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(54) Title: INSTALLATION FOR CARRYING OUT PHOTOCHEMICAL AND PHOTOCATALYTIC REACTIONS AND PHOTOINDUCED PROCESSES

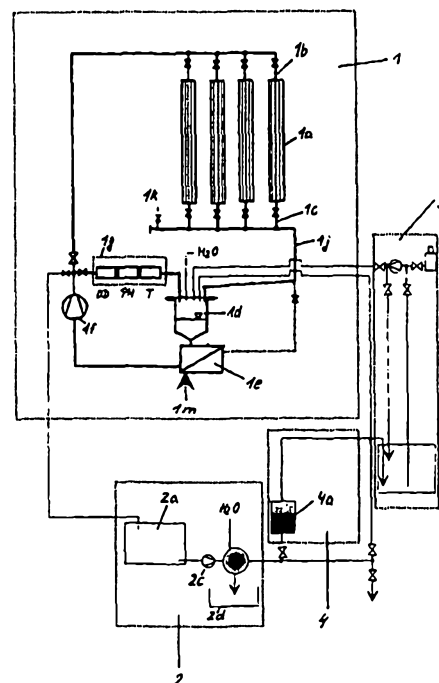
(54) Bezeichnung: ANLAGE ZUR DURCHFÜHRUNG VON PHOTOCHEMISCHEN UND PHOTOKATALYTISCHEN REAKTIONEN UND PHOTOINDUZIERBAREN PROZESSEN

(57) Abstract

The invention relates to an installation for carrying out photochemical and photocatalytic reactions and photoinduced processes, in particular for cultivating phototrophic organisms and cell cultures, e.g. microalgae. The invention seeks to improve known devices by the "thin-film principle" so that production of micro-organisms, in particular phototrophic micro-organisms, becomes so efficient as regards cultivating and harvesting processes that maximum production of micro-organisms is achieved all year round with optimal exploitation of natural sunlight and optimal conditioning of the culture suspension, in particular of the energy and material exchange processes as well as of the hydrodynamic flow conditions. At the same time, in co-operation with the peripheral equipment, optimal operating result is guaranteed.

(57) Zusammenfassung

Die Erfindung betrifft eine Anlage zur Durchführung von photochemischen und photokatalytischen Reaktionen und photoinduzierbaren Prozessen, insbesondere zur Kultivierung von phototrophen Organismen und Zellkulturen, z.B. Mikroalgen. Der vorliegenden Erfindung liegt die Aufgabe zugrunde, bekannte Einrichtungen nach dem "Dünnschichtprinzip" zur effizienten Produktion von Mikroorganismen, insbesondere von phototrophen Mikroorganismen hinsichtlich des Kultivations- und Ernteprozesses dahingehend zu verbessern, daß im gesamten Jahresverlauf unter optimaler Ausnutzung des natürlichen Sonnenlichts und optimaler Konditionierung der Kultursuspension, insbesondere der energetischen und stofflichen Austauschprozesse sowie der hydrodynamischen Strömungsverhältnisse, eine maximale Produktion von Mikroorganismen realisiert und gleichzeitig im Zusammenwirken der peripheren Ausrüstung ein optimales Betriebsergebnis gewährleistet wird.



**Apparatus to carry out photochemical and photocatalytic
reactions and photoinducible processes**

Description

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The invention concerns an apparatus to carry out photochemical and photocatalytic reactions and photoinducible processes, in particular for the cultivation of phototropic organisms and cell cultures, e.g. micro-algae.

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Known devices for the cultivation of phototropic organisms and cell cultures, like, for example, micro-algae, are constructed either as open ponds under the free sky or exclusively with artificial light, wherein the culture medium is conveyed in transparent tubes. In most cases devices having vessel-shaped reactors are also equipped with means for an intensive lighting for the conversion of the culture medium, the constructive effort thus increasing the associated costs.

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In the case of open ponds there is the constant danger of contamination, so that a pure cultivation is not possible. The management of temperature of such apparatus is also difficult under the prevailing climatic conditions.

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From the US published patent specification 4,473,970 a device operating with artificial light is known, in which the culture medium is conveyed in transparent tubes. The main disadvantage in the case of this device is the energy aspects with regard to the pumping and light capacities.

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Furthermore, devices are known, which consist of vessel-shaped reactors, into which the light is introduced by means of light guides. The light is introduced into the culture medium via a Fresnel lens which can follow the sun and/or via an artificial light source through the same light guide. Such plants are very cost intensive with regard to both manufacture and operation.

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In the US published patent specification 4,555,864 a device for cultivating chlorella algae is described, wherein the light guides are immersed directly into the culture medium in a cultivation vessel, while they are supplied with light energy via inclined positioned mirrors. This device is also very cost intensive and has an elaborate construction.

DE 41 34 813 A1 also describes a device for the cultivation of phototropic micro-organisms, which can be operated by using sunlight and/or artificial light and is flown through by a culture medium. This device is characterised by an arbitrary number of webs, which are arranged equidistant from each other between two parallel transparent plates in a fluid-tight manner and form chambers. At the end of each web connecting apertures are provided to the adjacent chamber, while the connecting apertures of each adjacent web are situated on the opposing ends, thus forming a channel which can be flown through in a meandering manner, the channel tightly enclosed on its faces and having a connecting socket at its beginning and end for the gas exchange. This device is unsuitable to be used on an industrial scale if only because of reasons of energy due to the expected high pump capacity necessary to overcome the channels' resistance to flow which can be flown through in a meandering manner and the high manufacturing costs of a plate having webs of this type.

Spectorova (1997) summarizes the status of the tubular bioreactors, which produce predominantly biologically active substances for the pharmaceutical and cosmetic industries. A horizontally arranged glass tube bioreactor having a capacity of 14 m³ culture has been installed in Uzbekistan. The glass tube bioreactor produces a biomass (*chlorella vulgaris*) of approx. 2 t/year with a maximum productivity of 43 g/m².d. At the same time a great problem is the distribution of the suspension in the individual glass tubes on the one hand, the tempering of the system and the very high concentration of oxygen in the suspension on the other. In addition, the assembly of the individual horizontal glass tubes is carried



out using flange connections, resulting in relatively high costs and gaps for the dead biomass or bacteria.

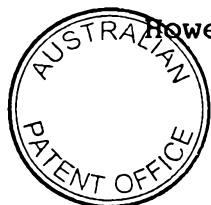
According to *Pulz* (1992 a, b) almost enclosed tubular glass tube fermenters (tube $d = 2.5$ cm) are used, whereby a centrifugal pump pumps 50 L of suspension of alga in a loop shape around the light source. If one bases the calculation on the jacket surface of the glass tube, then the surface-related growth rate of $14 \text{ g m}^{-2} \text{ d}^{-1}$ does not exceed the already mentioned productivities of the open systems.

Miyamoto et al. (1988) use the very cost-effective version, to cultivate micro-algae in vertical glass tubes with $d=5$ cm (surface/volume ratio 80 m^{-1}) and 2 m long, which are mass produced for fluorescent lights. By means of a plurality of parallel pistons an increase of capacity can be achieved without any effort, but the harvesting turned out to be difficult and there were considerable problems with heat when using natural lighting. Depending on the type cultivated, the maximum productivity extended from $0.3 \text{ g l}^{-1} \text{ d}^{-1}$ with the cyano bacterium *nostoc* to $0.6 \text{ g l}^{-1} \text{ d}^{-1}$ in the case of the green alga *monoraphidium*.

Jüttner (1977) has solved the problem of heat by installing a further internal tube, which held a cooling fluid on the one hand and reduced the non-illuminated areas of the suspension on the other. Due to this he achieved a further acceleration of the growth by a factor of 1.5. His idea, though technically elaborate and conditionally enlargeable, was used as a model for the construction of some Freiland tube apparatus with counter-current cooling (cf. published French patent specification 2 564 854).

Characterising for all cylindrical reactors is the high density of light relative to a small surface/volume ratio, so that high flow velocities are necessary to achieve an adequate light exposure of the alga cell or to prevent photoinhibition.

However, the strong turbulences produce corresponding loads by



gravitational forces, which have a counter-productive effect in the case of most types of micro-algae. In addition, an effective illumination of larger apparatus would be difficult to carry out or only with considerable technical effort.

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The object of this invention is to improve known devices using the "thin film principle" for the efficient production of micro-organisms, in particular of phototropic micro-organisms, with regard to the cultivation and harvesting process to that effect that a maximum production of micro-organisms is achieved during the entire year with the optimum use of the natural sunlight and optimum conditioning of the culture suspension, in particular of the energy and material exchange processes as well as of the hydrodynamic flow conditions, while simultaneously assuring an optimum operating result in conjunction with the peripheral equipment.

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According to the invention this objective is achieved by the features of claim 1. Advantageous developments of the invention are contained in claims 2 to 14.

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The device in accordance with the invention for the efficient production of micro-organisms comprises a bioreactor, a harvesting device, a nutrition solution and a parent solution feeding device and a nutrition solution preparing apparatus.

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The bioreactor comprises photosynthetically and hydrodynamically (turbular) active thin film systems, a forward and return flow system, a conditioning vessel, a system pump, a measuring section, a means for the quantitative and qualitative modification of the illumination of the vessel and a means for the gas supply and discharge. A photosynthetically and hydrodynamically active thin film system comprises a plurality of continuous tubes or hoses which are superposed and parallel to each other, whose inlet and outlet sides are connected with a forward module and a return, respectively, module by means of connecting pieces and a connector with reduced cross-section, wherein the individual superposed thin film vessels have

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connectors with reduced cross-section with a cross-section enabling an even flow pattern through all thin film vessels. According to a particular feature of the invention the connector with reduced cross-section has an increasing cross-section in the direction of flow of the forward module and/or a decreasing cross-section in the direction of flow of the return module.

According to a further feature of the invention the forward module and the return module consists of a tube. At the same time it is of advantage if in the direction of flow the forward module has a smaller diameter and the return module a larger diameter.

According to a particular feature of the invention the transparent thin film vessel has conditioning turbulators, which assure turbulent flow conditions even at a low flow velocity which increase the surface/base ratio, distribute the incident light in an optimum manner in the transparent thin film vessel and make possible the possibly occurring immobilisations on the surface of the vessel by means of an efficient cleaning effect by cleaning bodies in the suspension, while the conditioning turbulators are designed in such a manner that no hydraulically dead spaces will occur in the transparent thin film vessel.

According to a further feature of the invention the transparent thin film vessel comprises at least one glass tube module with superposed glass tubes, which contain the conditioning turbulators already during the manufacture of the glass tube.

According to a preferred embodiment of the invention the total length of the glass tube is made up from the individual tubes, which are bonded or welded together without any hydraulic interruption.

In an advantageous embodiment of the invention the means for the quantitative and qualitative modification of the



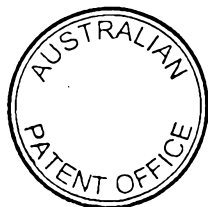
illumination of the vessel consist of UV-fluorescing and/or reflecting and/or anti-reflecting and/or transparent material, which increases the amount of radiation available in the photosynthetically active region for the illumination of the micro-organisms in the thin film vessels. At the same time it is an advantage if the surface and/or the material of the wall of the transparent thin film vessel is made of UV-fluorescing and/or anti-reflecting material, which increases the amount of radiation available in the photosynthetically active region for the illumination of the micro-organisms in the thin film vessels and/or between the thin film vessels UV-fluorescing and transparent material and/or below the thin film vessels UV-fluorescing material is provided.

According to a further feature of the invention the means for the quantitative and qualitative modification of the illumination of the vessel are from IR-reflecting and/or transparent material and are integrated in the surface or the material of the wall of the transparent thin film vessel, whereby an increase of the temperature of the suspension is reduced and a radiation cooling of the culture suspension is assured through the material of the wall of the transparent thin film vessel.

It is within the scope of the invention that the means for the gas supply and discharge comprise a return flow collector, which is provided in or on the bottom and is tempered, a CO₂ retainer in the culture suspension, an O₂ discharge from the culture suspension and a gas distributor in the return flow collector.

At the same time it is of advantage to use latent heat storage material for the means for the gas supply and discharge.

A particularly preferred embodiment of the invention provides that the conditioning vessel, the heat exchanger and the means for the gas supply and discharge are connected with each other and operate as a complex.



At the same time it is of advantage that the means for the gas supply and discharge, in particular the return flow collector, form simultaneously a part of the heat exchanger and/or of the conditioning vessel and thus a synergy effect is achieved as far as optimum conditioning of the culture suspension is concerned.

Furthermore, the invention is equipped with a harvesting device comprising a harvest container, a harvest pump, a separator and a biomass container.

In addition, the invention is equipped with a nutrition solution and parent solution feeding device, comprising a nutrition solution pump, a parent solution container with an agitator and a nutrition solution container.

According to a particular feature of the invention the harvesting device and/or the nutrition solution and the parent solution feeding device has a nutrition solution preparing device, by means of which a separated culture suspension from the harvesting device as nutrition solution and/or the nutrition and parent solution from the nutrition solution and parent solution feeding device can reach the bioreactor free of contamination.

It is also within the scope of the invention that the nutrition solution preparing device comprises a mechanical pre-filter and an activated carbon filter, which filters the contaminated materials and allows an unhindered passage of the nutrition materials for the micro-algae.

A particularly preferred embodiment of the invention provides that the harvesting device, the nutrition solution and parent solution feeding device and the nutrition solution preparing device are connected with each other and operate as a complex.

In the following the invention is explained in detail based on embodiments. The drawings show in:

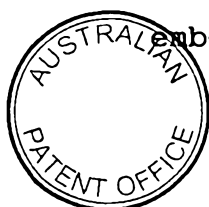


Fig.1 - the schematic illustration of a version of the apparatus according to the invention,

Fig.2 - a glass tube module with forward and return flow as well as conditioning turbulators,

Fig.3 - a glass tube module (in side view) with return collector and means for the quantitative and qualitative modification of the illumination of the vessel.

Fig. 1 shows a schematic illustration of a version of the apparatus according to the invention for the efficient production of phototropic organisms and cell cultures, in particular of micro-algae, under the influence of light.

The apparatus is made up from a bioreactor 1, the harvesting device 2, a nutrition solution and parent solution feeding device 3 and a nutrition solution preparing device 4.

First of all in Fig.1 by inoculating a nutrition solution with micro-organisms a culture suspension and parent solution is produced. The device 3 comprises essentially a nutrition solution container 3a, a nutrition solution pump 3b and a parent solution container 3c with an agitator .

The nutrition solution 1a is conveyed from here to the bioreactor 1. In this example the bioreactor 1 comprises essentially four modules of a photosynthetically and hydrodynamically active thin film system 1a, which modules form the reactor vessel, a forward and return flow system 1b and 1c, respectively, a conditioning vessel 1d, a heat exchanger 1e, a system pump 1f, a measuring section 1g, means 1h and 1i (see Figs.2 and 3) for the quantitative and qualitative modification of the illumination of the reactor vessel and means to supply and discharge gas. In the example described the means to supply and discharge the gas comprise a return flow collector 1j,



which in this case is provided on the bottom of the apparatus, a device (not illustrated) to retain the CO₂ in the culture suspension in the return flow collector 1j, an O₂ discharge 1k and a gas distributor 1l (Fig.3).

5 In the return flow collector 1j a gas distributor 1l is provided, wherein the gas distributor 1l apportions over the entire length of the return flow collector 1j a waste gas having a CO₂ concentration of 25 % into the culture suspension.
10 When the temperature is low, the waste gas has a maximum temperature of 38°C and heats the culture suspension 1e.

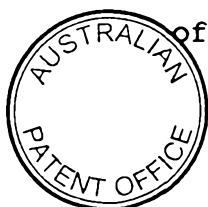
At the same time a heat exchanger 1e assures an optimum temperature waste gas and suspension temperature.

15 For the purpose of increasing the heat transfer the heat exchanger 1e is mounted in the suction line of the system pump 1f. The conditioning of the CO₂-containing waste gas takes place as a function of the pH, temperature and optical density
20 values of the culture suspension, which are determined in the measuring section 1g before the system pump 1f. At the same time the measuring, control, regulation and storage of the variable values of the culture suspension and of the CO₂-containing waste gas is carried out by means of a central
25 measuring, control, regulating and storage unit (not illustrated). The conditioned culture suspension is conveyed to the photosynthetically and hydrodynamically active thin film system 1a of the bioreactor 1 with the aid of a pump movement 1n. The thin film system 1a in this example comprises four
30 modules, while each module comprises a plurality of parallel thin film vessels (glass tubes) 1o superposed and at a distance from each other, whose inlet and outlet sides are joined by means of connecting pieces 1p and connectors 1q with a forward module 1r and a return module 1s, respectively. The individual
35 connectors 1q have a cross-section which enables an even flow pattern through all glass tubes 1o. At the same time the cross-sections of the connectors 1q increase in the direction of flow of the forward module 1r, whereas the cross-sections of the



connectors 1q of the return module 1s decrease in the direction of the flow. The forward module and return module 1r and 1s respectively, comprise a tube with connecting pieces 1t for the connector 1q, wherein the cross-section of the forward module 1r decreases in the direction of the flow and the cross-section of the return module 1s increases in the direction of the flow. An even flow pattern is achieved through all thin film vessels 1o by the connectors 1q with the reduced and increased, respectively, cross-sections and the forward and return modules having reduced and increased, respectively, cross-sections. In addition, the thin film vessels 1o are equipped with a plurality of conditioning turbulators 1u, which assure turbulent flow conditions even at a low flow velocity and increase the surface/base ratio as well as distribute the incident light in the thin film vessel in an optimum manner and prevent and/or reduce the immobilisations of the cleaning bodies in the suspension occurring on the surface of the vessel.

In this present case the thin film system 1a is constructed from four glass tube modules, each of which comprises a stand 1v with bilateral mountings offset relative each other for the tubes, in which mountings the tubes 1o are arranged superposed and offset relative each other. The surface and/or the material of the wall of the glass tubes 1o and/or the stand 1v or the mountings of the tubes (not illustrated) and/or the base on which the thin film system 1a is arranged, are provided with means, in particular from UV-fluorescing and/or transparent and/or reflecting material 1h and 1i for the quantitative and qualitative modification of the illumination of the reactor vessel. By virtue of this the amount of radiation available in the photosynthetically active region is increased for the illumination of the micro-organisms in the thin film vessels. However, the means may be also from IR-reflecting and/or transparent material, which prevents an increase of the temperature of the suspension and a radiation cooling of the culture suspension is assured through the material of the wall of the transparent thin film vessel. In the process of growth



of the biomass in the thin film system 1a energy-rich carbon compounds are formed. The harvesting of the biomass is carried out with a dry-mass concentration in the culture suspension of > 2 g/L.

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For this purpose the culture suspension is conveyed to the harvesting device 2 with the aid of the system pump 1f. The harvesting device consists of a harvest container 2a, a harvest pump 2b, a separator 2c and a biomass container 2d. The separator 2c separates the biomass from the nutrition solution, which is returned to the nutrition solution preparing device 4 and/or directly to the nutrition solution and parent solution feeding device 3.

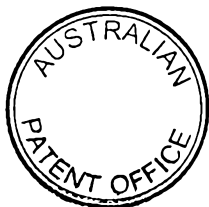
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The device 4 comprises a mechanical pre-filter (not illustrated) and an activated carbon filter 4a, which filters the contaminated materials and allows an unhindered passage of the nutrition materials for the micro-algae.

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The biomass obtained in this manner is conveyed to a preparing device (not illustrated), wherein energy-rich carbon compounds are extracted as energy carriers for the production of chemical and/or pharmaceutical basic and active materials and/or for the production of high-degree fodder or fodder additives.



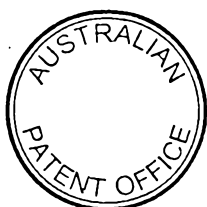
Patent claims

1. An apparatus for the production of phototropic organisms and cell cultures under the influence of light by photochemical and photocatalytic reactions and photoinducible processes, comprising a bioreactor (1), a harvesting device (2), a nutrition solution and parent solution preparing device (3) and a nutrition solution preparing device (4), wherein the bioreactor (1) has a photosynthetically and hydrodynamically active thin film system (1a), a forward and return flow system (1b, 1c), a conditioning vessel (1d), a heat exchanger (1e), a system pump (1f), a measuring section (1g), means (1h, 1i) for the quantitative and qualitative modification of the illumination of the reactor vessel and means to supply and discharge gas.

2. An apparatus according to claim 1, characterised in that the photosynthetically and hydrodynamically active thin film system (1a) comprises at least one module with a plurality of parallel tubular thin film vessels (1a) which are superposed and are at a distance from each other, whose inlet and outlet sides are joined by means of connecting pieces (1p, 1t) and connectors (1q) with a forward module (1r) and a return module (1s), respectively, while each individual connector (1q) for the superposed thin film vessels (1a) has a cross-section which enables an even flow pattern through all thin film vessels.

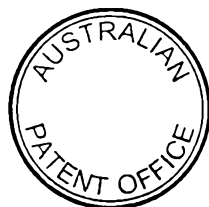
3. An apparatus according to claim 2, characterised in that the cross-section of the connector (1q) increases in the direction of flow of the forward module (1r) and decreases in the direction of flow of the return module (1s).

4. An apparatus according to any one of the preceding claims, characterised in that the forward and return module (1r, 1s) comprise a tube with connecting pieces (1p, 1t) for the connectors (1q), while the cross-section of the forward



module (1r) decreases in the direction of the flow and the cross-section of the return module (1s) increases in the direction of flow.

- 5 5. An apparatus according to any one of the preceding claims, characterised in that the thin film vessels (1o) have a plurality of conditioning turbulators (1u).
- 10 6. An apparatus according to any one of the preceding claims, characterised in that the thin film systems (1a) comprise at least one glass tube module, which comprises a stand (1v) with bilateral tube mountings provided offset relative each other in which the tubes (1o) are arranged superposed and offset relative each other.
- 15 7. An apparatus according to any one of the preceding claims, characterised in that the surface and/or the material of the wall of the thin film vessels and/or the stand (1v) or the mountings of the tubes of the glass modules and/or the base on which the glass modules are arranged and/or between the tubes means (1h, 1i) are provided for the quantitative and qualitative modification of the illumination of the reactor vessel.
- 20 8. An apparatus according to any one of the preceding claims, characterised in that the means (1h, 1i) for the quantitative and qualitative modification of the illumination of the reactor vessel consist of UV-fluorescing and/or reflecting and/or anti-reflecting and/or transparent material.
- 25 9. An apparatus according to any one of the preceding claims, characterised in that the means (1h, 1i) for the quantitative and qualitative modification of the illumination of the reactor vessel consist of IR-reflecting and/or transparent material.
- 30 35



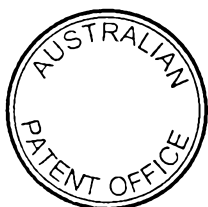
10. An apparatus according to any one of the preceding claims, characterised in that the means to supply and discharge the gas comprise a return flow collector (1j), a device to retain the CO₂ in the culture suspension, a device (1k) to discharge the O₂ from the culture suspension and a gas distributor (1l) in the return flow collector (1j).

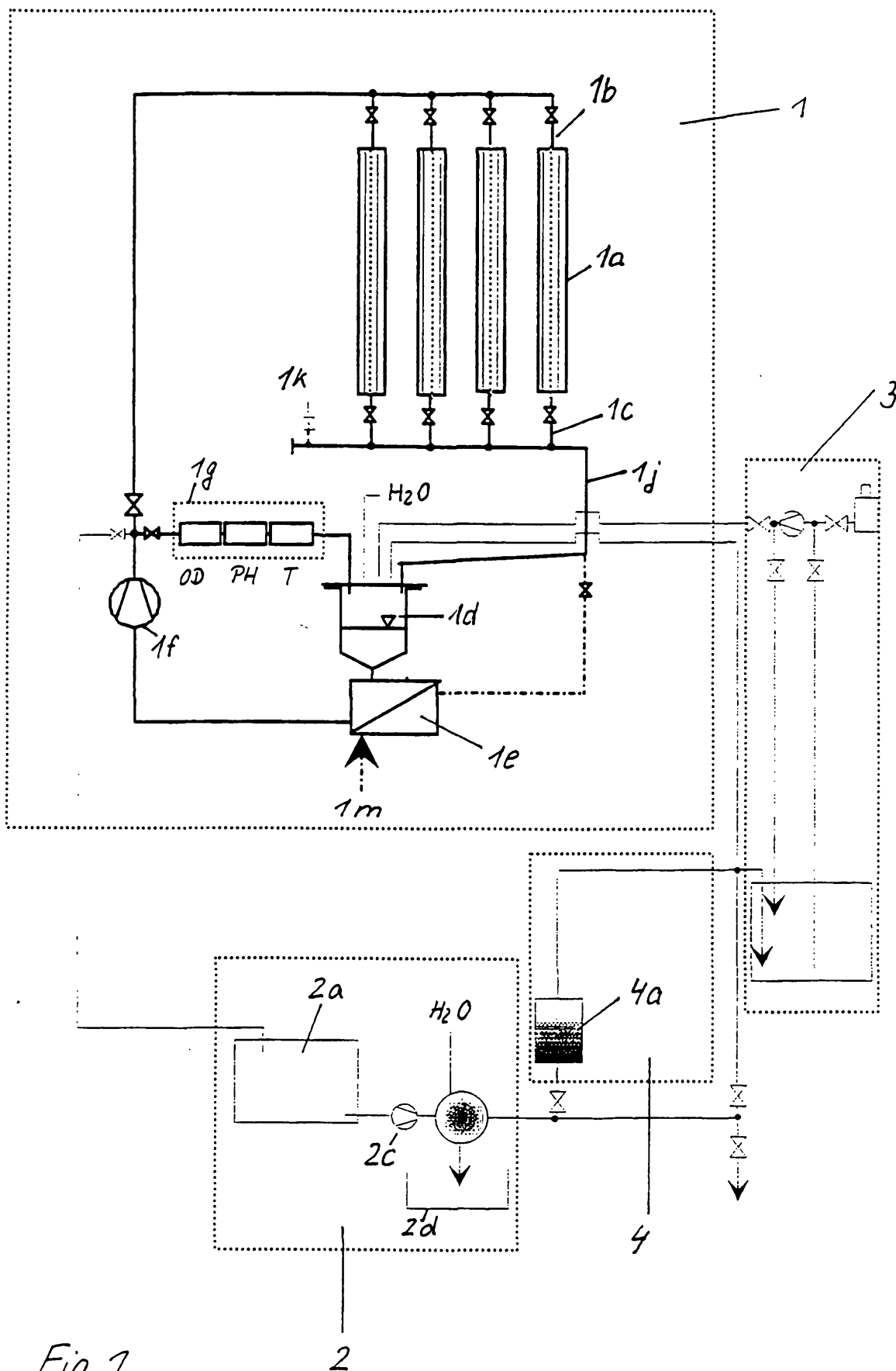
11. An apparatus according to any one of the preceding claims, characterised in that the conditioning vessel (1d), the heat exchanger (1e) and the means for the supply and discharge of the gas are connected with each other, the return flow collector (1j) simultaneously forming a part of the heat exchanger (1e) and/or of the conditioning vessel (1d).

12. An apparatus according to any one of the preceding claims, characterised in that the harvesting device (2) consists of a harvest container (2a), a harvest pump (2b), a separator (2c) and a biomass container (2d).

13. An apparatus according to any one of the preceding claims, characterised in that the nutrition solution and parent solution feeding device (3) comprises a nutrition solution pump (3a), a parent solution container (3b) with an agitator and a nutrition solution container (3c).

14. An apparatus according to any one of the preceding claims, characterised in that the nutrition solution preparing device (4) comprises a mechanical pre-filter and an activated carbon filter (4a) and is connected with the harvesting device (2) and/or the nutrition solution and parent solution feeding device (3).





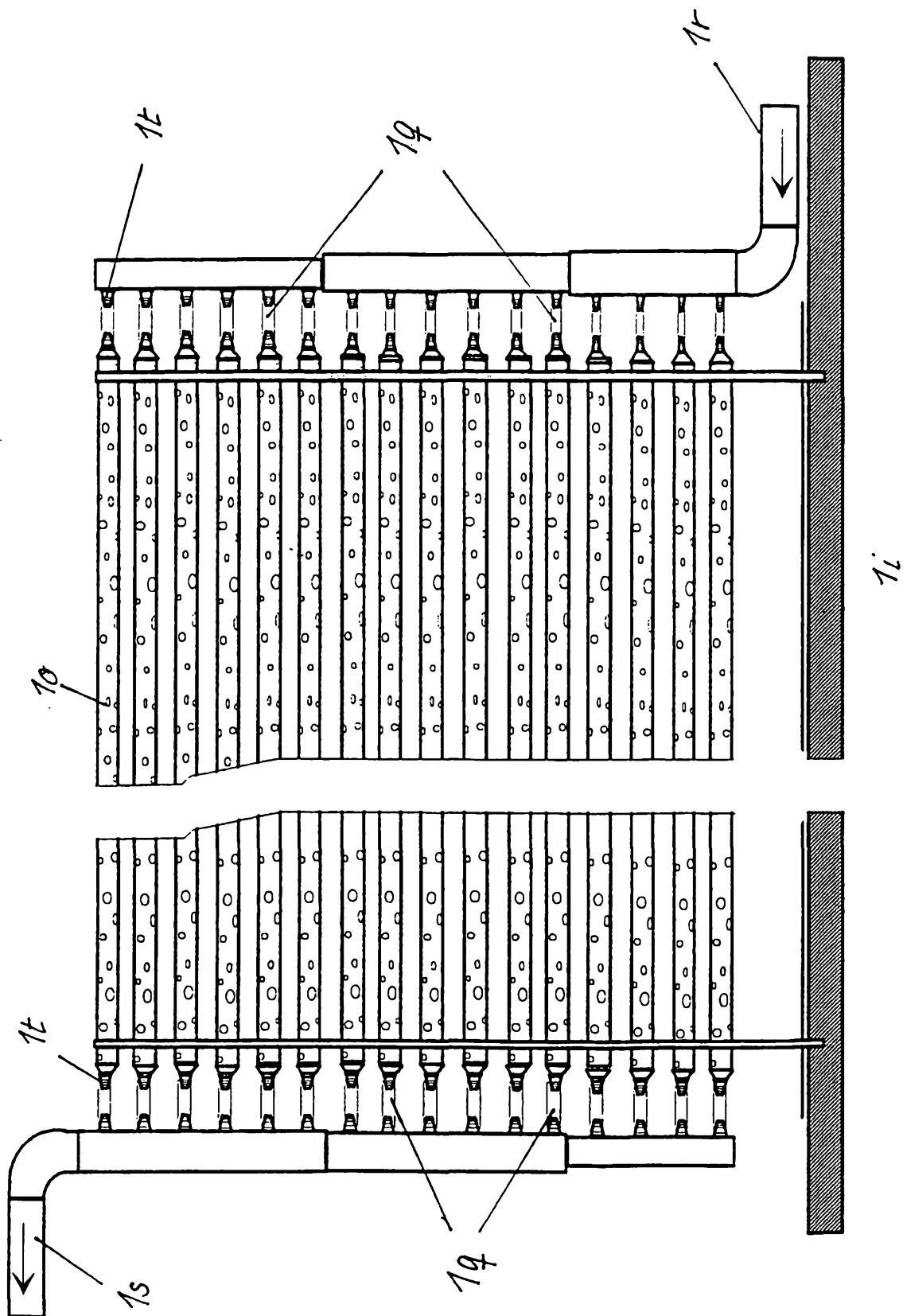


Fig. 2

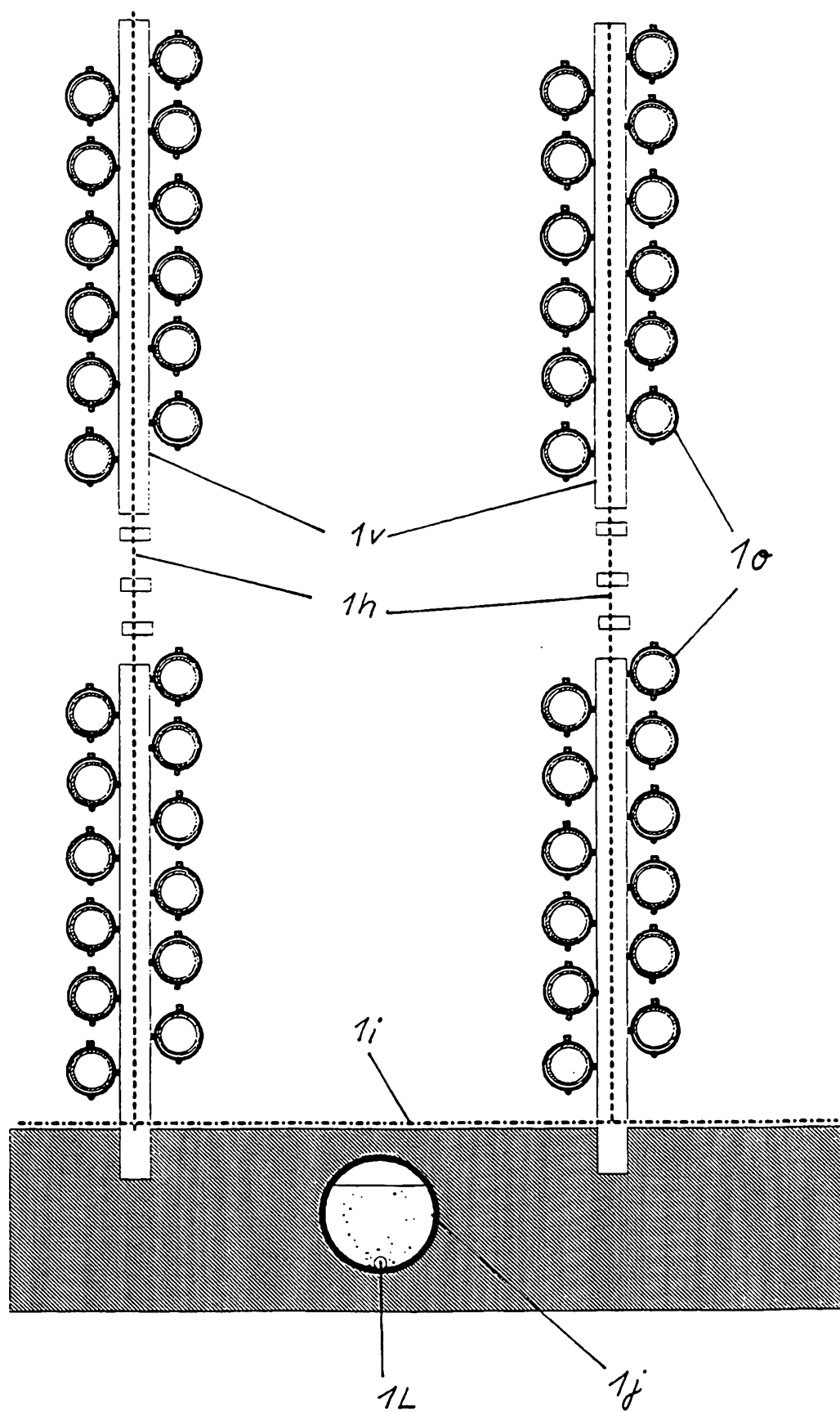


Fig. 3