

19



Octrooicentrum
Nederland

11

2034234

12 B1 OCTROOI

21

Aanvraagnummer: 2034234

51

Int. Cl.:
C01B 32/55 (2023.01) F25D 3/12 (2023.01)

22

Aanvraag ingediend: 28 februari 2023

62

30

Voorrang:

-

41

Aanvraag ingeschreven:
9 september 2024

43

Aanvraag gepubliceerd:
-

47

Octrooi verleend:
9 september 2024

45

Octrooischrift uitgegeven:
19 september 2024

73

Octrooihouder(s):
Universiteit Twente te Enschede

72

Uitvinder(s):
Srinivas Vanapalli te Enschede
Abhishek Sharad Purandare te Enschede

74

Gemachtigde:
dr. A. Ellens c.s. te Wageningen

54

Dry ice

57

The invention provides a system (100), wherein the system (100) comprises a device (10) comprising (i) a device wall (15) with a device opening (19), a (ii) flow channel element (20) comprising an element inlet (21) and an element outlet (29), wherein the flow channel element (20) is at least partly heat conductive, (iii) a confining wall (35) for defining a confined space (30), and (iv) a channel space (40), wherein the confining wall (35) comprises a plurality of openings (31) for allowing a gaseous fluid to leave the confined space (30), the device wall (15) and the confining wall (35) define the channel space (40), wherein the channel space (40) fluidically connects the confined space (30) to the device opening (19) via the plurality of openings (31) and wherein the flow channel element (20) is configured thermally coupled to the channel space (40), wherein the element inlet (21) is configured at an outer side (11) of the device (10) and the element outlet (29) is configured in open fluid connection with the confined space (30), and the element outlet (29) comprises a throttle element (22).

Dry ice

FIELD OF THE INVENTION

5 The invention relates to a system and a method for producing dry ice and cooling with dry ice.

BACKGROUND OF THE INVENTION

Methods and devices to produce dry ice are known in the art. WO2021078891,
10 for instance, describes a device for producing dry ice pellets comprising a pressing cylinder, into which an inlet opening for feeding liquid carbon dioxide leads and which is equipped, at an end, with a die, which has one or more openings, and with a piston, which is longitudinally movably accommodated in the pressing cylinder. During use, liquid carbon dioxide is fed to the pressing cylinder. Said carbon dioxide is expanded at the inlet opening, is at least partially
15 converted into carbon dioxide snow and is pressed against and through the die by the movement of the piston. According to the description, the die is in the form of a cylindrical body that can be connected to the pressing cylinder and is equipped with feed-throughs, which are oriented parallel to a longitudinal axis of the die and are dimensioned in such a way that the length and the diameters of the feed-throughs correspond to the length and the diameters of the dry ice
20 pellets to be produced.

SUMMARY OF THE INVENTION

Various applications require working with/at cryogenic temperatures. Examples are, e.g., found in cryopreservation and cold chain transportation. At cryogenic temperatures,
25 such as below 200 K, atoms are slowed down and therefore conservation of goods is possible for a longer duration. A common method to cool down to cryogenic temperatures is done by using cryogenic liquids or dry ice (carbon dioxide in the solid state). Cryogenic liquids are normally at their saturation temperature, i.e., boiling temperature. Any heat transferred to the liquid will cause evaporation, thereby reducing the amount of liquid but keeping it at a constant
30 temperature. Likewise, heat transferred to dry ice may induce sublimation of the dry ice, wherein the (solid) dry ice is kept at a constant temperature.

One of the challenges in the healthcare is the introduction of molecular diagnostic tests, which can improve the diagnosis of the disease and the choice of therapy in (cancer) patients. For this, it is important to prepare and transport frozen biomedical samples

(e.g. tissues and other human materials) in a controlled and safe manner, so that the desired diagnostic tests can be performed in specialized laboratories. Due to the increase in the number of molecular diagnostic tests, the demand for a reliable cold chain for preparation and transportation is increasing rapidly. The cornerstone for sample preparation and logistics in the cryopreservation field is to use dry ice (solid carbon dioxide with a sublimation temperature of -78 °C). Unfortunately, dry ice cannot be directly used in freezing samples as the cooling rate when a sample is gently placed on dry ice is low. Therefore, clinical staff use dangerous substances such as isopentane that is pre-cooled by dry ice to freeze the sample. Experimental work shows that the cooling speed of vials in an isopentane bath cooled by dry ice is one order of magnitude faster than by directly placing the vials in contact with the (densified) dry ice. After freezing, the samples are transported in insulation boxes containing densified dry ice.

In presently applied systems to produce dry-ice, high pressure CO₂ liquid at ambient temperature is normally expanded through an orifice. In this process, CO₂ the liquid state at high pressure and room temperature is converted to a combination of CO₂ vapor and solid CO₂, wherein the total enthalpy is maintained. Theoretically, at ambient pressure, around 30 weight% of the mixture is in the solid state, whereas the remaining about 70 weight% is cold vapor. Hence, 1 kg of liquid CO₂ may produce 0.3 kg of solid carbon dioxide (dry ice). In practice, the dry ice snow that is formed during the expansion process is captured in a mold and simultaneously compressed to form densified dry ice. Vast amount of CO₂ gas formed during the expansion process is exhausted into the atmosphere. This cold gas that exists the system contains about 87 kJ/kg of cold energy (between cold temperature and ambient). For a bottle of 20 kg, approximately 1200 kJ of energy is being released into the atmosphere (thus wasted). The conversion efficiency of the devices present in the market is approximately 20 %, which basically means that using these devices one can obtain approximately 4-5 kg of dry ice from 20 kg liquid CO₂ bottle. Next to the inefficient conversion of liquid CO₂ to dry ice, present systems and devices for cooling with dry ice may come with at least the several further relevant disadvantages given below:

The rigid way of sample preparation: The tight link between the sample freezing and the availability of dry ice means the sample preparation (of e.g. cells, tissues etc.) must be carefully planned. Ordering large boxes of dry ice as a buffer is a possible solution. However, this would mean obtaining and dedicating space for a storage of large quantities of dry ice. Moreover, losses are substantial and is a waste of cold resource. Added to this the lost euros.

Losses during transportation: Dry ice must be transported from the point of production to the end user, this leads to losses of dry ice as it continuously sublimates when it is simply waiting to be used.

Safety concerns with isopentane: It must be noted that isopentane is a dangerous substance (carcinogenic) and special training must be followed to use this substance. Preferably an alternative solution is found to freeze samples at the same speed as with pre-cooled isopentane.

Hence, it is an aspect of the invention to provide an alternative system, especially for producing solid CO₂ (dry ice), which preferably further at least partly obviates one or more of above-described drawbacks. It is a further aspect of the invention to provide a method for cooling, which preferably further at least partly obviates one or more of above-described drawbacks. In yet a further aspect, the invention provides a method for producing dry ice.

The present invention may have as object to overcome or ameliorate at least one of the disadvantages of the prior art, or to provide a useful alternative.

Hence, in a first aspect, the invention provides a system. The system is especially for producing solid CO₂ (dry ice). The system comprises a device, especially a device for producing solid CO₂ (dry ice). The device comprises a device wall. The device wall especially comprises a device opening. In further embodiments, the device (further) comprises a flow channel element, especially comprising an element inlet and an element outlet. The flow channel element may in further embodiments be at least partly heat conductive. The device further especially comprises (or defines) a confined space. The device may especially comprise a confining wall for defining a confined space. The confined space is especially defined during use (and may be open when the device is not used). Further, especially, the confined space comprises a confining wall. Further, in embodiments, the confining wall, comprises an opening, especially for allowing a gaseous fluid to leave the confined space. In specific embodiments, the confining wall comprises a plurality of openings (especially for allowing a gaseous fluid to leave the confined space. The device may in further embodiments comprise a channel space. In further specific embodiments, the device wall and the confining wall define the channel space. The device wall may especially enclose the channel space. In further embodiments, the channel space encloses the confining wall. In further specific embodiments, the channel space fluidically connects the confined space to the device opening, especially via the (plurality of) opening(s). In further embodiments, the flow channel element is configured thermally coupled to the channel space. In specific embodiments, at least part of the flow channel element is arranged in the channel space. Further, especially, the flow channel element may be configured

through the device wall. The element inlet may in embodiments be arranged at an outer side of the device, and especially the element outlet may be configured in open fluid connection with the confined space. The flow channel element may in embodiments define a flow path (especially a channel or duct) from a location external of the device to a location in the confined space. In further embodiments, the element outlet comprises a throttle element.

The invention further provides a method (for cooling). In embodiments, the method comprises a snow forming stage. In specific embodiments, the snow forming stage comprises: flowing (a flow of) liquid CO₂ through a flow channel element from an element inlet to an element outlet (of the flow channel element), especially while cooling the flow channel element. The (liquid) CO₂ may especially exit the flow channel element at the element outlet. The snow forming stage may further comprise throttling the flow of the liquid CO₂ at the element outlet. By throttling the flow of liquid CO₂, the liquid CO₂ may form a mixture comprising a particulate solid CO₂ (or “dry ice snow”) and a gaseous CO₂ (“CO₂ vapor”). In specific embodiments, the liquid CO₂ may form a mixture of particulate solid CO₂ and gaseous CO₂ (after throttling). The mixture especially sprays from the element outlet, especially into a confined space. The element outlet is especially configured in fluid connection with the confined space. The method may in specific embodiments further comprise a cooling stage. In embodiments, the cooling stage may comprise spraying the particulate solid CO₂ over a sample arranged in the confined space. In further specific embodiments the cooling stage comprises densifying the particulate solid CO₂ in the confined space and cooling a sample with the densified particulate solid CO₂. In specific embodiments, the cooling stage may comprise densifying the particulate solid CO₂ in the confined space (and cooling a sample with the densified particulate solid CO₂). The cooling stage may in embodiments comprise (directly) cooling the sample with dry ice snow (in the confined embodiment). Alternatively, the dry ice snow may be densified to form densified particulate solid CO₂ (“densified dry ice” or “dry ice pellets”) and especially the sample is cooled with the densified dry ice (after removing the densified dry ice from the confined space).

The device may allow cooling the sample by producing a spray of dry ice snow and spraying the dry ice snow over a sample arranged in a confined space. This way, cooling rates may be provided that are acceptable to the cryopreservation community while not requiring the use of isopentane or other alcohols anymore. Further, using the device and the method may result in a production of 70 % more densified dry ice per unit of a liquid CO₂ compared to the known devices and methods. This allows substantial saving in the operating costs and is good for the environment. Moreover, compared to the prior art, the system is safe

to use as cold surfaces that an operator may be exposed with may be eliminated. The invention may provide flexibility in sample preparation; with the system at the doorstep of a user, small quantities of dry ice can be produced as per the demand of the user, and at the time when it is actually needed. This allows the user more flexibility in preparation and packaging of temperature sensitive samples. Further, the invention may eliminate dependency on the dry ice supplier which reduces transportation of its need, the dry ice is not waiting for longer period. This eliminates losses of dry ice due to its sublimation during its storage.

The invention may come without safety concerns; with the system and the method samples may be frozen at high cooling rates using dry ice snow spray, eliminating the need to use isopentane, and thereby eradicate the risks associated with it. Moreover, in the state-of-the-art processes, CO₂ gas is released directly into the room and might cause asphyxiation hazard. In the present system/ device the CO₂ gas may be channeled through the system/device and may be vented through a hose outside the room.

A main aspect of the invention is the production of a greater amount of dry ice from a given amount of liquid CO₂ by precooling the liquid CO₂. In advantageous embodiments, the liquid CO₂ is precooled by recuperating the cold energy of the exhaust vapor. The incoming liquid CO₂ may be precooled by the cold vapor leaving the confined space (especially the confined space). As a result of the precooling, the isenthalpic expansion of liquid CO₂ takes place at a lower enthalpy (J/kg) of the CO₂ compared to present methods. This change in enthalpy results in higher fraction of solid CO₂. This way, in embodiments an increase of 70 % of the conversion efficiency from liquid CO₂ to dry ice may be obtained.

The system, especially the device, may be used for the method(s) described herein. Moreover, the method may be applied using the system (device) described herein. The method and the system may especially advantageously be used for cooling a temperature sensitive sample. The sample may e.g. comprise a biomedical sample, such as a tissue or other human material, especially for a diagnostic test. The sample may further comprise a biological product, such as a vaccine. In further embodiments, the sample may comprise a medical product, such as a medical drug, an auxiliary agent for a medical product, Hence, in embodiments, the sample comprise one or more of a biomedical sample a biological product, and a medical product.

Hence, in embodiments, the invention provides a system (for producing solid CO₂ “dry ice”), wherein the system comprises a device comprising (i) a device wall with a device opening, a (ii) flow channel element comprising an element inlet and an element outlet, wherein the flow channel element is at least partly heat conductive, (iii) a confining wall for

defining a confined space, and (iv) a channel space, wherein (1b) the confined space wall comprises an opening, especially a plurality of openings (for allowing a gaseous fluid to leave (an interior of) the confined space, (iib) the device wall and the confined space wall define the channel space, wherein the channel space fluidically connects (the interior of) the confined space to the device opening via the plurality of openings and wherein the flow channel element is configured thermally coupled to the channel space; (iiib) the flow channel element is configured through the device wall, wherein the element inlet is arranged at an outer side of the device and the element outlet is configured is in open fluid connection with the confined space, and (ivb) the element outlet comprises a throttle element. Especially the device is configured for producing dry ice.

In a further aspect, the invention provides in embodiments, a method (for cooling), wherein the method comprises a snow forming stage and a cooling stage, wherein the snow forming stage comprises: (i) flowing liquid CO₂ (1b) through a flow channel element from an element inlet to an element outlet (both of the flow channel element) while cooling the flow channel element, and (ii) throttling the flow of the liquid CO₂ at the element outlet, wherein (or “such that”) the liquid CO₂ (1b) forms a mixture comprising (a) particulate solid CO₂ (1a) and a gaseous CO₂, the mixture spraying from the element outlet into a confined space; and wherein cooling stage comprises one of (i) spraying the particulate solid CO₂ (1a) over a sample arranged in the confined space; and (ii) densifying the particulate solid CO₂ (1a) in the confined space and cooling a sample with the densified particulate solid CO₂ (1a).

Herein the term “confined space” is used especially referring to a space (or environment) restricted in volume (or area). The sample may in embodiments be placed in this space when being sprayed by the dry ice snow. The space may be restricted by restricting walls or other sealing elements (completely) enclosing the volume, herein indicated as a “confining wall”. In further embodiments the walls may not completely enclose the volume but especially may cover the volume. For instance, the walls may define a cover arranged over the sample. In specific embodiments the confined space may be a (closed) confined space or a closable confined space. The invention may especially be explained based on a chamber as an embodiment of the confined space. It will be understood that the term “chamber” in embodiments may be replaced by “confined space” and vice versa. Likewise the term “confining wall” may be replaced chamber wall”. Moreover the confining wall or cover may be arranged at a surface (not necessarily being part of the system) and as such may define the confined space or chamber. The terms “chamber” and “confined space” may thus especially refer to the device during use of the device. The device may especially comprise a “chamber

element” or a “confined space element” (i.e. the confining wall configured to define a (closed) confined space when being arranged at a surface, a plane, or a further cover (closing the confined space. In further specific embodiments, the confined space may comprise a (detachable) closing element closing the confined space (see further below). The term

5 “confined space” may in embodiments refer to an open confined space or more especially the confining wall. It is further noted that the confined space may not be hermetically closed; a fluid may in embodiments leave the confined space via one or more openings in the confining wall.

Herein the terms “liquid carbon dioxide” and “gaseous carbon dioxide”/“carbon

10 dioxide vapor” and comparable terms are used. Liquid carbon dioxide (CO_2) is the liquid state of carbon dioxide, The liquid state of carbon dioxide only exists at pressures in the range of 5.2 bar to 73.773 bar (i.e. the pressure at the triple point and pressure at the critical point, respectively) and in the temperature range of 216.55 K to 304.128 K. (the temperature of the triple point and the temperature of the critical point, respectively). Gaseous carbon dioxide and

15 carbon dioxide vapor are the gaseous state of carbon dioxide.

Herein, terms like “subcooled”, “saturated liquid”, etc. are used. The terms “subcooled” (also called “undercooled”), “subcooling”, “undercooling”, and the like, refer to a liquid at a temperature below its normal boiling point (at a given pressure). For instance water at ambient pressure boils at 373 K. Therefore, water at room temperature is indicated as

20 “subcooled”. Likewise, liquid carbon dioxide at a pressure of 5.2 bar (5.1 atm) has a boiling point of about 217 K (216.55 K) and is called subcooled at temperatures below 217 K (at 5.2 bar). Moreover, herein, the terms “subcool(ing)” and “sub-cool(ing)” may both be used. Likewise “sub-cooled” and “subcooled” may be used interchangeably. Further, the term “saturated”, such as in “saturated liquid” relates to a liquid at its boiling point. Liquid carbon

25 dioxide at about 217 K (216.55 K) and 5.2 bar is a saturated liquid.

It is noted that at atmospheric pressure gaseous CO_2 directly changes phase from the gaseous to the solid state when being cooled to below 195 K.

The term “flow channel element” especially refers to a continuous (hollow) element (or “channel”) allowing a fluid flowing through the flow channel element from one

30 end of the flow channel element to another end of the flow channel element, such as from the inlet of the element to the outlet of the element. A channel in the flow channel element may extend from the element inlet to the element outlet. Further, the term “channel” may refer to a plurality of channels, or a channel network, especially (all) being connected to the element inlet and the element outlet. Furthermore, herein, also the terms “channel”, “fluid (flow) channel”

and “flow channel” may be used in relation to (the channel in) the flow channel element. The term “channel” may refer to a plurality of channels. The flow channel element may in embodiments comprise a tube (defining the channel). Hence, at least part of a wall of the tube (“tube wall”) may be heat conductive. The terms “element inlet” and “element outlet” especially refer to an opening in the flow channel element, allowing a flow through the flow channel element (via the channel) to enter or exit the flow channel element. The terms “element inlet” and “element outlet” may especially relate to a direction of flow of the carbon dioxide during use. The element outlet is essentially configured downstream of the element inlet.

As discussed above, a main aspect of the invention is (sub)cooling a (saturated) carbon dioxide liquid before changing its state to solid carbon dioxide. The liquid carbon dioxide is normally provided at room temperature and at elevated pressure between the pressure at the triple point and the pressure at the critical point, e.g. in the range of 10-75 bar, such as between 20-70 bar, especially between 40 and 70 bar, such as around 60 bar. By throttling the flow at the element outlet, the pressure is reduced, wherein the carbon dioxide may change its physical state. The liquid CO₂ is especially cooled between the provision of the CO₂ (or the “CO₂ supply”) in the flow channel element via the at the element inlet and the leaving of the flow channel element at the element outlet. Additionally or alternatively, the liquid carbon dioxide may be cooled upstream of the element inlet (and downstream of the carbon dioxide supply), see further below.

The flow channel element is in embodiments especially configured for exchanging heat (from external of the flow channel element to the interior of the of the flow channel element (see further below)). Therefore, at least part of the flow channel element may be configured to conduct heat.

Herein, the terms “thermal contact” and “thermally coupled” may especially refer to an arrangement of elements that may provide a thermal conductivity of at least about 10 W/m/K, such as at least 20 W/m/K, such as at least 50 W/m/K. In embodiments, the terms may especially refer to an arrangement of elements that may provide a thermal conductivity of at least about 150 W/m/K, such as at least 170 W/m/K, especially at least 200 W/m/K. In embodiments, the terms “thermal contact” and “thermally coupled” may especially refer to an arrangement of elements that may provide a thermal conductivity of at least about 250 W/m/K, such as at least 300 W/m/K, especially at least 400 W/m/K. The flow channel element and the channels space may especially be configured such that a heat flow with a value described above may be transferred from the flow channel element to the channel space, especially a fluid flowing through the channel space. This may in embodiment be based on a physical contact

between the channel space and the flow channel element. Additionally or alternatively a further (heat conductive) element may be configured between the flow channel element and the channel space, in which the heat may flow via the further element. Further, the heat conductivity of the flow channel element may have a value described above with respect to the thermal contact.

Especially, a part of the channel element that is thermally coupled to (or arranged in) the channel space may be heat conductive. In embodiments at least 50%, such as at least 60%, especially at least 75% of the flow channel element may be thermally conductive. The given percentages may refer to volume percentages or e.g. percentages of a total length of the channel in the flow channel element. In embodiments substantially 100% of the flow channel element is thermally conductive. At least part of the flow channel element may e.g. be made of a heat conductive material, or “thermally conductive material”, e.g. a metal, such as copper, brass, and stainless steel. Further suitable thermally conductive materials, for the flow channel element may be selected from the group (of thermally conductive materials) consisting of aluminum, silver, gold, and an alloy of one or more of the metals and/or thermally conductive materials. The flow channel element may be cooled with the vapor CO₂ generated by expanding the liquid CO₂ at the throttle element (flowing through the channel space).

Additionally, or alternatively, the flow channel element may be cooled with a further coolant. The flow channel element, or a channel for connecting a carbon dioxide supply to the flow channel inlet, may e.g. be partly configured in (through) a container or cooler comprising the further coolant. The further coolant may comprise any arbitrary known coolant normally used for cooling below room temperature, especially below zero degrees Celsius. The further coolant may e.g. comprise a glycol water mixture. The further coolant may comprise an (aqueous) alcohol. Further, in embodiments the further coolant may be cooled by an external cooling device such as a thermoelectric device. The system may in embodiments comprise the external cooling device especially for cooling carbon dioxide at a location upstream of the element outlet external from the device. In embodiments, the element inlet is arranged upstream of the container comprising the further coolant. In further embodiments, the element inlet is fluidically coupled to a further channel, and the container comprising the further coolant is configured for cooling the further channel, especially at a location upstream of the element inlet. Using such further coolant for cooling the liquid carbon dioxide, especially in combination with cooling the liquid carbon dioxide via (the gaseous carbon dioxide in) the channel may improve the efficiency of the device and system in warm weather conditions.

In further embodiments, the flow channel element, or the channel for connecting a carbon dioxide supply to the flow channel inlet, may be configured in thermal connection with a channel connected to the device opening (for guiding carbon dioxide vapor flowing from the device opening). In embodiments, e.g. the system may comprise an external heat exchanger
5 configured for (pre) cooling the liquid CO₂ with the gaseous CO₂ being guided away from the device opening. In embodiments, the flow channel element or the channel for connecting the carbon dioxide supply to the flow channel inlet is configured through the external heat exchanger and the channel connected to the device opening is configured through the external heat exchanger, such that gaseous vapor flowing through the channel may (pre) cool the liquid
10 CO₂ flowing to (the element inlet and) the element outlet. By pre-cooling the liquid carbon dioxide this way, an additional cooling capacity may be obtained from the gaseous carbon dioxide.

Herein, the device is especially explained based on cooling the flow channel element via the channel space with gaseous CO₂. Yet, in embodiments the feature “channel
15 space” may be changed with the feature “container comprising a further coolant. In further embodiments, the container comprising a further coolant may be arranged upstream of a location where the flow channel element is configured leaving the device (especially where the flow channel element is configured through the device wall) (i.e. especially outside the device). The system may in further specific embodiments comprise a container for comprising a further
20 coolant, wherein a part of the flow channel element is configured in the container (for cooling the flow channel element with the further coolant). The container is especially arranged outside of the device. In yet further embodiments, at least part of a further channel fluidically connected to the element inlet is configured in the container (for cooling the further channel with the further coolant).

25 The terms “upstream” and “downstream” relate to an arrangement of items or features relative to the propagation of an element such as a particle or a fluid in a channel, flow path, or hydraulic circuit, wherein relative to a first position within the channel, flow path or circuit, a second position in the channel, flow path or hydraulic circuit to an inlet (for the fluid or particle) of the channel, flow path or hydraulic circuit is “upstream”, and a third position
30 within the channel, flow path or circuit further away from the inlet “downstream”. For instance, herein a supply of the liquid CO₂ may be arranged upstream of the element inlet. The throttle element is especially arranged downstream of the element inlet.

The liquid CO₂ (1b) (provided) at the element inlet is especially a saturated liquid. Moreover, the saturated liquid CO₂ is in embodiment (provided) at room temperature

(20°C). The liquid CO₂ is in further embodiments subcooled in the flow channel element to a temperature selected from the range of 3.7-10°C, especially from the range of 5-10°C (based on a room temperature of 20°C). A temperature of the liquid CO₂ may in embodiments decrease 10-15 °C between the element inlet and the element outlet during cooling the flow channel
5 element.

In further specific embodiments, the flow channel element comprises, especially is made of, one or more materials selected from the group of a metal, copper, brass, and stainless steel. It may further be advantageous to optimize a total surface area for heat transfer from the flow channel element to the channel space.

10 The flow channel element may especially comprise a flow channel fluidically connecting the element inlet to the element outlet. A total length of the channel in the flow channel element may e.g. be optimized, especially to be large enough to allow a desired heat transfer. The term “length of the channel” may in embodiments refer to the length of the plurality of channels or the length of the channel network (comprised by the flow channel
15 element). In embodiments, e.g. a total length of the channel in the flow channel element is at least 0.1 meter, especially at least 0.25 meter, such as at least 0.5 meter or at least 0.6 meter. The total length may be limited for practical reasons. For instance, in embodiments, the total length of the channel is 5 meter at maximum, such as 3 meter at maximum, more especially 2 meter at maximum, or even 1 meter at maximum. In further embodiments, the total channel
20 length is no larger than 0.75 meter, such as 0.5 meter at maximum. Hence, in embodiments, a total length of the (flow) channel in the flow channel element is selected from the range of 0.1-5 meter, such as 0.1-2 meter, especially 0.5-2 m, especially 0.6-1 meter. The flow channel in the flow channel element may be a straight flow channel. The device may in embodiments be circular round. Such configuration may e.g. provide advantages with respect to handling high
25 pressures, or with respect to better conserving cold in the device. The flow channel element may be adapted to such device configuration. In specific embodiments the flow channel, especially the flow channel element is at least partly coiled. The heat exchanging surface of the flow channel element may further be extended by configuring fins or heat conductive extension at the flow channel element. Hence, in embodiments, the flow channel element comprises one
30 or more heat conductive extensions extending from the flow channel element.

Further, in specific embodiments, the flow channel element comprises a flow channel fluidically connecting the element inlet to the element outlet, wherein a total length of the flow channel is selected from the range of 0.1-5 meter, such as 0.1-2 meter, especially 0.5-2 m, especially 0.6-1 meter.

Because the flow channel element may exchange heat, the flow channel element, or at least part of the flow channel element may herein also be referred to as heat exchanging element or “heat exchanger”. Especially, the heat exchanging part or heat exchanger may be configured thermally coupled to the channel space, more especially may be arranged in the
5 channel space.

When flowing a saturated carbon dioxide (liquid) through the throttle element, the carbon dioxide may at least partly change phase to the solid state at the moment the carbon dioxide leaves the throttle element (and enters the confined space). Especially, based on the throttling, a part of the saturated liquid may change phase from the liquid phase (or “liquid
10 state”) to the solid phase (or “solid state”) and another part may change phase to the gas phase (or “gaseous state”). Herein, the saturated carbon dioxide (liquid) may also be referred to with the term “saturated liquid”.

The (single-phase) saturated liquid may change phase to a mixture of liquid and solid (a two-phase mixture) when reducing the pressure. The mixture will then have a reduced
15 temperature relative to the temperature of the (single-phase) saturated liquid. This process may also be known as the Joule-Thomson process, which describes the temperature change when a liquid expands from a high pressure to a low pressure in an isenthalpic process.

The system (device) is especially configured for using this temperature reduction. The vapor with the reduced temperature is especially used to cool the saturated liquid
20 in the flow channel element. More especially, the vapor is allowed to leave the confined space through the opening in the confining wall via the channel space and may ultimately leave the device through the device opening in the device wall (especially functioning as a wall of the channel space). The vapor may further be guided away through a further channel or vent coupled to the device opening. Said further channel may e.g. guide the carbon dioxide vapor
25 away from a room the device is arranged in, e.g. to prevent accumulation of CO₂ in the room.

The element outlet may comprise a (fluid) flow restriction. The flow restriction may comprise or define the throttle element. The flow restriction may comprise a reduction in cross-sectional (flow-through) area of the flow channel element. The flow restriction may in embodiments comprise a porous element configured at the end of the element outlet. The
30 porous element may comprise pores allowing a fluid to flow through. The porous element may in embodiments comprise a sintered metal plate. In further embodiments, the porous element may comprise an inorganic porous material. The flow restriction may in further embodiments comprise a cavity or narrowing. The flow restriction may comprise a capillary or capillary flow channel element arranged. The element outlet may comprise the capillary or narrowing. The

flow restriction may especially be configured for resulting in a pressure drop over the flow restriction when a fluid flows through the flow restriction.

In further embodiments, the element outlet comprises an orifice defining the throttle element. The flow-through area of the flow restriction, especially the orifice, may in further embodiments be selected based on the desired pressure drop. For instance, in
5 embodiments the orifice may have a diameter of 100 μm to provide the desired pressure drop. The orifice may in further embodiments have a diameter in the range of 1-1000 μm , especially 10-1000 μm , such as 10-50 μm . A further embodiment may comprise a plurality of orifices (in a single throttle element or in a plurality of throttle elements).

10 Essentially any flow restriction may throttle the flow of the saturated liquid. The throttle element may especially comprise a flow restriction. The flow restriction may, e.g., in embodiments be defined by the element outlet. In specific embodiments, the throttle element (especially the element outlet) may comprise an (outlet) orifice. The throttle element may in further embodiments comprise a cavity. The throttle element may comprise a porous element.
15 The throttle element may comprise a capillary flow channel element. In further embodiments, the throttle element may comprise a controllable flow restriction. The throttle element may e.g. comprise a valve. The valve may in embodiments be set (or controlled) before providing the saturated liquid to the element inlet. In further specific embodiments, the valve may be configured for actuating (i.e. changing the flow restriction) during use. Hence, in embodiments,
20 the flow restriction is controlled (changed) during flowing the liquid CO_2 through the throttle element (i.e. especially during throttling the flow of the liquid CO_2 at the element outlet). Moreover, in embodiments the method, especially the snow forming stage, comprises controlling (a flow-through area of) the flow restriction.

Hence, the throttle element may in embodiments comprise one or more of a
25 cavity, a porous element, and a capillary flow channel element.

The terms "flow restriction" and "throttle element" may independently from each other refer to a plurality of (different) flow restrictions and a plurality of (different) throttle elements, respectively. Further, in embodiments a single throttle element may comprise a plurality of flow restrictions. The throttle element may e.g. comprise a capillary flow channel
30 element comprising a porous element optionally further including a valve.

The element outlet comprises the throttle element, especially for throttling a fluid leaving the flow channel element at the element outlet. Hence, in embodiments the throttle element comprises one or more of a cavity, a porous element, a capillary flow channel element, and a controllable flow restriction.

The element outlet with the throttle element (e.g. the orifice) may in embodiments be arranged in the confined space. The element outlet may extend into confined space. Alternatively, the element outlet may be arranged in a wall of confined space (the confining wall), such that a fluid flowing from the element outlet may flow into the confined space. Essentially, the element outlet is arranged such that the element outlet, especially the throttle element, is configured in direct fluid connection with the confined space.

The term “flow-through” in expressions like “flow-through channel” or “flow-through area” especially refers to a structure or device such as a channel wherein a fluid may enter the structure or device at a first location (or end) and exit the structure or device at a further location (or end), different from the first location.

In this application, the feature “cross-sectional area” is used. In that respect, the cross-sectional area can also be indicated as the flow-through cross-sectional area or cross section. This cross-sectional area can be rectangular, elliptic, or round (such as a ring), for instance.

As discussed above, based on throttling the flow of liquid CO₂, in the confined space dry ice snow may be formed in combination with CO₂ vapor. The dry ice snow may be sprayed (directly from the throttle element) over a sample arranged in the confined space. In further embodiments, the dry ice snow may be compacted or “densified” in the confined space. The vapor may leave the confined space, via the opening in the confining wall. However, preferably, the dry ice is kept in the confined space. The opening (in the confining wall may in embodiments be configured for allowing a vapor to pass through the opening, and especially for (at least partly) blocking a transport of solid CO₂ through the opening. The opening may e.g. comprise a pore. The opening may in embodiments have a cross-sectional area, or flow-through area of 1 mm² at maximum, such as 0.5 mm² at maximum, especially 0.25 mm² at maximum. In further embodiments, the flow-through area may be in the range of 0.01-2 mm², such as in the range of 0.01 – 1 mm², especially in the range of 0.1 – 1 mm². The flow through area may especially be at least 0.01 mm². A small opening may restrict a total flow of gaseous CO₂ away from the confined space. Therefore, in embodiments, the confining wall may comprise more than one opening. The opening and/or plurality of openings may be configured to control a flow of the gaseous carbon dioxide. The confining wall may in embodiments comprise a porous wall.

By allowing the gaseous carbon dioxide to leave the confined space while blocking substantially all of the solid carbon dioxide, the particulate solid CO₂ may be densified in the confined space. In further embodiments the device may further comprise a densifying

element configured for densifying solid CO₂ provided in the confined space. In embodiments, e.g., alternately dry ice snow may be sprayed in the confined space, and the dry ice snow may be densified with the densifying element. The densifying element may, e.g., in embodiments comprise a piston or plunger.

5 It will be understood that at least during use of the device, especially during spraying the dry ice and or densifying the dry ice, the confined space is preferably closed, except for the opening in the confining wall. Yet, for placing a sample in the confined space and/or removing densified dry ice from the confined space, the confined space (or confining wall) may especially also be opened. In further embodiments, the device further comprises a
10 closing element for closing the confined space. The closing element may in embodiments comprise a removable closing element. The closing element may in further embodiments close the confining wall. In embodiments, the closing element may be screwed in the confining wall. In further embodiment, the closing element and the confining wall define a screw mount. They may further define a bayonet mount. The closing element may especially seal the confined
15 space at the confining wall.

The gaseous CO₂ may flow from the confined space into the channel space and may successively exit the channel space via the device opening. The channel space is especially thermally coupled to the flow channel element, indicating that while being in (flowing through) the channel space, the gaseous CO₂ may cool the flow channel element. In the channel space,
20 the gaseous CO₂ especially may in embodiments contact the flow channel element (wall). While contacting the flow channel element, the gaseous CO₂ may cool the flow channel element, and especially based on that, the liquid CO₂ flowing through the flow channel element may be cooled. In embodiments, the snow forming stage comprises cooling the flow channel element (especially being at least partly conductive) with (the) gaseous CO₂ (formed in the confined
25 space) to cool the liquid CO₂ flowing through the flow channel element.

Hence, the term “cooling the flow channel element” may in embodiments be replaced by (sub)cooling the liquid CO₂ (flowing) in (or flowing through) the flow channel element.

Preferably, the cold in the gaseous CO₂ is used efficiently for cooling the liquid
30 CO₂ flowing through the flow channel element. The device opening may therefore in embodiments be configured to optimize a flow of the gaseous CO₂ along the flow channel element. In embodiments, a flow path from the opening in the confining wall to the device opening is maximized. In further embodiments, the flow channel element is arranged in the flow path. Further a heat transfer from external of the device into the device may also be

reduced. In further embodiments the device wall comprises an insulated device wall. The insulated device wall may save energy. A further advantage of insulating the device is that operating the device is safer since cold surfaces an operator may be exposed with are reduced or eliminated.

5 Hence, in embodiments the snow forming stage comprises throttling the flow of liquid CO₂ (1b) at the element outlet, thereby forming a mixture a particulate solid CO₂ flow (1a) and a gaseous CO₂ (1c), and guiding the gaseous CO₂ (1c) away from the confined space along the flow channel element to cool the flow channel element. Especially, cooling the flow channel element may in embodiments comprise guiding the gaseous CO₂ (1c) away from the
10 confined space along the flow channel element. It is noted that optionally the particulate solid CO₂ sprayed from the element outlet may sublime providing further gaseous CO₂.

The term “opening” such as relation with the device opening and the opening in the confined wall may refer to a plurality of (different) openings.

The pressure in the confined space is especially controlled for having CO₂ in the
15 solid state (and the gaseous state). The pressure is especially in the range of the pressure of the triple point and the pressure of the critical point of CO₂. In embodiments, one or more of the device opening and the opening (in the confining wall) may be configured for maintaining the pressure in the confined space in this range.

In embodiments, especially wherein the cooling stage comprises spraying the
20 particulate solid CO₂ over the sample arranged in the confined space, the method comprises maintaining a pressure in the confined space above a pressures of the triple point pressure of CO₂ and below the pressure of the critical point of CO₂.

In further embodiments, especially wherein the cooling stage comprises densifying the particulate solid CO₂ (1a) in the confined space, during the snow forming stage
25 and during densifying the particulate solid CO₂, the pressure in the confined space is maintained above the pressure of the triple point pressure of CO₂ and below the pressure of the critical point of CO₂.

The method, especially wherein the cooling stage comprises densifying the particulate solid CO₂ (1a) in the confined space, may in further embodiments comprise
30 collecting the densified particulate solid CO₂ (1a) in a container, especially a heat insulated container, and providing the sample in the container to cool the sample with the densified particulate solid CO₂.

Herein the term “cooling” may include, maintaining a predetermined (cool) temperature. The sample may, e.g., be precooled and maintained at a temperature using the densified dry ice.

The pressure in the device, especially in the channel space, may in embodiments
 5 be controlled. For instance, a back pressure device (back pressure regulator, back pressure valve) is in embodiments configured downstream of the device opening. The back pressure device may be set for controlling the pressure in the device, especially in the channel space between a pressure of the liquid CO₂ provided at the element inlet (“inlet pressure”) and the pressure of the triple point. The pressure may in embodiments be set at a pressure in the range
 10 of the 50-99% of the inlet pressure, such as in the range of 50-90% of the inlet pressure. In further specific embodiments, the method comprises controlling the pressure in the confined space while providing liquid CO₂ (1b) to the element inlet, wherein the particulate solid CO₂ is densified by an increasing pressure in the confined space. The pressure in the channel space may especially be in the range of the atmospheric pressure, such as in the range of 1-1.5,
 15 especially 1-1.2 times atmospheric pressure,

Hence, in embodiments, the sample may be arranged in the confined space prior to the snow forming stage. The sample may be removed after the cooling stage and may e.g. be stored in a cooled environment, such as in a container comprising (densified) dry ice, or a container comprising liquid nitrogen.

20 In further embodiments the device is used to provide (produce) densified particulate CO₂ and the densified particulate CO₂ (dry ice pellets) is used to cool the sample. The sample thus not necessarily is arranged in the confined space.

In a further aspect, the invention also provides a method for producing dry ice. The dry ice production method especially comprises flowing liquid CO₂ (especially at room
 25 temperature) through a flow channel element from an element inlet to an element outlet (of the flow channel element) while cooling the flow channel element. The dry ice production method may further comprise throttling the flow of the liquid CO₂ at the element outlet, wherein the liquid CO₂ forms mixture comprising (or “of”) (a) particulate solid CO₂ and (a) gaseous CO₂, the mixture spraying from the element outlet into a confined space. In further embodiments,
 30 the dry ice method comprises densifying the particulate solid CO₂ in the confined space. Further, especially the dry ice method may comprise collecting the densified solid CO₂ to provide the dry ice.

The dry ice production method may in embodiment be part of the method for cooling. The dry ice production method may be performed using the system described herein.

The method and the system are especially for subcooling liquid carbon dioxide and successively expanding the liquid carbon dioxide, wherein the carbon dioxide changes phase.

Herein, the term “phase” such as in phrases like “wherein the liquid changes
5 phase”, “phase changing of the liquid”, and the like, especially relate to a state of a substance that changes. The substance (carbon dioxide) may e.g. change from the liquid state to the gaseous state (“vaporize”; from a liquid to a vapor), from a liquid state to a solid state (“solidify”), or e.g. from a solid state to a gaseous state (also named “sublime”). The terms “gas phase”, “gaseous phase” and “vapor phase” may be used interchangeably, especially referring
10 to a state (or phase) of the substance at a temperature above the boiling point of the substance (at a given pressure). The term “changing phase” may further especially refer to a change from a single phase (liquid) to a two-phase system. As is indicated above, the single liquid phase may in embodiments change to a two-phase mixture comprising liquid and vapor (CO₂). Hence, changing phase may also refer to a single (liquid) phase changing to a (mixture of) a solid phase
15 and a gaseous phase. Further, the phrase “wherein at least part of the cryogenic liquid changes phase” may refer to the (single phase) cryogenic liquid changes to a two-phase mixture”. The term “two-phase mixture may refer to a mixture comprising the liquid and vapor (of the carbon dioxide).

20 BRIEF DESCRIPTION OF THE DRAWINGS

Embodiments of the invention will now be described, by way of example only, with reference to the accompanying schematic drawings in which corresponding reference symbols indicate corresponding parts, and in which Figs 1-2 and 3A and 3B schematically depict embodiments of the device (and the system); Fig. 4 schematically depicts a further
25 embodiment of the system; and Figs 5A and 5B depict some further aspects of the system. The schematic drawings are not necessarily on scale.

DETAILED DESCRIPTION OF THE EMBODIMENTS

Figs. 1-3 schematically depict embodiments of the system 100. They also depict
30 embodiment of the device 10. The system 100 (and the device 10) may be used for producing solid CO₂ (“dry ice”) 1, 1a and for cooling,

In the figures, the reference number 1 is used to indicate the substance carbon dioxide in general. Further, the state of CO₂ is also indicated. Reference 1a, refers to the solid state of CO₂ 1, or “solid CO₂” 1, 1a. Reference 1b, refers to the liquid state of CO₂ 1, or “liquid

CO₂” 1,1b, and reference 1c, refers to the gaseous state of CO₂ 1, also indicated as CO₂ vapor 1,1c.

The depicted devices 10 comprise a device wall 15, a flow channel element 20, a confining wall 35, and a channel space 40. The confining wall 35 during use defines a confined space 30. Yet when not in use, the defining wall 35 may be open (such as in the depicted embodiments) e.g., at the bottom. Herein, the confined space 30 may also be indicated as chamber 30. Further, the terms “chamber” in relation to phrases like “entering the chamber”, “arranging a sample in the chamber”, “leaving the chamber”, “closing the chamber”, etc. may especially refer to the interior of the chamber.

The device wall 15 comprises a device opening 19. In the figures only one opening 19 is depicted. Yet, in further embodiments, the wall 15 may comprise a plurality of device openings 19. The flow channel element 20 is at least partly heat conductive. In the depicted embodiments, the flow channel element 20 comprises a (coiled) tube, comprising an at least partly heat conductive tube wall 25. The confining wall 35 encloses the confined space 30. In Fig. 1, at the top side of the device 10 a volume 41 is indicated. This volume 41 may in embodiments be an open space being part of the channel space 40. In further embodiments, the volume 41 may be closed and may e.g. be part of the confining wall 35.

Further, the confining wall 35 and the device wall 15 define the channel space 40. The confining wall 35 further comprises a plurality of openings 31 for allowing a gaseous fluid, such as gaseous CO₂ 1,1c to leave (the interior of) the confined space 30. The openings 31 are further especially configured for blocking solid carbon dioxide 1a

The gaseous CO₂ 1,1c may enter the channel space 40 via the openings 31 and leave the space 40 via the device opening 19. Herein, this may also be indicated that the openings 31 may be arranged at an upstream side of the channels space 40, and the device opening 19 is arranged at a downstream side of the channel space 40. The channel space 40, thus, fluidically connects the confined space 30 to the device opening 19 via the plurality of openings 31. In the depicted embodiments of Figs 1-2, the element inlet 21 is configured through the device wall 15 and almost the entire flow channel element 20 is arranged in the channel space 40. In further embodiments, see e.g. Fig 3, the element inlet 21 may be configured as an opening in the device wall 15 (in fluid connection with the remainder of the flow channel element 20) and may substantially not extend from the device wall 15. This may also be indicated as the element inlet 21 is configured at an outer side 11 of the device 10. This term “at an outer side 11” especially indicates that the element inlet 21 is configured such that

a fluid, especially a liquid carbon dioxide 1,1b, may be provided from the outside 11 of the device 10 to the flow channel element 20 at the element inlet 21.

Further, the element outlet 29 comprises a throttle element 22 that extends in the confined space 30. As such, the confined space 30 is in open fluid connection with the flow channel element 20 at the element outlet 29. Based on the throttle element, the CO₂ 1 changes phase from the single liquid phase 1b to a mixture comprising CO₂ 1 in the gaseous phase 1c, and in the solid phase 1a (especially having a particulate appearance). Moreover, the flow of the liquid CO₂ 1b may especially be throttled at the element outlet 29, wherein the liquid CO₂ 1b forms a mixture 2 comprising particulate solid CO₂ 1a and a gaseous CO₂ 1c spraying from the element outlet 29 into the confined space 30. The throttle element 22 is especially a flow restriction and may e.g. comprise a fixed restriction such as a cavity, a porous element, and/or a capillary flow channel element. The restriction may also be controllable flow restriction, such as a valve.

Further, the gaseous CO₂ 1,1c (coming from the confined space 30) and flowing along the flow channel element 20 (in the channel space 40) may cool the liquid CO₂ flowing through the flow channel element 20. The figures, especially depict (during use) is an embodiment of “flowing liquid CO₂ 1b through the flow channel element 20 from an element inlet 21 to the element outlet 29 while cooling the flow channel element 20”. The carbon dioxide vapor 1,1c may cool the flow channel element 20 because the channel space 40 and the flow channel element 20 are especially thermally coupled to each other, especially allowing a heat (or cold) flow between these elements 40,20.

To allow a good heat transfer, at least part of the flow channel element 20, is made of a thermally conductive material, such as a metal. The metal may e.g. be selected from the group of a metal, copper, brass, and stainless steel. In the embodiments of Figs 1-2, the flow channel element 20 is coiled around the confining wall 35 comprising the volume 41, illustrated by the visible cross sections of the flow channel element 20 in the channel space 40. The flow channel element 20 may in embodiments comprise a channel 24 fluidically connecting the inlet 21 to the outlet 29. The channel 24 may be a single channel 24, such as in the depicted embodiments comprising the coiled tube. Yet, the term “channel” 24 may also refer to a plurality of channels 24 or a network of channels 24. Hence, in alternative embodiments not depicted, the flow channel element 20 may comprise a network of channels 24. The flow channel element 20 may essentially comprise any arbitrary shape. Also in embodiments with a plurality of channels 24 and/or a channel network 24, the flow channel element 20 is configured

in thermal contact with the channel space 40 (and the element inlet 21 is arranged at the outer side 11 of the device 10, and the element outlet in fluid connection with the confined space 30).

The channel 24 may have a predetermined length. The total length of the channel 24 (including the channels 24 in the network of channels 24) may in embodiments, e.g., be 0.1-5 m, or 0.1-2 m, especially 0.5-2 m, even more especially 0.6-1 m. The channel 24 (and the flow channel element 20) may further have any arbitrary cross section and not necessary is based on a round channel 24 as depicted in the Fig 1-3. To increase the area that may transfer heat from the flow channel element 20 to the gaseous CO₂ 1c, the flow channel element 20 may comprise one or more heat conductive extensions 26 extending from the flow channel element 20 (the tube in the depicted embodiments), as is very schematically indicated at locations of the tube wall 25 in Fig 2. In the embodiment depicted in Fig. 3 other extension 26 are shown extending from the channel element 20. Further, the device wall 15 may be insulated and may have a heat insulating layer 15a. Moreover, in the depicted embodiments, the wall 15 comprises, especially is, an (heat) insulated device wall 15.

The confined space 30 may further be open(ed) and closed, for instance for arranging a sample 5 in the confined space 30 and/or for removing densified CO₂ 1,1a from the confined space 30. Therefore, the device 10 may further comprise a removable closing element 16 for opening the confined space 30 and for closing the confined space 30. Yet, in the embodiment depicted in Fig. 1, the confined space 30 may be closed by arranging the device 10 at a surface 6.

In Fig. 3a embodiment of the device 10 is depicted. In Fig. 3, the channel 24 of the flow channel element 20 comprises extensions 26 shaped like fins and attached to the channel 24. The extensions 26 are arranged radially extending from the channel 24 and allow a high heat transfer between the gaseous CO₂ 1c, flowing in the channel space 20, with the liquid CO₂ 1a flowing through the channel 24 of the channel element 20.

In Fig 4, very schematically a system 100 is depicted comprising a plurality of devices 10 all coupled to a (single) liquid CO₂ supply 60. This may e.g. allow operating the respective devices 10 sequentially, resulting is a semi-continuous process. Switching between the devices 10 may e.g. be done manually. Additionally, or alternatively, the system further comprises a control system 70 and control devices 80 configured to control a flow of liquid carbon dioxide 1,1b to a respective device 10. The control system 70 may be functionally coupled to the control devices 80. The control system 70 may further be functionally coupled to other controllable elements of the device 10, e.g. for controlling a pressure in the confined space 30.

Fig. 5A and 5B schematically depict some further embodiments of the system 100. In the depicted embodiments, the device 10 is indicated only very schematically. Fig 5A, schematically depicts an embodiment wherein the liquid CO₂ 1,1b is further cooled with a further coolant 51. In the embodiment a channel for providing the liquid CO₂ 1,1b to the device 10 is partly configured in (through) a container 50 comprising the further coolant 51. This channel may be (part of) the flow channel element 20 but may also be a channel connecting a liquid carbon dioxide 1,1b supply to the flow channel inlet 21. The further coolant 51 may e.g. comprise a glycol water mixture. The further coolant 51 is cooled (in the container 50) by an external cooling device 52, e.g., a thermoelectric device.

In Fig 5B the flows of the carbon dioxide 1 are very schematically depicted (without directly indicating the structural elements guiding the carbon dioxide 1). In the embodiment, the flow channel element 20, or the channel for connecting a carbon dioxide supply 60 to the flow channel inlet 21 is configured in thermal connection with a channel connected to the device opening 19 (for guiding carbon dioxide vapor 1,1c flowing from the device 10). In the embodiment, the system 10 comprises an external heat exchanger 55 for precooling the liquid CO₂ 1,1b flow with the gaseous CO₂ 1,1c flow. Such external heat exchanger 55 may be combined with the channel element 20, or also indicated as (internal) heat exchanger 20. Yet in embodiments, the device 10 may only comprise the external heat exchanger 55. The flow channel element 20 may in embodiments directly provide the liquid carbon dioxide 1,1b into the confined space 30, without exchanging heat with a channel space 40. (The device 10 may lack the channel space 40) The embodiment of Fig. 5B also comprises the container or cooler 50 with the further coolant 51 for cooling the liquid CO₂ 1,1b upstream of the device 10. In further embodiments, that cooler 50 may be left out.

Hence in embodiments, the system 100 comprises the external heat exchanger 55 and/or the cooler /container 50 for pre-cooling the liquid carbon dioxide 1,1b.

The figures further illustrate embodiments of the methods of the invention. The method may especially comprise a snow forming stage and a cooling stage.

In the snow forming stage, liquid CO₂ 1,1b, may be provided to the element inlet 21. The liquid CO₂ 1,1b may e.g. be stored in a gas cylinder 60 at room temperature and at elevated pressure, e.g. at 60 bar, and may especially comprise a saturated liquid. The liquid CO₂ 1b is further especially flown through the flow channel element 20 from the element inlet 21 to the element outlet 29. At the same time, the flow channel element 20 may be cooled, e.g. based on a flow of the gaseous CO₂ 1c along the flow channel element 20 and/or based on a further cooling of the flow channel element 20. Such further cooling is not depicted and may

e.g. comprise arranging the flow channel element is a further coolant like liquid nitrogen instead of or additionally to arranging the flow channel element 20 in thermal contact with the channel space 40. The system 100 may in embodiments comprise a further cooling device comprising the further coolant. The snow forming stage further comprises throttling the flow of the liquid CO₂ 1,1b at the element outlet 29. This way the liquid CO₂ 1b may form a particulate solid CO₂ 1,1a spraying from the element outlet 29 into a confined space 30. It is noted that at the same time gaseous CO₂ 1,1c is formed in the confined space 30. The gaseous CO₂ 1c is especially guided away from the confined space 30 (through the openings 31 in confining wall 35) along the flow channel element 20 to cool the flow channel element 20 and the liquid CO₂ 1,1b flowing through the flow channel element 20. The liquid CO₂ 1,1b in the flow channel element 20 may in embodiments be cooled with 10 to 15 °C, or even more (temperature difference between the CO₂ at the element inlet 21 and at the element outlet 29). Based on the cooling of the liquid CO₂ 1,1b, the ratio gaseous CO₂ 1,1c to solid CO₂ 1,1a in the mixture 2 being formed at the element outlet 29 is reduced.

The cooling stage may in embodiments (depicted in Fig. 1), comprise spraying the mixture 2 (particulate solid CO₂ 1,1a, especially dry ice snow 1,1a, in combination with the gaseous CO₂ 1,1c) over the sample 5 arranged in the confined space 30. During spraying the mixture 2, the pressure in the confined space 30 may be maintained between the triple point pressure of CO₂ 1 and the critical point pressure of CO₂ 1.

In the embodiment depicted in Fig. 2, a part of the cooling stage is depicted, comprising densifying the particulate solid CO₂ 1,1a (to densified dry ice 1a, or dry ice pellets 1a) in the confined space 30. During the cooling stage a pressure in the confined space 30 may be controlled (increased) to further facilitate densifying the dry ice 1,1a in the confined space 30.

The cooling stage may further comprise cooling a sample 5 (external from the device 10) with the densified particulate solid CO₂ 1,1a, especially after first removing the densified dry ice 1,1a from the device 10. In such cooling stage, the pressure in the confined space 30 is especially maintained above the pressure of the triple point of CO₂ 1 and below the pressure of the critical point of CO₂ 1 during densifying the CO₂ 1,1a. The method may in embodiments (not depicted) comprise collecting the densified dry ice 1,1a in a heat insulated container, and providing an optionally precooled sample 5 in the container to (further) cool (including controlling a temperature of the sample 5 at a predetermined value) the sample 5.

The snow forming stage depicted in the Figs 1-2 comprises cooling the flow channel element 20 with gaseous CO₂ 1,1c.

The method may be used for any temperature sensitive sample, such as tissue or other human material for diagnostic tests, a vaccine or other biological product, and a medical product or (medical) drug.

Fig. 2 further schematically also illustrates the method for producing dry ice, described herein. In that method, liquid CO₂ 1,1b is flown in (through) the flow channel element 20 at (from) an element inlet 21 (especially at room temperature) to an element outlet 29 while cooling the flow channel element 20. The flow of the liquid CO₂ 1,1b is throttled at the element outlet 29, and the liquid CO₂ 1,1b forms the mixture 2 comprising a dry ice snow 1,1a spraying from the element outlet 29 into the confined space 30. Next, the method comprises densifying the particulate solid CO₂ 1,1a in the confined space 30 (obtaining densified dry ice 1,1a), and the densified solid CO₂ (dry ice) 1,1a is collected to provide the dry ice 1,1a.

The technology and the process of making dry ice 1a using the system 100 and the method of the invention especially makes use of a recuperation step is described herein. The main elements may in embodiments be described as: At first, a high-pressure liquid CO₂ 1,1b, passing through an inner flow channel element 20 of a finned heat exchanger flow channel element 20 (with fins 26), expands through an orifice 22 or throttle element 22. This creates dry ice snow 1a and cold CO₂ gas 1c as schematically shown in Figs. 1-2. The snow 1b starts to accumulate in the expansion chamber 30 and the cold CO₂ gas 1c escapes through small holes 31 present on the periphery or wall 35 of the expansion chamber 30. The cold gas 1c coming out of the expansion chamber 30 travels upwards and passes over (the fins 26 of) the heat exchanger 20, thereby transferring energy and precooling the incoming liquid CO₂ 1b. The precooled liquid 1b then expands through the orifice 22 to form more solid CO₂ 1a snow. This process continues until the chamber 30 is full of CO₂ snow. Because of the clogging of the orifice 22 (and the holes 31 on the expansion chamber 30) the (snow forming) process stops. The bottom lid 16 is then opened (after stopping the liquid carbon dioxide 1,1b supply) to obtain the densified form of dry ice 1a to be used for packaging and transportation.

A further advantageous aspect of this device 10 is to facilitate high cooling rates that may be required to prepare the temperature sensitive bio samples 5. This can be easily done by opening the bottom lid 16 in the process shown in Fig. 2 or lifting the device 10 of Fig. 1, allowing CO₂ snow 1a and cold gas 1c to directly cool the samples. Low temperature of CO₂ snow 1a, high heat transfer coefficient, and high velocities of CO₂ snow spray 1a ensures high cooling rates of samples 5.

The device 10 may producing higher amount of dry ice 1a than the existing solutions from a given amount of liquid CO₂ 1b. The existing solutions limits dry ice 1a

production to a maximum of 4.5 kg from 20 kg of liquid CO₂ 1b bottle. Whereas, using technology of the invention, one may produce approximately 8 to 9 kg of dry ice 1a, thereby improving the conversion efficiency by 100 % compared to existing solutions. The method and device have an added advantage of being operated in dual mode, namely, to produce densified
 5 dry ice ($\Delta T = 0$) for packaging of bio samples 5, and to prepare the bio samples 5 ($\Delta T < 0$) by freezing them at the cooling rates required in a medical setting. It is noted that herein both processes (packaging of bio samples 5 (with $\Delta T = 0$)), and preparing the bio samples 5 ($\Delta T < 0$) by freezing) are indicates as cooling processes. In the existing solutions, after making dry ice 1a, the user will come in direct contact with the expansion chamber 30 to get the dry ice out
 10 of their device.

Therefore, to avoid any burns from very cold temperatures of the chamber 30 (-78°C) of present systems, user of the existing solution needs safety gloves. In the present device 10, the expansion chamber 30 may be isolated by insulation material 15a, making the production of dry ice safe and ergonomic.

15 The device 10 may produce dry ice 1a in a clinical setting to freeze and package temperature sensitive samples 5. It not only reduces user dependency on dry ice supplier but also allows on-demand and efficient production of dry ice 1a. Some key features of embodiments of the inventions are: Dual functionality - Cooling ($\Delta T < 0$) + packaging ($\Delta T = 0$) samples; Less maintenance - No moving parts; Ergonomic and Safe — No cold fingers and
 20 no need of safety gloves; Portable — tabletop arrangements possible; Can be re-used; Can be produced in different sizes.

The devices, apparatus, or systems may herein amongst others be described during operation. As will be clear to the person skilled in the art, the invention is not limited to methods of operation, or devices, apparatus, or systems in operation.

25 The system, or apparatus, or device may execute an action in a “mode” or “operation mode” or “mode of operation”. Likewise, in a method, an action or stage, or step may be executed in a “mode” or “operation mode” or “mode of operation”. The term “mode” may also be indicated as “controlling mode”. This does not exclude that the system, or apparatus, or device may also be adapted for providing another controlling mode, or a plurality
 30 of other controlling modes. Likewise, this may not exclude that before executing the mode and/or after executing the mode one or more other modes may be executed.

However, in embodiments a control system may be available, that is adapted to provide at least the controlling mode. Would other modes be available, the choice of such modes may especially be executed via a user interface, though other options, like executing a

mode in dependence of a sensor signal or a (time) scheme, may also be possible. The operation mode may in embodiments also refer to a system, or apparatus, or device, that can only operate in a single operation mode (i.e. “on”, without further tunability).

The term “controlling” and similar terms herein especially refer at least to determining the behavior or supervising the running of an element. Hence, herein “controlling” and similar terms may e.g. refer to imposing behavior to the (controllable) element (determining the behavior or supervising the running of an element), etc., such as e.g. measuring, displaying, actuating, opening, shifting, changing temperature, etc., especially actuating. Beyond that, the term “controlling” and similar terms may additionally include monitoring. Hence, the term “controlling” and similar terms may include imposing behavior on an element and also imposing behavior on an element and monitoring the element. The controlling of the element can be done with the control system. The control system and the (controllable) element may thus at least temporarily, or permanently, functionally be coupled. The element may comprise at least part of the control system. In embodiments, the control system and element may not be physically coupled. Control can be done via wired and/or wireless control. The term “control system” may also refer to a plurality of different control systems, which especially are functionally coupled, and of which e.g. one control system may be a master control system and one or more others may be slave control systems.

The term “plurality” refers to two or more. Furthermore, the terms “a plurality of” and “a number of” may be used interchangeably.

The terms “substantially” or “essentially” herein, and similar terms, will be understood by the person skilled in the art. The terms “substantially” or “essentially” may also include embodiments with “entirely”, “completely”, “all”, etc. Hence, in embodiments the adjective substantially or essentially may also be removed. Where applicable, the term “substantially” or the term “essentially” may also relate to 90% or higher, such as 95% or higher, especially 99% or higher, even more especially 99.5% or higher, including 100%. Moreover, the terms “about” and “approximately” may also relate to 90% or higher, such as 95% or higher, especially 99% or higher, even more especially 99.5% or higher, including 100%. For numerical values it is to be understood that the terms “substantially”, “essentially”, “about”, and “approximately” may also relate to the range of 90% - 110%, such as 95%-105%, especially 99%-101% of the values(s) it refers to.

The term “comprise” also includes embodiments wherein the term “comprises” means “consists of”.

The term “and/or” especially relates to one or more of the items mentioned before and after “and/or”. For instance, a phrase “item 1 and/or item 2” and similar phrases may relate to one or more of item 1 and item 2. The term “comprising” may in an embodiment refer to “consisting of” but may in another embodiment also refer to “containing at least the
5 defined species and optionally one or more other species”.

Furthermore, the terms first, second, third and the like in the description and in the claims, are used for distinguishing between similar elements and not necessarily for describing a sequential or chronological order. It is to be understood that the terms so used are interchangeable under appropriate circumstances and that the embodiments of the invention
10 described herein are capable of operation in other sequences than described or illustrated herein.

The term “further embodiment” and similar terms may refer to an embodiment comprising the features of the previously discussed embodiment but may also refer to an alternative embodiment.

It should be noted that the above-mentioned embodiments illustrate rather than
15 limit the invention, and that those skilled in the art will be able to design many alternative embodiments without departing from the scope of the appended claims.

In the claims, any reference signs placed between parentheses shall not be construed as limiting the claim.

Use of the verb “to comprise” and its conjugations does not exclude the presence
20 of elements or steps other than those stated in a claim. Unless the context clearly requires otherwise, throughout the description and the claims, the words “comprise”, “comprising”, “include”, “including”, “contain”, “containing” and the like are to be construed in an inclusive sense as opposed to an exclusive or exhaustive sense; that is to say, in the sense of “including, but not limited to”.

25 The article “a” or “an” preceding an element does not exclude the presence of a plurality of such elements.

The invention may be implemented by means of hardware comprising several distinct elements, and by means of a suitably programmed computer. In a device claim, or an apparatus claim, or a system claim, enumerating several means, several of these means may be
30 embodied by one and the same item of hardware. The mere fact that certain measures are recited in mutually different dependent claims does not indicate that a combination of these measures cannot be used to advantage.

The invention also provides a control system that may control the device, apparatus, or system, or that may execute the herein described method or process. Yet further,

the invention also provides a computer program product, when running on a computer which is functionally coupled to or comprised by the device, apparatus, or system, controls one or more controllable elements of such device, apparatus, or system.

The invention further applies to a device, apparatus, or system comprising one
5 or more of the characterizing features described in the description and/or shown in the attached drawings. The invention further pertains to a method or process comprising one or more of the characterizing features described in the description and/or shown in the attached drawings. Moreover, if a method or an embodiment of the method is described being executed in a device, apparatus, or system, it will be understood that the device, apparatus, or system is suitable for
10 or configured for (executing) the method or the embodiment of the method, respectively.

The various aspects discussed in this patent can be combined in order to provide additional advantages. Further, the person skilled in the art will understand that embodiments can be combined, and that also more than two embodiments can be combined. Furthermore, some of the features can form the basis for one or more divisional applications.

Conclusies

1. Een systeem (100), waarbij het systeem (100) een inrichting (10) omvat, de inrichting (10) omvattende (i) een inrichtingswand (15) met een inrichtingsopening (19), een (ii) stromingskanaalelement (20) omvattende een elementinlaat (21) en een elementuitlaat (29), waarbij het stromingskanaalelement (20) ten minste gedeeltelijk warmtegeleidend is, (iii) een begrenziingswand (35) voor het definiëren van een beperkte ruimte (30), en (iv) een kanaalruimte (40), waarin
 - 10 - de begrenziingswand (35) een aantal openingen (31) omvat om een gasvormig fluïdum de beperkte ruimte (30) te laten verlaten,
 - de inrichtingswand (15) en de begrenziingswand (35) de kanaalruimte (40) definiëren, waarbij de kanaalruimte (40) de beperkte ruimte (30) met de inrichtingsopening (19) via het aantal openingen (31) fluïde verbindt en waarbij
 - 15 het stromingskanaalelement (20) thermisch gekoppeld met de kanaalruimte (40) is geconfigureerd,
 - waarbij de elementinlaat (21) is uitgevoerd aan een buitenzijde (11) van de inrichting (10) en de elementuitlaat (29) is uitgevoerd in open fluïdumverbinding met de beperkte ruimte (30), en
 - 20 - de elementuitlaat (29) een smoorelement (22) omvat.
2. Het systeem (100) volgens conclusie 1, waarbij ten minste een deel van het stromingskanaalelement (20) is aangebracht in de kanaalruimte (40).
- 25 3. Het systeem (100) volgens één van de voorgaande conclusies, waarbij het stromingskanaalelement (20) een of meer warmtegeleidende uitbreidingen (26) omvat die zich uitstrekken vanaf het stromingskanaalelement (20).
4. Het systeem (100) volgens één van de voorgaande conclusies, waarbij het
 - 30 smoorelement (22) een of meer van een holte, een poreus element, een capillair stromingskanaalelement en een regelbare stromingsbeperking omvat.

5. Het systeem (100) volgens één van de voorgaande conclusies, waarbij het stromingskanaalelement (20) een stromingskanaal (24) omvat dat de elementinlaat (21) op fluïde wijze verbindt met de elementuitlaat (29), waarbij een totale lengte van het stroomkanaal (24) wordt gekozen uit het bereik van 0,1-5 meter, zoals 0,1-2 meter, in het bijzonder 0,5-2 m, in het bijzonder 0,6-1 meter.

6. Het systeem volgens één van de voorgaande conclusies, waarbij het stroomkanaalelement (20) is gemaakt van een of meer materialen gekozen uit de groep van metaal, koper, messing en roestvrij staal.

10

7. Het systeem (100) volgens één van de voorgaande conclusies, waarbij de inrichtingswand (15) een geïsoleerde inrichtingswand (15) omvat.

8. het systeem (100) volgens één van de voorgaande conclusies, waarbij de inrichting (10) verder een verwijderbaar sluitelement (16) omvat voor het afsluiten van de beperkte ruimte (30).

9. Een werkwijze voor het koelen, waarbij de werkwijze een sneeuwvormingsstadium en een koelstadium omvat, waarbij het sneeuwvormingsstadium omvat:

- het stromen van vloeibaar CO₂ (1b) door een stroomkanaalelement (20) van een elementinlaat (21) naar een elementuitlaat (29) terwijl het stroomkanaalelement (20) wordt gekoeld, en

- het smoren van de stroom van het vloeibare CO₂ (1b) bij de elementuitlaat (29), waarbij het vloeibare CO₂ (1b) een mengsel (2) vormt dat een deeltjesvormig vast CO₂ (1a) en gasvormig CO₂ (1c) omvat, waarbij het mengsel (2) vanaf de elementuitlaat (29) in een beperkte ruimte (30) sproeit;

en waarbij het koelstadium er één omvat van

(i) het sproeien van het mengsel (2) over een monster (5) gerangschikt in de beperkte ruimte (30); en

(ii) het verdichten van het deeltjesvormige vaste CO₂ (1a) in de beperkte ruimte (30) en het koelen van een monster (5) met het verdichte deeltjesvormige vaste CO₂ (1a).

5 10. De werkwijze volgens conclusie 9, waarbij het stromingskanaalelement (20) ten minste gedeeltelijk warmtegeleidend is, waarbij het sneeuwvormingsstadium het koelen van het stromingskanaalelement (20) met gasvormig CO₂ (1c) omvat om het door het stromingskanaalelement (20) stromende vloeibare CO₂ af te koelen (1b).

10

11. De werkwijze volgens één van de conclusies 9-10, waarbij het koelen van het stromingskanaalelement (20) het uit de beperkte ruimte (30) wegleiden van het gasvormige CO₂ (1c) langs het stromingskanaalelement (20) omvat.

15 12. De werkwijze volgens één van de conclusies 9-11, waarbij de koelstadium het sproeien van het mengsel (2) over het in de beperkte ruimte (30) opgestelde monster (5) omvat, waarbij de werkwijze het handhaven van een druk in de beperkte ruimte (30) boven een druk van een tripelpuntdruk van CO₂ (1) en onder een druk van een kritisch punt van CO₂ (1) omvat.

20

13. De werkwijze volgens één van de conclusies 9-11, waarbij de koelstadium het verdichten van het deeltjesvormige vaste CO₂ (1a) in de beperkte ruimte (30) omvat, waarbij tijdens het sneeuwvormingsstadium en tijdens het verdichten van het deeltjesvormige vaste CO₂, (1a) een druk in de beperkte ruimte (30) wordt
25 gehandhaafd boven een druk van een tripelpuntdruk van CO₂ (1) en onder een druk van een kritisch punt van CO₂ (1), waarbij de werkwijze verder het verzamelen van het verdichte deeltjesvormige vaste CO₂ (1a) in een container, en het monster (5) in de container verschaffen om het monster (5) af te koelen met het verdichte deeltjesvormige vaste CO₂ (1a) omvat.

30

14. De werkwijze volgens conclusie 13, waarbij de werkwijze het beheersen van een druk in de beperkte ruimte (30) omvat terwijl het vloeibare CO₂ (1b) door

het stroomkanaalelement (21) stroomt, waarbij het deeltjesvormige vaste CO₂ (1a) wordt verdicht door een toenemende druk in de beperkte ruimte (30).

15. De werkwijze volgens één van de conclusies 9-14, waarbij het vloeibare CO₂ (1b) bij de elementinlaat (21) een verzadigde vloeistof is.

16. De werkwijze volgens één van de conclusies 9-15, waarbij het monster (5) een of meer van een biomedisch monster, een biologisch product en een medisch product omvat.

10

17. Een werkwijze voor het produceren van droogijs (1a), waarbij de werkwijze omvat:

- het stromen van vloeibaar CO₂ (1b) door een stroomkanaalelement (20) van een elementinlaat (21) naar een elementuitlaat (29) terwijl het stroomkanaalelement (20) wordt gekoeld, en
- het smoren van de stroom van het vloeibare CO₂ (1b) bij de elementuitlaat (29), waarbij het vloeibare CO₂ (1b) een mengsel (2) vormt dat een deeltjesvormig vast CO₂ (1a) en een gasvormig CO₂ (1c) omvat, waarbij het mengsel (2) vanaf de elementuitlaat (29) in een beperkte ruimte (30) sproeit;
- het verdichten van het deeltjesvormige vaste CO₂ (1a) in de beperkte ruimte (30);
- het verzamelen van het verdichte vaste CO₂ (1a) om het droogijs (1a) te verschaffen.

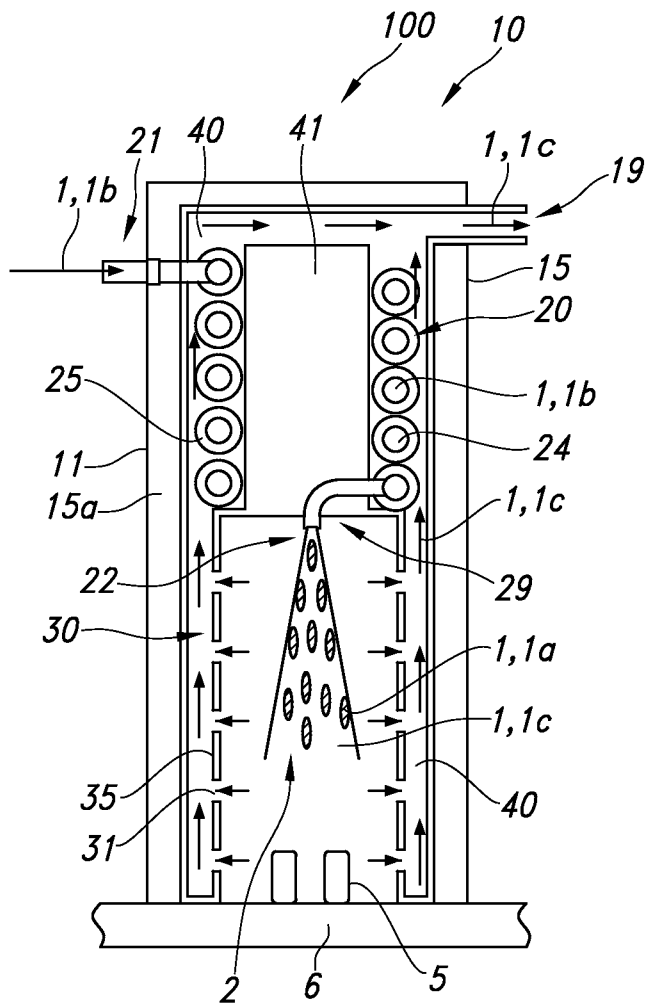


FIG. 1

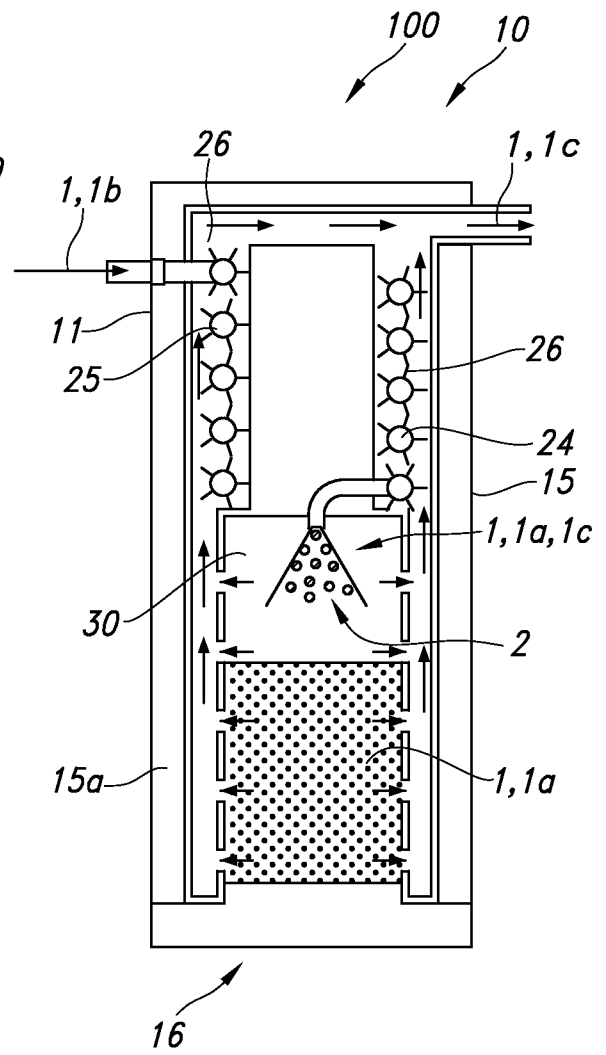
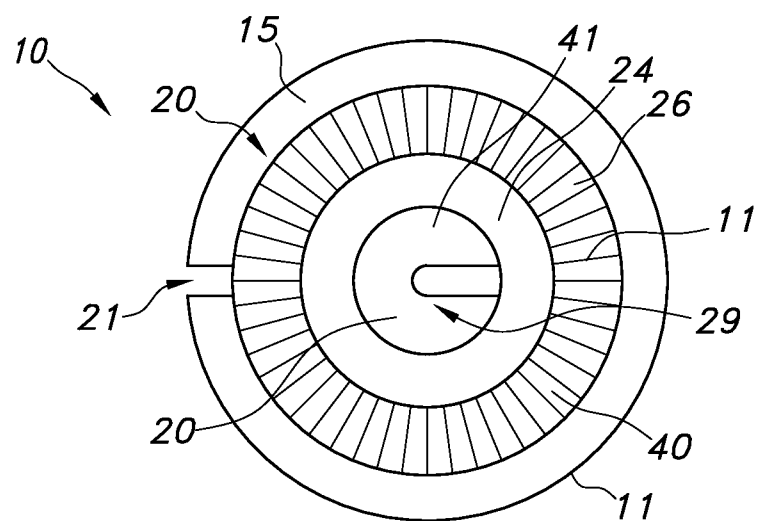
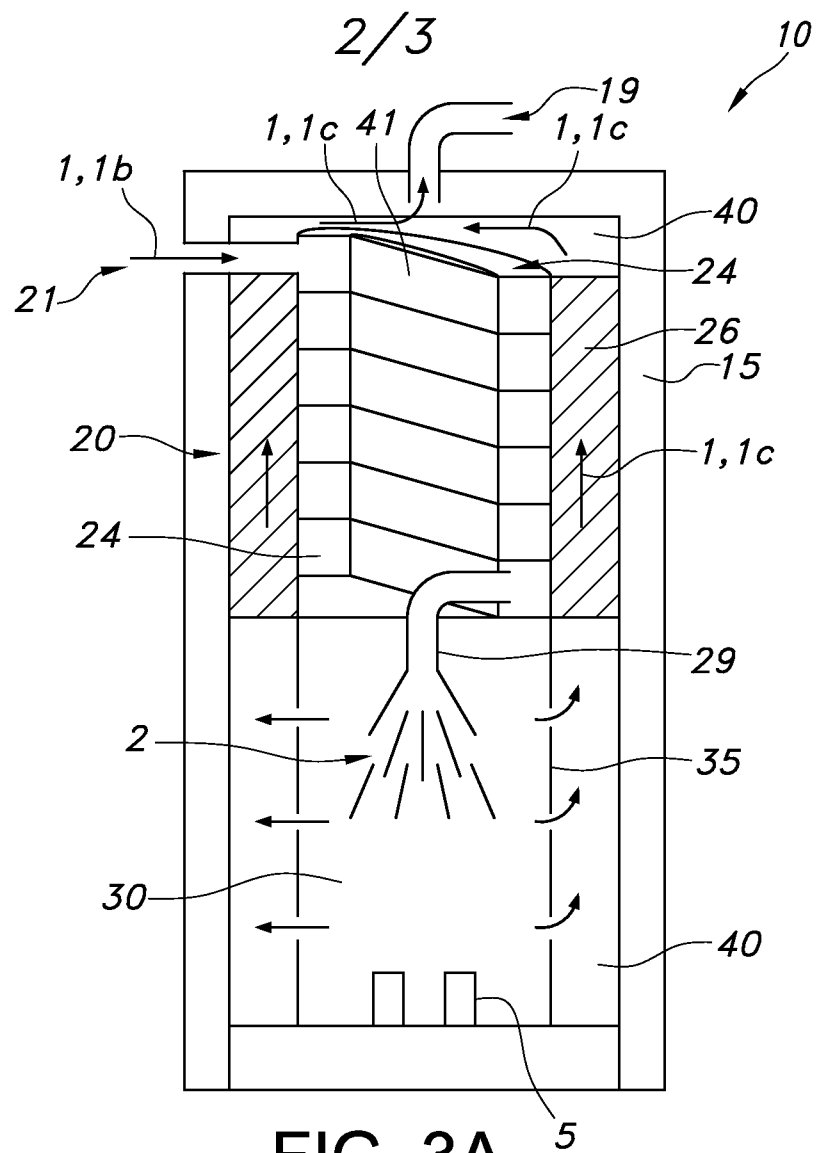


FIG. 2



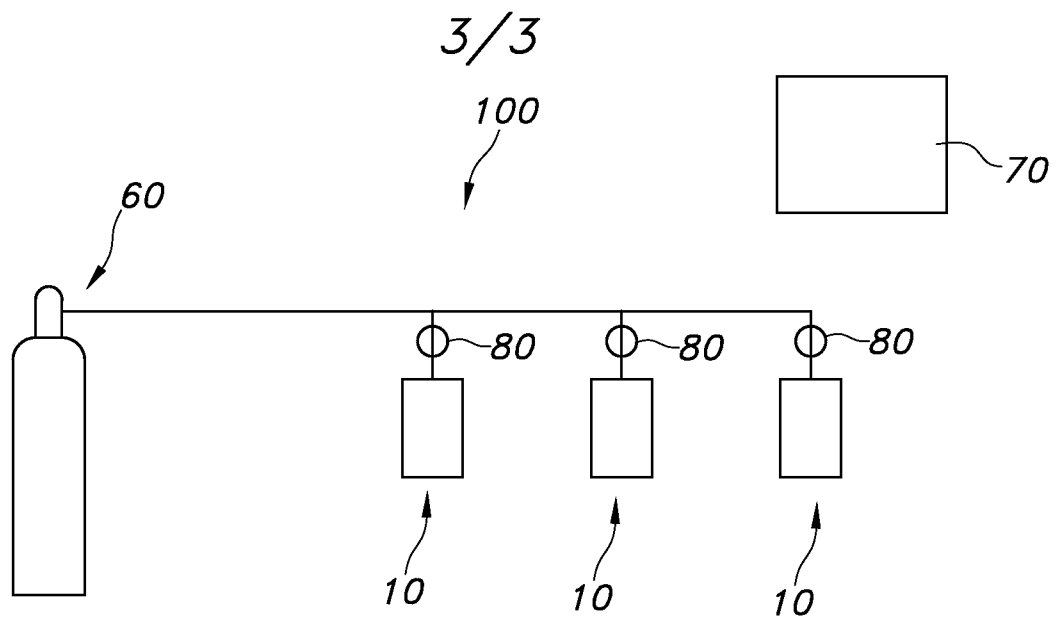


FIG. 4

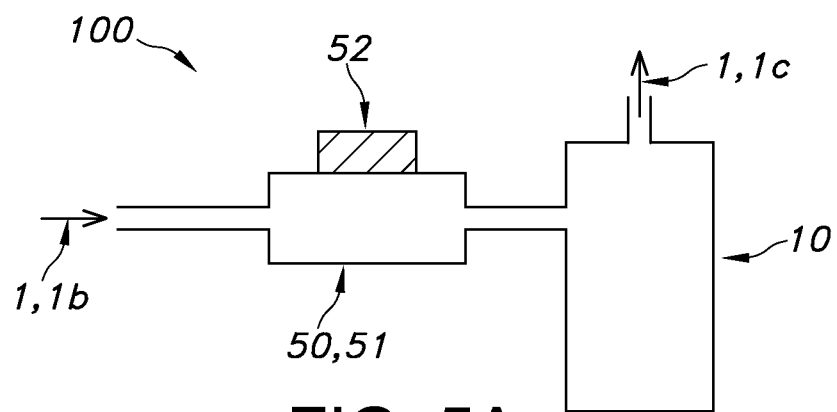


FIG. 5A

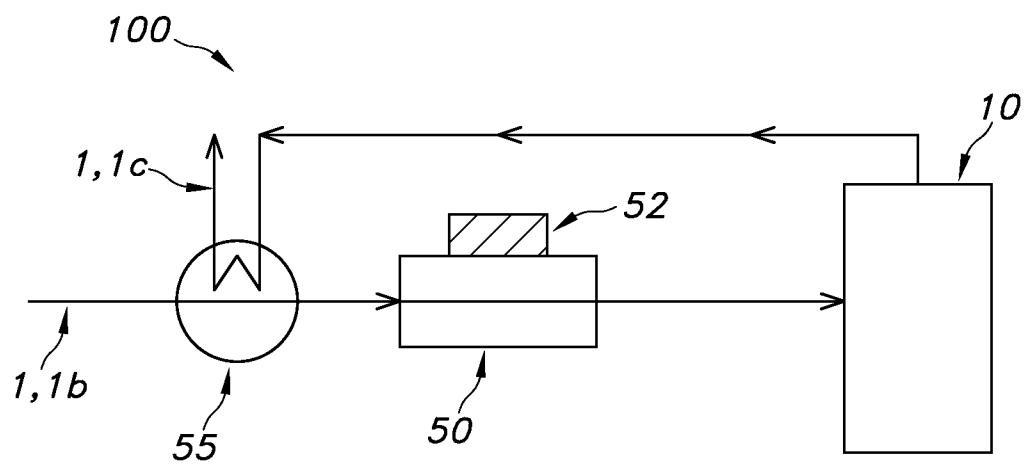


FIG. 5B

SAMENWERKINGSVERDRAG (PCT)

RAPPORT BETREFFENDE NIEUWHEIDSONDERZOEK VAN INTERNATIONAAL TYPE

IDENTIFICATIE VAN DE NATIONALE AANVRAGE		KENMERK VAN DE AANVRAGER OF VAN DE GEMACHTIGDE	
Nederlands aanvraag nr. 2034234		Indieningsdatum 28-02-2023	
		Ingeroepen voorrangsdatum	
Aanvrager (Naam) Universiteit Twente			
Datum van het verzoek voor een onderzoek van internationaal type 06-05-2023		Door de Instantie voor Internationaal Onderzoek aan het verzoek voor een onderzoek van internationaal type toegekend nr. SN83756	
I. CLASSIFICATIE VAN HET ONDERWERP (bij toepassing van verschillende classificaties, alle classificatiesymbolen opgeven)			
Volgens de internationale classificatie (IPC) Zie onderzoeksrapport			
II. ONDERZOCHE GEBIEDEN VAN DE TECHNIEK			
Onderzochte minimumdocumentatie			
Classificatiesysteem	Classificatiesymbolen		
IPC	Zie onderzoeksrapport		
Onderzochte andere documentatie dan de minimum documentatie, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen			
III.	GEEN ONDERZOEK MOGELIJK VOOR BEPAALDE CONCLUSIES		(opmerkingen op aanvullingsblad)
IV.	GEBREK AAN EENHEID VAN UITVINDING		(opmerkingen op aanvullingsblad)

**ONDERZOEKSRAPPORT BETREFFENDE HET
RESULTAAT VAN HET ONDERZOEK NAAR DE STAND
VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Nummer van het verzoek om een onderzoek naar
de stand van de techniek

NL 2034234

A. CLASSIFICATIE VAN HET ONDERWERP

INV. F25D3/12 C01B32/55

ADD.

Volgens de Internationale Classificatie van octrooien (IPC) of zowel volgens de nationale classificatie als volgens de IPC.

B. ONDERZOCHETE GEBIEDEN VAN DE TECHNIEK

Onderzochte minimum documentatie (classificatie gevolgd door classificatiesymbolen)

F25D C01B

Onderzochte andere documentatie dan de minimum documentatie, voor dergelijke documenten, voor zover dergelijke documenten in de onderzochte gebieden zijn opgenomen

Tijdens het onderzoek geraadpleegde elektronische gegevensbestanden (naam van de gegevensbestanden en, waar uitvoerbaar, gebruikte trefwoorden)

EPO-Internal

C. VAN BELANG GEACHTE DOCUMENTEN

Categorie °	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
X	US 2 071 302 A (REUBEN HILL) 16 februari 1937 (1937-02-16) * figuren 1-12 * -----	1-17
X	FR 709 021 A (GUGLIELMO WEBER) 1 augustus 1931 (1931-08-01) * het gehele document * -----	1-17
X	US 3 670 516 A (DURON PAUL P ET AL) 20 juni 1972 (1972-06-20) * figuren 1-3 * -----	1, 17
A	US 1 843 397 A (MARCUS DAVID A ET AL) 2 februari 1932 (1932-02-02) * het gehele document * ----- -/-	1, 9, 17



Verdere documenten worden vermeld in het vervolg van vak C.



Leden van dezelfde octroofamilie zijn vermeld in een bijlage

° Speciale categorieën van aangehaalde documenten

"A" niet tot de categorie X of Y behorende literatuur die de stand van de techniek beschrijft

"D" in de octrooiaanvraag vermeld

"E" eerdere octrooi(aanvraag), gepubliceerd op of na de indieningsdatum, waarin dezelfde uitvinding wordt beschreven

"L" om andere redenen vermelde literatuur

"O" niet-schriftelijke stand van de techniek

"P" tussen de voorrangsdatum en de indieningsdatum gepubliceerde literatuur

"T" na de indieningsdatum of de voorrangsdatum gepubliceerde literatuur die niet bezwarend is voor de octrooiaanvraag, maar wordt vermeld ter verheldering van de theorie of het principe dat ten grondslag ligt aan de uitvinding

"X" de conclusie wordt als niet nieuw of niet inventief beschouwd ten opzichte van deze literatuur

"Y" de conclusie wordt als niet inventief beschouwd ten opzichte van de combinatie van deze literatuur met andere geciteerde literatuur van dezelfde categorie, waarbij de combinatie voor de vakman voor de hand liggend wordt geacht

"&" lid van dezelfde octroofamilie of overeenkomstige octrooipublicatie

Datum waarop het onderzoek naar de stand van de techniek van internationaal type werd voltooid

6 september 2023

Verzenddatum van het rapport van het onderzoek naar de stand van de techniek van internationaal type

Naam en adres van de instantie

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040,
Fax: (+31-70) 340-3016

De bevoegde ambtenaar

de Graaf, Jan Douwe

**ONDERZOEKSRAPPORT BETREFFENDE HET
RESULTAAT VAN HET ONDERZOEK NAAR DE STAND
VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Nummer van het verzoek om een onderzoek naar
de stand van de techniek
NL 2034234

C.(Vervolg). VAN BELANG GEACHTE DOCUMENTEN		
Categorie °	Geciteerde documenten, eventueel met aanduiding van speciaal van belang zijnde passages	Van belang voor conclusie nr.
A	WO 2021/078891 A1 (MESSER GROUP GMBH [DE]) 29 april 2021 (2021-04-29) * het gehele document * -----	1-17
A	US 6 761 043 B1 (REZNIKOV LEV [US] ET AL) 13 juli 2004 (2004-07-13) * figuur 9 * -----	9

**ONDERZOEKSRAPPORT BETREFFENDE HET
RESULTAAT VAN HET ONDERZOEK NAAR DE STAND
VAN DE TECHNIEK VAN HET INTERNATIONALE TYPE**

Informatie over leden van dezelfde octrooifamilie

Nummer van het verzoek om een onderzoek naar
de stand van de techniek

NL 2034234

In het rapport genoemd octrooigeschrift	Datum van publicatie	Overeenkomend(e) geschrift(en)	Datum van publicatie
US 2071302	A	16-02-1937	GEEN
FR 709021	A	01-08-1931	GEEN
US 3670516	A	20-06-1972	CA 950615 A 09-07-1974
		DE 2106477 A1 19-08-1971	
		FR 2078442 A5 05-11-1971	
		GB 1301783 A 04-01-1973	
		JP S5384643 U 12-07-1978	
		JP S5436126 Y2 01-11-1979	
		US 3670516 A 20-06-1972	
		ZA 71749 B 27-09-1972	
US 1843397	A	02-02-1932	GEEN
WO 2021078891	A1	29-04-2021	BR 112022007614 A2 19-07-2022
		CA 3155583 A1 29-04-2021	
		CN 114599606 A 07-06-2022	
		DE 102019007439 A1 29-04-2021	
		EP 4048633 A1 31-08-2022	
		US 2022379276 A1 01-12-2022	
		WO 2021078891 A1 29-04-2021	
US 6761043	B1	13-07-2004	GEEN

WRITTEN OPINION

File No. SN83756	Filing date (<i>day/month/year</i>) 28.02.2023	Priority date (<i>day/month/year</i>)	Application No. NL2034234
International Patent Classification (IPC) INV. F25D3/12 C01B32/55			
Applicant Universiteit Twente			

This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☐ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the application
- ☐ Box No. VIII Certain observations on the application

	Examiner de Graaf, Jan Douwe
--	---------------------------------

WRITTEN OPINION

Application number

NL2034234

Box No. I Basis of this opinion

1. This opinion has been established on the basis of the latest set of claims filed before the start of the search.
2. With regard to any **nucleotide and/or amino acid sequence** disclosed in the application, this opinion has been established on the basis of a sequence listing:
 - a. ☐ forming part of the application as filed.
 - b. ☐ furnished subsequent to the filing date for the purposes of search,
 - ☐ accompanied by a statement to the effect that the sequence listing does not go beyond the disclosure in the application as filed.
3. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the application, this opinion has been established to the extent that a meaningful opinion could be formed without a WIPO Standard ST.26 compliant sequence listing.
4. Additional comments:

Box No. V Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty	Yes: Claims	6, 7, 9-16
	No: Claims	1-5, 8, 17
Inventive step	Yes: Claims	
	No: Claims	1-17
Industrial applicability	Yes: Claims	1-17
	No: Claims	

2. Citations and explanations

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1) Reference is made to the following documents:

D1) US 2 071 302 A (REUBEN HILL) 16 februari 1937 (1937-02-16)

D2) FR 709 021 A (GUGLIELMO WEBER) 1 augustus 1931 (1931-08-01)

D3) US 3 670 516 A (DURON PAUL P ET AL) 20 juni 1972 (1972-06-20)

D4) US 6 761 043 B1 (REZNIKOV LEV [US] ET AL) 13 juli 2004 (2004-07-13)

2) The present application does not meet the criteria of patentability, because the subject-matter of claim 1 is not new.

Document D1 discloses (the numbers in brackets are referring to D1): "

Een systeem (fig. 1), waarbij het systeem een inrichting omvat, de inrichting omvattende (i) een inrichtingswand (20,21) met een inrichtingsopening (97,98), een (ii) stromingskanaalelement (99) omvattende een elementinlaat (97,98) en een elementuitlaat (100,44), waarbij het stromingskanaalelement (pre-cooling coil 99) ten minste gedeeltelijk warmtegeleidend is, (iii) een begrenziingswand (upper mold body 34,35,36,37, lower mold body inc. screen 39) voor het definiëren van een beperkte ruimte (mold cavity), en (iv) een kanaalruimte (23), waarin

- de begrenziingswand (34,35,36,39) een aantal openingen omvat om een gasvormig fluïdum de beperkte ruimte (mold cavity) te laten verlaten,
- de inrichtingswand (20,21) en de begrenziingswand (34,35,36,37,39) de kanaalruimte (23) definiëren, waarbij de kanaalruimte (23) de beperkte ruimte (mold cavity) met de inrichtingsopening (97,98) via het aantal openingen fluïde verbindt en waarbij het stromingskanaalelement (99) thermisch gekoppeld met de kanaalruimte (23) is geconfigureerd,
- waarbij de elementinlaat (97,98) is uitgevoerd aan een buitenzijde van de inrichting en de elementuitlaat (103) is uitgevoerd in open fluïdumverbinding met de beperkte ruimte (mold cavity), en
- de elementuitlaat (44) een smoorelement (jet 44) omvat."

Accordingly, the features of claim 1 are disclosed in D1, the subject matter of claim 1 is therefor deemed **not new**.

Documents D2 and D3 also disclose the features of claim 1.

Each of documents D1 - D3 further disclose the features of corresponding method claim 17, which is therefor also considered not new. Producing dry ice using a cooled (such as in a countercurrent flow) liquid CO₂ supply is well known in the art.

3) The present application does not meet the criteria of patentability, because the subject-matter of independent method claim 9 does not involve an inventive step.

Document D1 is regarded as being the prior art closest to the subject-matter of claim 9 , and discloses all the method features disclosed in claim 9, except for :

"stap (i) het sproeien van het mengsel over een monster gerangschikt in de beperkte ruimte en het koelen van het monster met het verdichte deeltjesvormige vaste CO₂", and is therefor deemed new.

The problem to be solved by the present invention may therefore be regarded as to improve the fast cooling of a sample by installing the sample inside the dry ice pellet maker mold cavity and cooling the sample accordingly during dry ice making.

The solution proposed in claim 9 of the present application cannot be considered as involving an inventive step for the following reasons: To add the sample inside the dry ice making cavity (either in D1, or in D2) is merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

It is well known in the prior art to directly add produced dry ice upon or around a product to be cooled (see e.g. document D4, fig. 9).

4) Dependent apparatus claims 2-8 do not appear to contain any additional features which, in combination with the features of any claim to which they refer, meet the requirements of novelty (claims 2-5 and 8) and/or inventive step (claims 6,7), the reasons being as follows:

Documents D1 and D2 both disclose the additional features of claims 2 - 5 and 8 (numbers apply to D1), claims 2-5 and 8 are thus deemed not new:

Claim 2: " ... waarbij ten minste een deel van het stromingskanaalelement (99) is aangebracht in de kanaalruimte (23).

Claim 3: "... waarbij het stromingskanaalelement (99) een of meer warmtegeleidende uitbreidingen (item 99 is made coiled inside space 23) omvat die zich uitstrekken vanaf het stromingskanaalelement (99)."

Claim 4: " waarbij het smoorelement (jet 44) een capillair stromingskanaalelement omvat."

Claim 5: " ...waarbij het stromingskanaalelement (99) een stromingskanaal (99) omvat dat de elementinlaat (97) op fluïde wijze verbindt met de elementuitlaat (103), waarbij een totale lengte van het stroomkanaal (99) wordt gekozen uit het bereik van 0,1-5 meter.

Claim 8: " ... waarbij de inrichting verder een verwijderbaar sluitelement (31) omvat voor het afsluiten van de beperkte ruimte (mold cavity)."

The features of dependent claim 6 and 7 are deemed merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

Choosing e.g. a metal for a coil (as in claim 6) is simply not inventive in this set-up, there is no surprising effect to be foreseen.

Dependent method claims 10-16 do not appear to contain any additional features which, in combination with the features of any claim to which they refer, meet the requirements of inventive step, the reasons being as follows:

Claims 10-15: The features mentioned in claims 10-15 are merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

Claim 16: Taking a medical sample (or a sample, or a product) and cooling this inside the dry ice maker cannot be regarded as an inventive activity (refer to for example D6, fig. 9), as the skilled person would select this, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed.

5) The relevant background art disclosed in D1, D2 and D6 is not mentioned in the description, nor are these documents identified therein.

6) Independent claims 1 ,9 and 17 are not in the two-part form, which in the present case would be appropriate, with those features known in combination from the prior art being placed in the preamble and the remaining features being included in the characterising part.