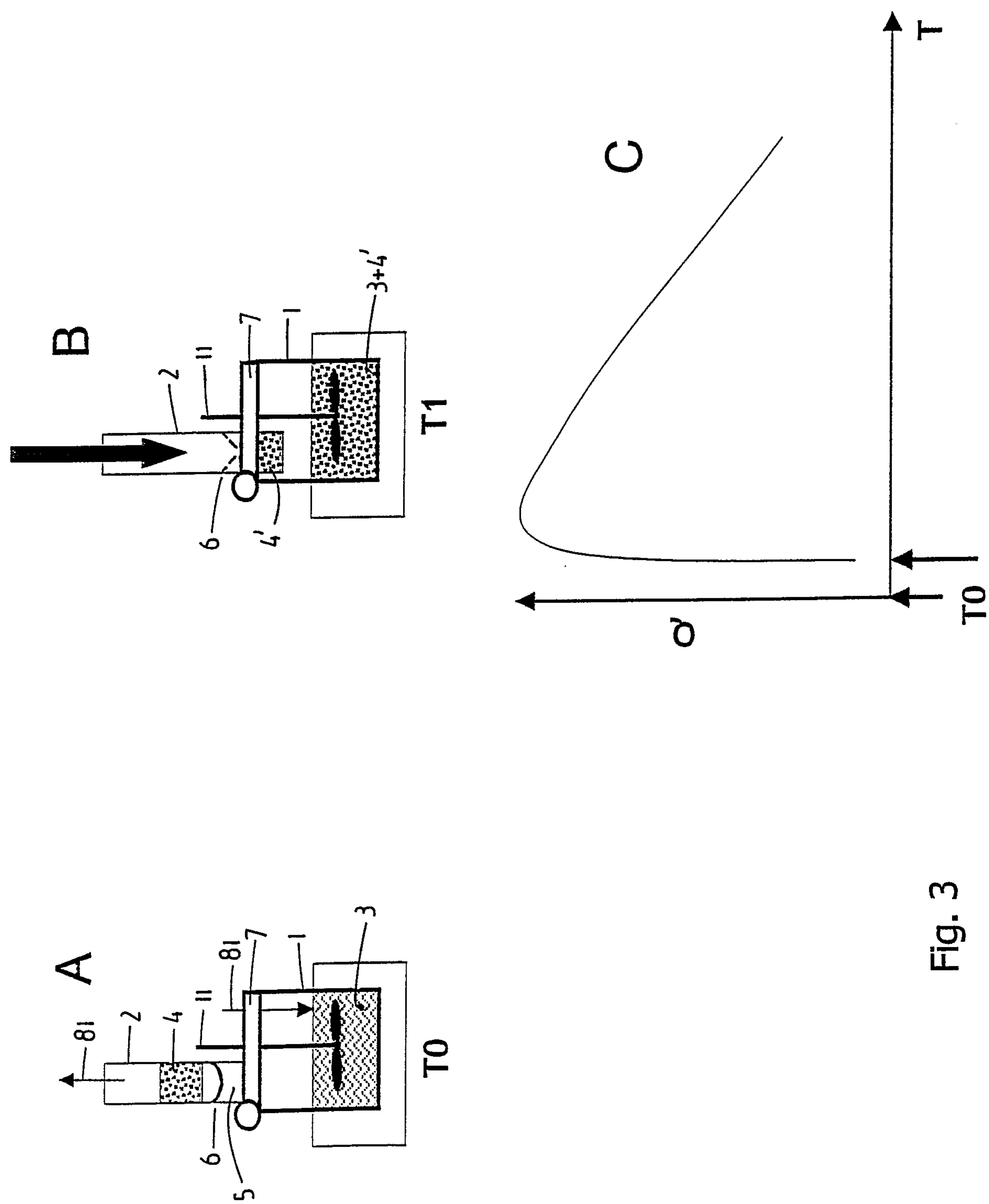


Fig. 3

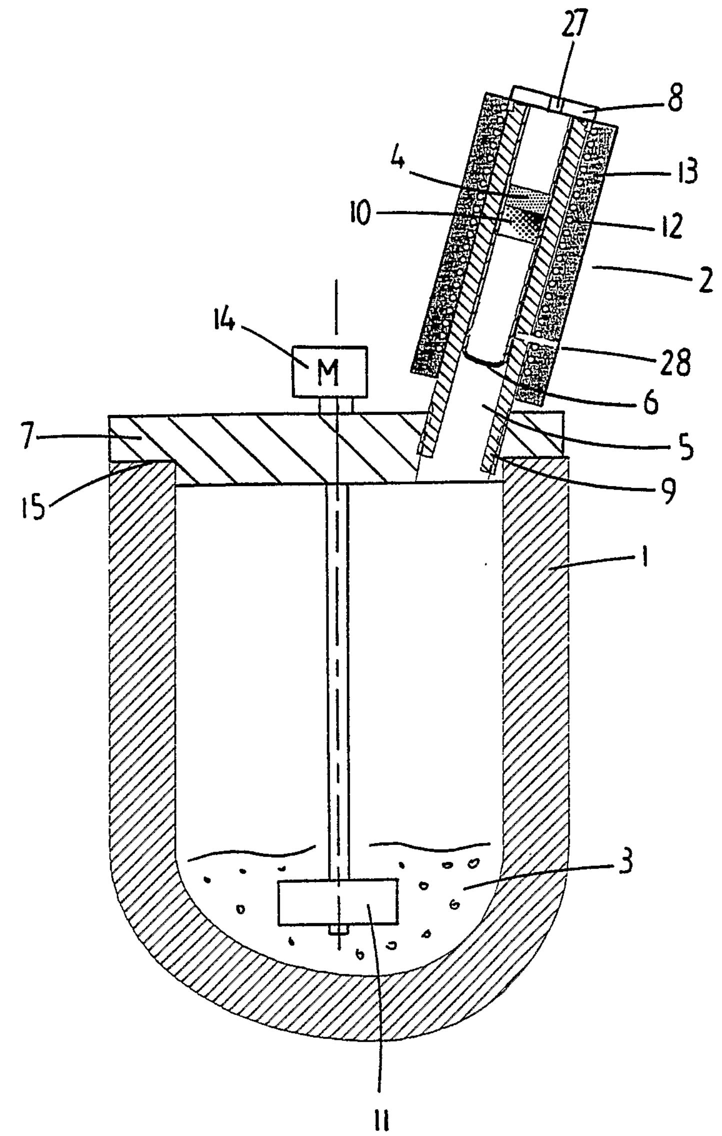


Fig. 4

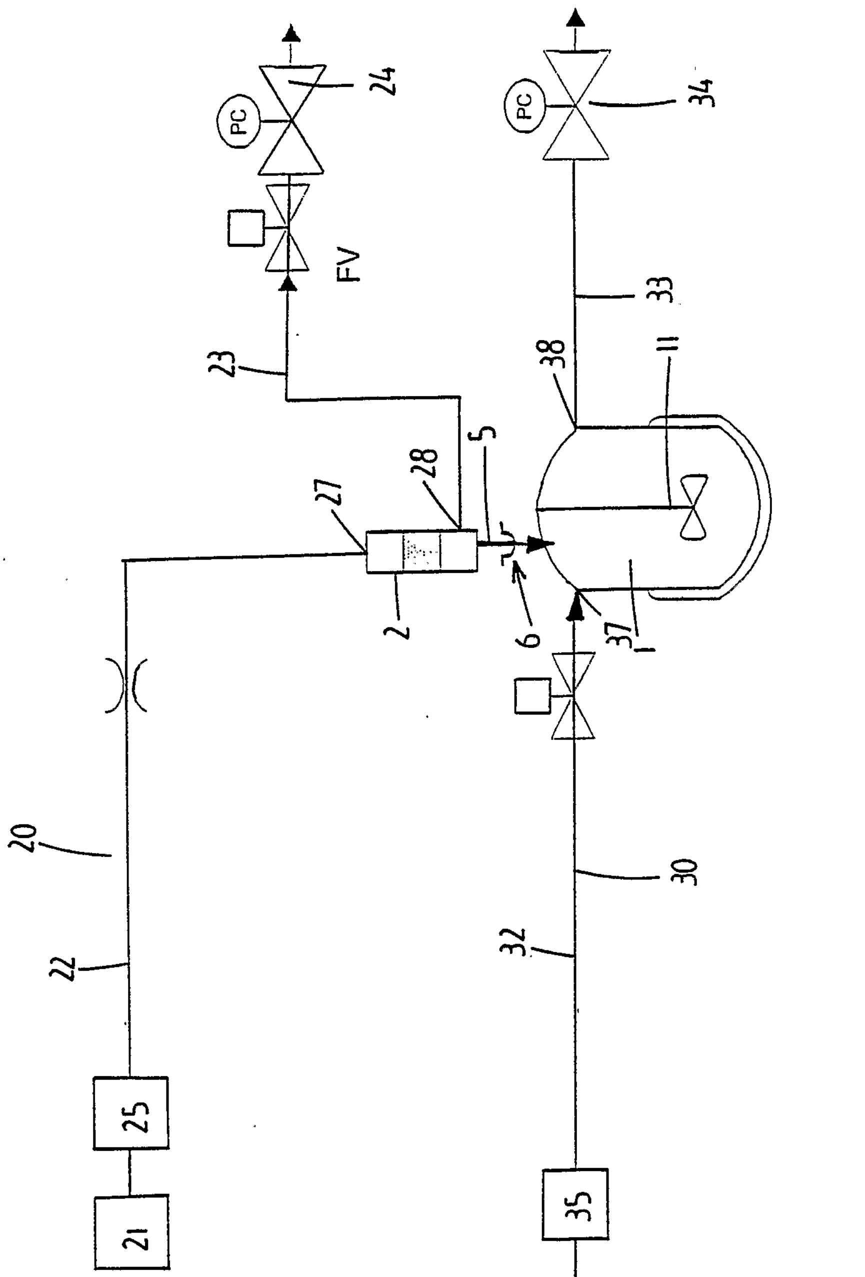


Fig. 5

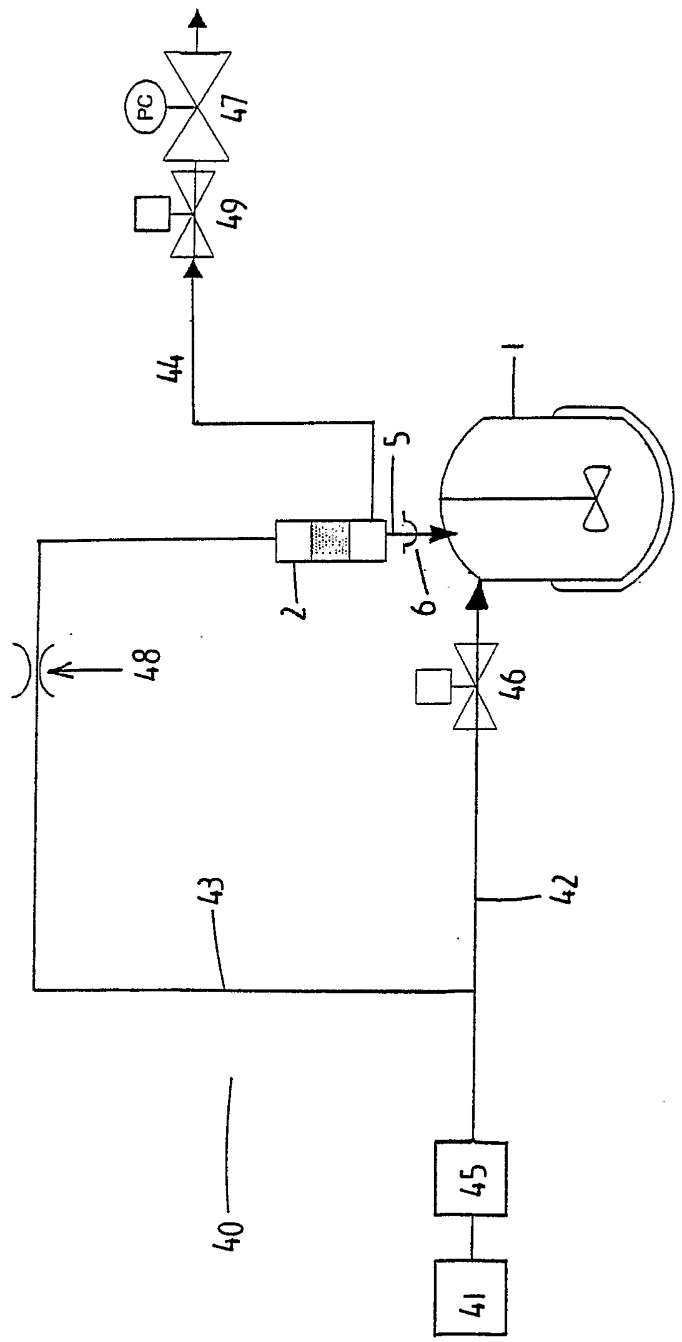


Fig. 6

INTERNATIONAL SEARCH REPORT

International Application No
PCT/NL 02/00537

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B01J19/00 B01L3/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B01J B01L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 01 36087 A (KOBYLECKI RYSZARD ;BRUCE MICHAEL (GB); PARTINGTON ROY (GB); MILLEN) 25 May 2001 (2001-05-25) page 28, line 18 -page 30, line 30; figures 1,3,4	1-33
X	WO 02 43854 A (HTE AG ;SCHUNK STEPHAN ANDREAS (DE)) 6 June 2002 (2002-06-06) page 9, line 14 -page 11, line 16; claim 15; figures 3-7	26

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
9 April 2003	23/04/2003

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Veefkind, V

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	STOREY R F ET AL: "A MODIFIED GLASS REACTOR FOR THE PREPARATION OF LINEAR AND STAR- BRANCHED BLOCK COPOLYMERS VIA LIVING ANIONIC POLYMERIZATION" JOURNAL OF APPLIED POLYMER SCIENCE, JOHN WILEY AND SONS INC. NEW YORK, US, vol. 66, no. 1, 3 October 1997 (1997-10-03), pages 151-159, XP000802670 ISSN: 0021-8995 the whole document ---	1,26
X	DATABASE WPI Section Ch, Week 199641 Derwent Publications Ltd., London, GB; Class J04, AN 1996-406592 XP002237780 -& JP 08 196897 A (TAKEDA CHEM IND LTD), 6 August 1996 (1996-08-06) abstract; figures 2-4 ---	1,26
A	WO 98 10866 A (ALPHAHELIX AB ;MALMQVIST MATS (SE)) 19 March 1998 (1998-03-19) the whole document -----	1,26

INTERNATIONAL SEARCH REPORT

Int onal Application No
PCT/NL 02/00537

Patent document cited in search report		Publication date	Patent family member(s)			Publication date
WO 0136087	A	25-05-2001	AU	1529601	A	30-05-2001
			EP	1230011	A1	14-08-2002
			WO	0136087	A1	25-05-2001
			GB	2372506	A	28-08-2002
WO 0243854	A	06-06-2002	DE	10059633	A1	20-06-2002
			AU	1707902	A	11-06-2002
			WO	0243854	A2	06-06-2002
			US	2002089868	A1	11-07-2002
JP 8196897	A	06-08-1996	NONE			
WO 9810866	A	19-03-1998	AT	220951	T	15-08-2002
			AU	725668	B2	19-10-2000
			AU	4406897	A	02-04-1998
			CN	1230904	A ,B	06-10-1999
			DE	69714269	D1	29-08-2002
			DE	69714269	T2	27-03-2003
			DK	925113	T3	18-11-2002
			EP	0925113	A1	30-06-1999
			ES	2181028	T3	16-02-2003
			JP	2001508644	T	03-07-2001
			PL	332047	A1	16-08-1999
			WO	9810866	A1	19-03-1998
			US	6432694	B1	13-08-2002