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Method for Flow Rate Control of Clustering Fluid and Device for Flow Rate Control of Clustering Fluid Employed in the Method

Field of the Invention

5 The present invention is utilized mainly in the fields of the production of pharmaceuticals, nuclear fuels, chemicals, semiconductors and the like, and is concerned with the method for the fluid flow rate control which can control the flow rate of a fluid as supplied such as hydrofluoric gas or hydrogen fluoride (hereinafter called the HF gas), ozone, water and the like (hereinafter called a
10 clustering fluid) with high precision using a flow rate control device. The present invention is also concerned with a flow rate control device employed in the method for the flow rate control of the clustering fluid.

Background of the Invention

15 The clustering fluid such as the HF gas and the like has been widely utilized in the fields of the production of chemicals, semiconductors and the like. Accordingly, a wide variety of flow rate control devices are used in the facilities to supply the clustering fluid such as the HF gas and the like. Among others, a mass flow controller, a pressure difference type flow rate control device and the like are the ones comparatively widely utilized.

20 The clustering fluid such as the HF gas and the like is known as a substance having peculiar properties such as high density, high specific heat ratio, high dependence of the molecular association on the temperature and pressure, high dependence of an endothermic amount on the temperature at the time of the molecular dissociation, and the like.



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Therefore, Boyle's and Charles' law does not apply to the clustering fluid unlike ideal gases like the oxygen gas or the nitrogen gas, and in regard with the flow control there exist many problems which are not encountered when controlling the flow rate control of the oxygen gas or the nitrogen gas. This is
5 because the molecular weight of the clustering fluid such as the HF gas and the like greatly changes depending on the pressure fluctuations and others due to dependence of the molecular association on the temperature and pressure, and even if the clustering fluid such as the HF gas and the like has the same weight, the number of mole of the clustering fluid changes when the molecular weight M
10 changes.

Because the association heat of the molecules of the clustering fluid such as the HF gas and the like is great, the bigger the pressure difference is when it is supplied, the larger endothermic effects result. In the event that the endothermic effects are remarkable, a problem such as liquefaction will occur
15 because of the low steam pressure.

Hence, with the conventional supply systems for the clustering fluid such as the HF gas and the like, it has been tried to improve precision of the flow rate control precision of the clustering fluid such as the HF gas and the like by mixing the clustering fluid in a large amount of the diluting gas and controlling the
20 diluting gas so that a high precision flow rate control of the clustering fluid such as the HF gas and the like can be achieved. Or, a heating device is installed at the supply source of the clustering fluid such as the HF gas and the like so that the clustering fluid such as the HF gas and the like is supplied under a relatively high pressure and in a state in which the clustering of the fluid is made difficult by

heating the fluid at a high temperature.

Description of Prior Art

However, if a large amount of the diluting gas is used, or the clustering fluid heated to a high temperature is supplied under high pressure, its cost effectiveness and controllability may be hindered. Furthermore, in the event that the process side, for instance the semiconductor manufacturing facilities, to which the clustering fluid is supplied, is low-pressurized to $10^{-2} \sim 10^2$ Torr forming a near-vacuum, the high precision flow rate control can be never achieved with the conventional mass flow controller due to various difficulties caused in the practical use.

On the other hand, in the field of semiconductor manufacturing in recent years, the pressure type flow rate control device is widely used for the flow rate control of the gases as supplied, replacing the conventional mass flow rate controller in which the flow rate is basically controlled by detecting the transfer of heat quantity. With the said pressure type flow rate control device, in the event that the fluid passing through an orifice is under the so-called critical condition, the flow rate of the fluid Q can be obtained from the formula, $Q=KP_1$ (where K represents a constant determined by a nozzle, and P_1 represents the gas pressure on the upstream side of the nozzle). In the semiconductor manufacturing facilities and the like where the pressure on the downstream side of the nozzle (i.e., the process side to which the gas is supplied) is generally low-pressurized to almost a vacuum, use of the pressure type flow rate control device is favored from the view point of maintaining the afore-mentioned critical condition.

However, with the pressure type flow rate control device in which the gas flow rate is computed on the basis that the gas to be controlled passes through an orifice under the so-called critical condition holding the ratio P_2/P_1 between the pressure P_1 on the upstream side of the orifice and the pressure P_2 on the downstream side of the orifice at a value less than about 0.5, the degree of the molecular association of the cluster-making fluid such as the HF gas and the like changes due to fluctuation of the temperature caused by the endothermic action at the time of the dissociation of molecules, or due to fluctuation of the pressure on the downstream side of the orifice (or the process side). If, as a result, the critical condition of the clustering fluid such as the HF gas and the like is not satisfied, the high precision flow rate control of the clustering fluid such as the HF gas and the like becomes basically impossible, thus causing various difficulties.

For example, if the ratio Q_{HF}/Q_N (the flow factor F.F.) is obtained in advance from the actual measurement of the flow rates where the flow rate Q_N (SCCM) is the flow rate of the N_2 gas and the flow rate Q_{HF} is the flow rate of the clustering fluid, and N_2 and the clustering fluid are passed to the same pressure type flow rate control device under the same conditions, the actual flow rate Q_{HF} of the clustering fluid such as the HF gas and the like which is passed to the pressure type flow rate control device graduated for the N_2 gas can be determined by multiplying the flow rate value indicated when the clustering fluid is passed to the said pressure type flow rate control device by the afore-mentioned flow factor value F.F. In the event that the flow factor F.F. of the clustering fluid such as the HF gas and the like is always held at a certain

value, the flow rate control of the clustering fluid such as the HF gas and the like can be effected with a comparatively high precision using the pressure type flow rate control device.

On the other hand, the flow rate factor F.F. of the afore-mentioned various
 5 gases can be computed by the theoretical formula, and normally it can be computed by the following formula:

[Formula 1]

$$F.F. = (K / r_s) \{ 2 / (\kappa + 1) \}^{1/(\kappa - 1)} [\kappa / \{ (\kappa + 1) \cdot R \}]^{1/2}$$

where r_s represents density of the gas in the normal state, κ a specific
 10 heat ratio of gas, R a gas constant, and K a proportional constant not depending on the type of gas (TOKU-KAI No. 2000-322130).

The flow factor F.F. of the HF gas at the room temperature can be obtained by the afore-mentioned theoretical formula as follows:

$$F.F. = 0.440100$$

15 On the contrary, when the flow factor F.F. is actually measured by using the flow rate test device in Figure 1 to be explained below, it is found that the value is as follows:

Temperature actually measured	45°C
Flow factor F.F. actually measured	1,1665

The temperature is that of the heated body part of the pressure type flow
 20 rate control device.

As apparent from the afore-mentioned flow factor value, due to the peculiar

properties of the HF gas which permit the HF gas to form a state of molecules as bound together depending on the conditions of pressure and temperature, the flow factor value of the HF gas, as actually measured, is vastly different from that determined by the theoretical formula. It is, thus, found that the concept that
5 applies to ordinary gases such as N₂ gas, O₂ gas and the like will not be applicable to the flow rate control of the HF gas when the pressure type flow rate control device is in use.

The above explanation was given with respect to the case where the pressure type flow rate control device is employed. However, it should be
10 noted that the explanation can also apply to the cases where the heat type flow rate control device such as a mass flow controller, or the flow rate control device using a flow rate control valve is employed. In these cases too, the concept applicable to the ordinary gases such as the O₂ gas, N₂ gas and the like cannot apply to the flow rate control of the cluster fluid such as the HF gas and the like.

15 Object of the Invention

It is an object of the present invention to solve such problems as encountered when the flow rate control device is used for controlling the flow rate of the clustering fluid such as the HF gas and the like, that is, the problem that the conventional flow rate control device can not achieve a high precision
20 flow rate control of the clustering fluid such as the HF gas and the like to be supplied to a low pressurized chamber due to the changes in the molecular weight and the number of mole caused by the molecular association and the like, the changes in the temperature caused by the endothermic action at the time of dissociation of molecules, and the changes in the degree of the molecular

association caused by the fluctuations of the pressure and temperature. It is also an object of the present invention to provide a method for the flow rate control of the clustering fluid such as the HF gas and the like in which the temperature of a main part of the flow rate control device is maintained at 40°C ~ 85°C, more preferably higher than 65°C, or the clustering fluid is added with a dilute gas to keep the fluid not higher than a partial pressure by making use of the fact that the clustering fluid has not only temperature dependency but also pressure dependency so that the association of the clustering fluid such as the HF gas and the like is dissociated to keep the clustering fluid in a theoretically monomolecular state (or a state of a small number of cluster molecules) with the clustering fluid being permitted to pass through the flow rate control device, thereby making it possible to control the flow rate of the clustering fluid such as the HF gas and the like with a high precision and to supply the clustering fluid to a low pressurized chamber. It is also an object of the present invention to provide a device for the flow rate control of the clustering fluid to be used in the above mentioned method.

Disclosure of the Invention

The inventors of the present invention first conceived an idea of using the pressure type flow rate control device and heating a major part of the pressure type flow rate control device so that the flow rate control device can supply the clustering fluid with the flow rate of 3~300SCCM to a low pressurized process chamber used in the semiconductor manufacturing facilities while subjecting the fluid to a high precision flow rate control. And, then, based on the afore-mentioned idea, tests were conducted to determine the relationship

between the heating temperature and flow rate control characteristics (flow rate control precision) with respect to a plural number of set flow rates (75%, 60%, 45%, 30% and 15%) of the pressure type flow rate control device.

The present invention has been created based on the afore-mentioned test
5 results. The present invention in Claim 1 relates to a method for supplying a clustering fluid from a gas supply source to desired instruments or apparatuses by using a flow rate control device in which the association of molecules of the clustering fluid passing through the said flow rate control device is dissociated by heating the afore-mentioned flow rate control device so that the clustering fluid
10 kept in a monomolecular state is supplied by the flow rate control device while subjected to the flow rate control.

The present invention in Claim 2 relates to a method for supplying a clustering fluid from a gas supply source to desired instruments or apparatuses by using a flow rate control device in which the association of molecules of the
15 clustering fluid passing through the afore-mentioned flow rate control device is dissociated by diluting the afore-mentioned clustering fluid to a level lower than a partial pressure so that the clustering fluid kept in a monomolecular state is supplied by the flow rate control device while subjected to the flow rate control.

The present invention in Claim 3 relates to a method as claimed in Claim 1
20 or Claim 2 wherein the flow rate control device is a flow rate control valve, a heat type flow rate control device or a pressure type flow rate control device.

The present invention in Claim 4 relates to a method as claimed in Claim 1 wherein the temperature for heating of a main body of the flow rate control device is not lower than 40°C.

The present invention in Claim 5 relates to a method for the flow rate control of a clustering fluid employing a pressure type flow rate control device in which the flow rate Q of the gas passing through an orifice is computed as $Q=KP_1$ (where K is a constant) with the gas passing through the orifice in a state
5 where the ratio P_2/P_1 between the gas pressure P_1 on the upstream side of the orifice and the gas pressure P_2 on the downstream side of the orifice is held at a value lower than the critical pressure ratio of the gas wherein the clustering fluid is permitted to pass through the afore-mentioned orifice while the afore-mentioned pressure type flow rate control device is heated to the
10 temperature not lower than 40°C .

The present invention in Claim 6 relates to a method as claimed in Claim 5 wherein a chamber which is connected to the downstream side of the pressure type flow rate control device and receives the clustering fluid is a vacuum device.

The present invention in Claim 7 relates to a method as claimed in Claim 5
15 wherein the graduation of the pressure type flow rate control device is determined based on the N_2 gas, and the flow rate factor F.F. of the clustering fluid against the N_2 gas is appropriately chosen according to the temperature of the main body of the pressure type flow rate control device so that the flow rate of the clustering fluid is obtained by multiplying the measured flow rate value of
20 the passing clustering fluid by the afore-mentioned flow factor F.F.

The present invention in Claim 8 relates to a method as claimed in Claim 6 wherein the pressure type flow rate control device is heated to the temperature of $40^{\circ}\text{C} \sim 85^{\circ}\text{C}$, and the vacuum device which is connected to the downstream side of the pressure type flow rate control device and receives the clustering

fluid is a vacuum chamber whose pressure is 10^{-3} Torr $\sim 10^2$ Torr.

The present invention in Claim 9 relates to a method as claimed in Claim 5 wherein the range of the flow rate control of clustering fluid to be supplied from the pressure type flow rate control device to the vacuum chamber is
5 3~300SCCM.

The present invention in Claim 10 relates to a method as claimed in Claim 1 to Claim 9 wherein the clustering fluid is hydrogen fluoride gas, water or ozone.

The present invention in Claim 11 relates to a method as claimed in Claim 7 wherein the temperature of a main body of the pressure type flow rate control
10 device is 45°C while the flow factor is F.F. is 1.1665 when hydrogen fluoride is in use for the fluid.

The present invention in Claim 12 relates to a flow rate control device for a clustering fluid which supplies the clustering fluid from a fluid supply source to desired instruments or apparatuses in which the association of molecules of the
15 clustering fluid passing through the said flow rate control device is dissociated by heating a main body of the afore-mentioned flow rate control device so that the flow rate control device is permitted to control the flow rate of the clustering fluid kept in a monomolecular state.

The present invention in Claim 13 relates to a flow rate control device for a
20 clustering fluid which supplies the clustering fluid from a fluid supply source to desired instruments or apparatuses in which the association of molecules of the clustering fluid passing through the said flow rate control device is dissociated by diluting the afore-mentioned clustering fluid to a level lower than a partial pressure so that the flow rate control device is permitted to control the flow rate

of the clustering fluid kept in a monomolecular state.

The present invention in Claim 14 relates to a flow rate control device as claimed in Claim 12 or Claim 13 wherein any one of a flow rate control valve, a heat type flow rate control device or a pressure type flow rate control device is
5 chosen for the flow rate control device.

The present invention in Claim 15 relates to a flow rate control device as claimed in Claim 12 wherein a main body of the flow rate control device is heated to the temperature not lower than 40°C.

The present invention in Claim 16 relates to a pressure type flow rate
10 control device in which the flow rate Q of the gas passing through an orifice is computed as $Q=KP_1$ (where K is a constant) with the gas passing through the orifice in a state where the ratio P_2/P_1 between the gas pressure P_1 on the upstream side of the orifice and the gas pressure P_2 on the downstream side of the orifice is held at a value not higher than the critical pressure ratio of the gas
15 wherein the fluid to be controlled is the clustering fluid, and a heating device is installed in the pressure type flow rate control device so that the heating device is permitted to heat a main body of the pressure type flow rate control device to the temperature not lower than 40°C.

The present invention in Claim 17 relates to a pressure type flow rate
20 control device as claimed in Claim 16 wherein the clustering fluid flow factor F.F. based on the N₂ gas is appropriately chosen according to the temperature of the main body of the pressure type flow rate control device, and the graduation of the pressure type flow rate control device is determined using the said chosen flow factor F.F. value.

The present invention in Claim 18 relates to a flow rate control device as claimed in Claim 13 to Claim 17 inclusive wherein the clustering fluid is hydrogen fluoride gas, water or ozone.

5 The present invention in Claim 19 relates to a pressure type flow rate control device as claimed in Claim 17 wherein the temperature of the main body of the pressure type flow rate control device is 45°C while the flow factor F.F. is 1.1665 when the hydrogen fluoride gas is in use for the fluid.

Brief Description of the Drawings

10 Figure 1 is a whole block diagram of the test device to find various characteristics of the pressure type flow rate control device with regard to the clustering fluid such as the HF gas and the like.

Figure 2 illustrates the flow rate characteristics of the pressure type flow rate control device, (a) showing the case where the test specimen F115A is used for the device, (b) the case where the test specimen F65A is used, and (c) the case where the test specimen F20A is used.

Figure 3 illustrates the flow rate errors caused by changes of the heating temperature ($45^{\circ}\text{C} \pm 2^{\circ}\text{C}$ and $85^{\circ}\text{C} \pm 2^{\circ}\text{C}$) in the cases where the test specimens are F115A and F115A-HT types.

20 Figure 4 is a diagram to show the flow rate as measured employing the test specimens, F115A, F65A and F20A where the heating condition is a parameter.

Figure 5 is a explanatory drawing of the method for the flow rate control of the clustering fluid in which the clustering fluid such as the HF gas and the like is permitted to be lower than a partial pressure by dilution.

List of Reference Letters and Numerals

1 designates a N2 gas supply line

2 a supply line for the clustering fluid such as the HF gas and the like

3 a pressure type flow rate control device (a test specimen)

4 a build-up chamber

5 5 a vacuum pump

6 a heating device

P a pressure sensor

T a temperature sensor

S a build-up flow rate measurement device

10 7 a diluting gas supply line

8 a dilution apparatus

9 a pressure type flow rate control device (non-heating type)

10 an apparatus using the clustering fluid such as the HF gas and the like

Mode of Carrying Out the Invention

15 The mode of carrying out the present invention is explained hereunder.

Figure 1 illustrates a whole construction of a test device to find various characteristics of the pressure type flow rate control device, which forms a basis of the present invention, with regard to the clustering fluid such as the HF gas and the like. Specifically, the following tests ①~⑦ were implemented using
20 the said test device.

① The flow rate control characteristics shown when the HF gas is passed at the flow rate of 4 -100SCCM are confirmed using 2 sets of the pressure type flow rate control device (for the N2 gas).

② There are confirmed the state of the flow rate control in which the build-up

chamber is heated and the state of the flow rate control in which the build-up chamber is not heated.

③ The flow rate control error shown when the HF gas is passed at the same flow rate to the flow rate control devices (for the N2 gas) having different flow rate
5 ranges is confirmed.

④ A linearity of the flow rate control shown when the flow rate of the HF gas is controlled (at the time of the intermediate setting measurement and at the time when the measurement is conducted by the build-up flow rate measurement device) is confirmed.

10 ⑤ Flow rate control precision shown when the temperature for heating the pressure type flow rate control device (for the N2 gas) is changed ($45^{\circ}\text{C} \pm 2$) is confirmed.

⑥ The flow rate shown when the heating temperature is raised to a high temperature (85°C) is confirmed using the pressure type flow rate control device
15 for high temperature.

⑦ Flow rate precision shown when the heating temperature is changed to a high temperature ($85^{\circ}\text{C} \pm 2^{\circ}\text{C}$) is confirmed using the pressure type flow rate control device for high temperature.

On the basis of the results of the afore-mentioned tests ①~⑦, an optimum
20 range of the heating condition is determined examining the relationship between the flow rate measurement precision and temperature conditions where the flow rate of the HF gas is controlled using the pressure type flow rate control device.

Referring to Figure 1, 1 designates a N2 gas supply line, 2 a HF gas supply line, 3 a pressure type flow rate control device (a test specimen), 4 a build-up

chamber, 5 a vacuum pump, 6 a heating device, P a pressure sensor, T a temperature sensor and V1~V6 valves. The pressure type flow rate control device 3 and a build-up chamber 4 are temperature-adjusted by the heating device (heater) 6.

5 The N₂ gas of 201kPa abs. and the HF gas are supplied to the afore-mentioned N₂ gas supply line 1 and the HF gas supply line 2 respectively.

Furthermore, a capacitance manometer (rated as 13.3kPa abs.) is used for the pressure sensor P, and a resistance thermometer (Pt100) is used for the temperature sensor T.

10 K.K. FUJIKIN-made FCS-4WS-F115A (115SCCMF.S.), FCS-4WS-F65A (65SCCMF.S.), FCS-4WS-F20A (20SCCMF.S.) and FCS-4JR-124-F115A-HT (115SCCMF.S. for high temperature) are used for the pressure type flow rate control device 3 which is the afore-mentioned test specimen. The build-up chamber 4 has an inside volume of 1000cc. And, an EDWARDS INC.-made
15 vacuum pump is used for the vacuum pump 5.

A so-called build-up flow rate measurement device S is formed of the pressure type flow rate control device 3, the build-up chamber 4, the pressure gauge P and valves V1~V5 and the like.

The pressure type flow rate control device 3 and the build-up chamber 4
20 are temperature-adjusted by the heat adjuster 6 (K.K. REKEN-KEIKI-made CB100) at a prescribed temperature determined by the heating conditions, and the temperature is measured at some parts (totaling 8 points) using the sheath type K thermocouple as mentioned later.

The heating conditions in the tests as will be mentioned below are follows

with respect to the pressure type flow rate control device 3 and the build-up chamber 4:

Test Specimen 3	Temperature	Build-up Chamber 4	Temperature
FCS(F115A・F65A・F20A)	45°C	Downstream side chamber	Rm Temp. 40°C
FCS(F115A)	43°C、47°C	Downstream side chamber	Rm Temp. 40°C
FCS(F115A-HT)	85°C	Downstream side chamber	40°C
FCS(F115A-HT)	83°C、87°C	Downstream side chamber	40°C

Each test specimen 3 has 5 measurement points (6 measurement points
5 only for F20A), which are the points of 75%, 60%, 45%, 30%, 15%, (90% only for F20A) of the respective rated set values. The set value (%) means the setting against the input signal 0-100% and the input voltage signal 0-5V to the pressure type flow rate control device 3.

Furthermore, when the measurement is conducted at the same flow rate
10 point, 3 points of 48.8SCCM, 20.0SCCM and 9.75SCCM (based on N2) are measured respectively.

The places for measuring the afore-mentioned temperature are as listed in Table 1. Point A designates a temperature monitor display of the leak port part of the main body of the pressure type flow rate control device, B a thermistor fitting
15 part of the main body of the pressure type flow rate control device, Point C a lower part of the base plate of the computing control part, Point D the room temperature in the test room, Point E a temperature monitor display of the

build-up chamber, Point F is a temperature displayed on the temperature adjuster 6 mounted on the build-up chamber, and Point G is a temperature displayed on the temperature adjuster 6 mounted on the pressure type flow rate control device.

5 The afore-mentioned tests ①~⑦ were conducted in the following order.

① The pressure type flow rate control device 3 which is the test specimen is connected as illustrated in Figure 1 (i.e., connected to the build-up flow rate measurement device S).

② The warming-up operation of the pressure type flow rate control device
10 3 and other measurement instruments is effected for more than 1 hour before the test is started.

③ Atmospheric gas is evacuated to a vacuum state up to the upstream side of the pressure type flow rate control device 3 by the vacuum pump 5.

④ A cycle purge using the N₂ gas is conducted for a gas substitution.

15 ⑤ Valves placed forwardly and backwardly of the pressure type flow rate control device 3 are closed while the system is kept in the vacuum state. The system is left for 10 minutes as it is, in order to confirm that there is no rise in the pressure command output (no leak is confirmed).

⑥ Next, the gas (the HF gas or the N₂ gas) to be measured is allowed to
20 pass to the pressure type flow rate control device 3, to measure the flow rate by the build-up method. The measurement is conducted 3 times to determine the average of the measurements, which will be the measurement result. It should be noted that the pressure inside the build-up chamber 4 is evacuated to a vacuum degree of 10^2 Torr before the start of the measurement.

⑦ A special heater is used for heating the pressure type flow rate control device 3.

⑧ Ample time should be given for the temperature adjustment, and the measurement should start after confirming that the temperature has been stabilized.

The results of the temperature measurement are as shown in Table 1 ~ Table 3. It should be noted that, in the Tables, the heating conditions ① are that the temperature of the test specimen 3 is 45°C and the build-up chamber is not heated while the heating conditions ② are that the temperature of the test specimen 3 is 45°C and the temperature of the build-up chamber is 40°C.

[Result of Temperature Measurement]

[Table 1] Test Specimen F115A

[°C]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
A. Downstream side leak port temp monitor	46.0	45.4	45.7	45.2
B. Thermistor part	47.2	46.6	46.9	46.4
C. Bottom part of substrate	43.1	42.8	43.1	42.5
D. Room temperature	24.5	25.3	24.5	24.8
E. Chamber temp. monitor	23.7	24.4	44.4	43.0
F. Chamber temp. adjustor display	24	25	40	40
G. FCS temp. adjustor display	45	45	45	45

[Table 2] Test Specimen F65A

[°C]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
A. Downstream side leak port temp monitor	45.9	45.8	45.8	45.7
B. Thermistor part	46.9	46.8	46.7	46.8
C. Bottom part of substrate	442.5	42.7	42.2	42.8
D. Room temperature	23.9	24.1	23.1	23.2
E. Chamber temp. monitor	22.9	23.3	42.7	43.8
F. Chamber temp. adjustor display	23	23	40	40
G. FCS temp. adjustor display	45	45	45	45

[Table 3] Test Specimen F20A

[°C]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
A. Downstream side leak port temp monitor	45.8	46.0	45.8	45.5
B. Thermistor part	47.2	47.2	46.8	47.0
C. Bottom part of substrate	42.9	43.1	43.8	43.4
D. Room temperature	23.3	24.5	25.6	26.1
E. Chamber temp. monitor	23.3	23.7	44.0	43.9
F. Chamber temp. adjustor display	22	24	40	40
G. FCS temp. adjustor display	45	45	45	45

As apparent from the results in Table 1 to Table 3 inclusive, there is seen a
 5 difference of about 0.2~2.2°C between the value displayed on the FCS
 temperature adjustor 6 in the item G and the temperature of the thermistor part
 in the item B. Therefore, this should be noted in the event that the strict
 temperature management is required fore electronic components and the like.

Table 4-1 shows the results of the measurement of the flow rate, which
 10 demonstrate that even under the same heating conditions, there are differences
 between the N₂ gas and the HF gas in the respective flow rate settings (75%,
 60%, 45%, 30% and 15%). The measured flow rates are the results of the
 measurements conducted 3 times. It is learned, then, that there is no

fluctuation of the flow rate value between the measurements with the pressure type flow rate control device. Furthermore, Table 4-2 shows the flow rates as measured to conduct the test stated in the afore-mentioned Item③ (Page 14) using the pressure type flow rate control device shown in Figure 1.

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[Table 4-1] Measurement Result of Flow Rate (Test Specimen F115A)

[SCCM]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
75% Ave.	<u>85.8</u>	<u>101.9</u>	<u>84.3</u>	<u>100.6</u>
75% 1st time	85.7	101.8	84.3	100.5
75% 2nd time	85.8	101.8	84.4	100.6
75% 3rd time	85.8	101.9	84.4	100.7
60% Ave.	<u>68.3</u>	<u>78.9</u>	<u>67.3</u>	<u>78.3</u>
60% 1st time	68.3	78.9	67.3	78.1
60% 2nd time	68.4	78.9	67.3	78.3
60% 3rd time	68.4	79.0	67.3	78.4
45% Ave.	<u>51.5</u>	<u>59.1</u>	<u>50.7</u>	<u>58.5</u>
45% 1st time	51.5	59.1	50.7	58.5
45% 2nd time	51.4	59.1	50.7	58.5
45% 3rd time	51.4	59.1	50.7	58.5
30% Ave.	<u>34.1</u>	<u>39.6</u>	<u>33.6</u>	<u>39.1</u>
30% 1st time	34.1	39.6	33.6	39.1
30% 2nd time	34.1	39.6	33.7	39.1
30% 3rd time	34.1	39.5	33.6	39.1
15% Ave.	<u>16.7</u>	<u>19.6</u>	<u>16.4</u>	<u>19.3</u>
15% 1st time	16.7	19.6	16.4	19.4
15% 2nd time	16.6	19.5	16.4	19.3
15% 3rd time	16.7	19.6	16.4	19.3

[Table 4-2] Measurement Result of Same Flow Rate

(Test Specimen F115A)

[SCCM]

Heating Conditions	②	
Gas Type	N ₂	HF
42.4% Ave.	<u>47.9</u>	<u>55.3</u>
42.4% 1st time	47.8	55.3
42.4% 2nd time	47.9	55.3
42.4% 3rd time	47.9	55.3
17.4% Ave.	<u>19.2</u>	<u>22.6</u>
17.4% 1st time	19.2	22.6
17.4% 2nd time	19.2	22.6
17.4% 3rd time	19.2	22.6

Table 5-1 shows the flow rate measurement results where F65A is used for
 5 the test specimen (the pressure type flow rate control device) 3, and Table 5-2
 shows the measurement results of the same flow rates with F65A.

Similarly, Table 6-1 shows the flow rate measurement results where F20A
 is used for the test specimen (the pressure type flow rate control device)3, and
 Table 6-2 shows the measurement results of the same flow rate with F20A.

[Table 5-1] Flow Rate Measurement (Test Specimen F65A)

[SCCM]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
75% Ave.	<u>48.3</u>	<u>55.4</u>	<u>46.3</u>	<u>56.1</u>
75% 1st time	48.3	55.4	46.3	56.2
75% 2nd time	48.3	55.4	46.4	56.0
75% 3rd time	48.3	55.3	46.3	56.0
60% Ave.	<u>38.7</u>	<u>43.8</u>	<u>37.1</u>	<u>43.1</u>
60% 1st time	38.7	43.8	37.1	43.1
60% 2nd time	38.6	43.8	37.1	43.1
60% 3rd time	38.7	43.8	37.1	43.0
45% Ave.	<u>28.9</u>	<u>32.7</u>	<u>27.8</u>	<u>32.2</u>
45% 1st time	28.9	32.7	27.8	32.2
45% 2nd time	28.9	32.8	27.8	32.2
45% 3rd time	28.9	32.7	27.8	32.2
30% Ave.	<u>19.0</u>	<u>21.8</u>	<u>18.3</u>	<u>21.4</u>
30% 1st time	19.0	21.7	18.3	21.4
30% 2nd time	19.0	21.7	18.2	21.4
30% 3rd time	19.0	21.8	18.3	21.4
15% Ave.	<u>9.1</u>	<u>10.6</u>	<u>8.8</u>	<u>10.4</u>
15% 1st time	9.1	10.6	8.8	10.4
15% 2nd time	9.1	10.6	8.8	10.4
15% 3rd time	9.1	10.6	8.8	10.4

[Table 5-2] Measurement of Same Flow Rate (Test Specimen F65A)

[SCCM]

Heating Conditions	②	
Gas Type	N ₂	HF
30.8% Ave.	<u>18.8</u>	<u>22.0</u>
30.8% 1st time	18.8	22.0
30.8% 2nd time	18.8	22.0
30.8% 3rd time	18.8	22.0

[Table 6-1] Flow Rate Measurement (Test Specimen F20A)

[SCCM]

Heating Conditions	①		②	
Gas Type	N ₂	HF	N ₂	HF
90% Ave.	<u>17.7</u>	<u>20.6</u>	<u>17.7</u>	<u>21.5</u>
90% 1st time	17.7	20.6	17.7	21.5
90% 2nd time	17.7	20.6	17.7	21.5
90% 3rd time	17.6	20.6	17.7	21.5
75% Ave.	<u>14.7</u>	<u>16.6</u>	<u>14.8</u>	<u>17.3</u>
75% 1st time	14.7	16.6	14.8	17.3
75% 2nd time	14.7	16.6	14.8	17.3
75% 3rd time	14.7	16.6	14.8	17.3
60% Ave.	<u>11.7</u>	<u>13.1</u>	<u>11.8</u>	<u>13.6</u>
60% 1st time	11.7	13.1	11.8	13.6
60% 2nd time	11.7	13.1	11.8	13.6
60% 3rd time	11.7	13.1	11.8	13.6
45% Ave.	<u>8.6</u>	<u>9.7</u>	<u>8.7</u>	<u>10.1</u>
45% 1st time	8.6	9.7	8.7	10.1
45% 2nd time	8.6	9.7	8.7	10.1
45% 3rd time	8.6	9.7	8.7	10.1
30% Ave.	<u>5.6</u>	<u>6.4</u>	<u>5.7</u>	<u>6.6</u>
30% 1st time	5.6	6.4	5.7	6.6
30% 2nd time	5.6	6.4	5.7	6.6
30% 3rd time	5.6	6.4	5.7	6.6
15% Ave.	<u>2.7</u>	<u>3.1</u>	<u>2.8</u>	<u>3.3</u>
15% 1st time	2.7	3.1	2.8	3.3
15% 2nd time	2.7	3.1	2.8	3.3
15% 3rd time	2.7	3.1	2.8	3.3

[table 6-2] Flow Rate Measurement (Test Specimen F20A)

[SCCM]

Heating Conditions	②	
Gas Type	N ₂	HF
48.8% Ave.	9.4	10.9
48.8% 1st time	9.4	10.9
48.8% 2nd time	9.4	10.9
48.8% 3rd time	9.4	10.9

Table 7 shows the flow factor F.F. for each test specimen (the pressure type flow rate control device) 3 computed on the basis of the actually measured flow rate values where the formula $F.F. = \text{HF gas flow rate} / \text{N}_2 \text{ gas flow rate}$.

The average value of F.F. is computed as the average of the F.F. values in the flow ranges of 14~60%. Under the heating conditions ②, it is learned that the difference is more than 1/100 order at respective flow rate ranges (%).

Table 8 ~Table 10 show the measurement errors or discrepancies between the test specimens 3 at the same set flow rate. Even if the same flow rate value is measured with the test specimens 3, it is learned that there occurs the error of 0.8SCCM at the maximum between the specimen types in the case of the HF gas.

[Table 7] Result of F.F. Measurement (Heating Conditions ②)

Flow Rate Range	F115A	F65A	F20A
75%	1.1928	1.2101	1.1714
60%	1.1626	1.1602	1.1543
45%	1.1545	1.1592	1.1545
3%	1.1634	1.1713	1.1646
15%	1.1791	1.1866	1.1738
F.F. Average	1.1649	1.1693	1.1618
F.F. 3 sets Ave.	1.1653		

15

[Table 8] Comparison Result of Same Setting Flow Rate

(Heating Conditions ②)

[SCCM]

Flow Rate Range	F115A	F65A	Error
Setting	42.4%	75.0%	(F65A-F115A)
N ₂	47.9	46.3	-1.5
HF	55.3	56.1	0.8

5

[Table 9] Comparison Result of Same Setting Flow Rate

(Heating Conditions ②)

[SCCM]

Flow Rate Range	F115A	F65A	Error
Setting	17.4%	38.0%	(F65A-F115A)
N ₂	19.2	18.8	-0.4
HF	22.6	22.0	-0.6

[Table 10] Comparison Result of Same Setting Flow Rate

(Heating Conditions ②)

[SCCM]

Flow Rate Range	F65A	F20A	Error
Setting	15.0%	48.8%	(F20A-F65A)
N ₂	8.8	9.4	0.6
HF	10.4	10.9	0.8

10

Figure 2 (a) is the graphic data of the flow rate measurement tests under the heating conditions ② with the test specimen F115A. Similarly, Figure 2 (b) and Figure 2 (c) are the graphic data with the test specimen F65A and the test specimen F20A, respectively.

15

As apparent from Figure 2, in the case of the pressure type flow rate

control device, the closer the set flow rate range gets to 100%, the greater the measured flow rate gets, compared with the case of the N₂ gas.

With Table 11 and Table 12, it was confirmed on the basis of the flow rate measurement by the build-up method whether there is any flow rate change or
5 not in each pressure range to find if the afore-mentioned critical condition is always realized at the time of the flow rate measurement of the HF gas. This is because the specific heat ratio of the HF gas is greater than those of normal gases, and there is a possibility that the critical pressure ratio becomes smaller.

More concretely, with calculation of the flow rate correction, it was
10 assumed that the pressure range is 30Torr~90Torr, selecting the pressure interval of 20Torr.

As apparent from the results in Table 11 and Table 12, there was observed little difference in the calculated flow rate ratio over all the pressure ranges. Accordingly, it is understood that even with the HF gas having a great gas
15 specific heat, the flow rate measurements are conducted under the situation where the critical condition is satisfied over the entire ranges of the said flow rate measurement tests.

[Table 11] Sample Data

No. of Measurement	Pressure Range: ΔP [Torr]	Computed Time Range [sec]	Computed Flow Rate Ratio
1st time	10-30	14.5	1.005
	30-50	14.5	1.004
	50-70	15.0	0.999
	70-90	14.5	0.998
	30-90	44.0	1.000
2nd time	10-30	14.5	1.002
	30-50	14.5	1.002
	50-70	15.0	1.001
	70-90	14.5	0.999
	30-90	44.0	1.000
3rd time	10-30	14.5	1.003
	30-50	14.5	1.002
	50-70	15.0	1.001
	70-90	14.5	0.997
	30-90	44.0	1.000

(F115A, HF gas, Heating conditions ②, Setting signal 75%)

[Table 12] Sample Data

No. of Measurement	Pressure Range: ΔP [Torr]	Computed Time Range [sec]	Computed Flow Rate Ratio
1st time	10-30	26.5	1.004
	30-50	26.0	1.001
	50-70	26.5	1.001
	70-90	26.5	0.997
	30-90	79.0	1.000
2nd time	10-30	26.5	1.006
	30-50	26.5	1.002
	50-70	26.5	0.999
	70-90	26.5	0.997
	30-90	79.5	1.000
3rd time	10-30	26.5	1.005
	30-50	26.5	1.002
	50-70	26.5	1.001
	70-90	26.5	0.997
	30-90	79.5	1.000

(F65A, HF gas, heating conditions ②, Setting signal 75%)

Table 13-1 ~ Table 13-4 inclusive show the results of the flow rate measurements with F114A and F115A-HT for the test specimen (the pressure type flow rate control device) 3, and the heated temperature of the test specimen 3 was $45^{\circ}\text{C} \pm 2^{\circ}\text{C}$ and $85^{\circ}\text{C} \pm 2^{\circ}\text{C}$. In the flow rate measurement in Table 13-1 to Table 13-4, the temperature of the build-up chamber 4 is fixed at 40°C .

ΔQ in Table 13-1 ~ Table 13-5 indicates the flow rate error between the flow rate at the time of the set temperature and the flow rate at the time of the base temperature (45°C and 85°C).

Table 13-5 shows the flow factor F.F. where F115A-HT is used for the test specimen 3. Here, the F.F. (measured value) is HF gas flow rate/N₂ gas flow rate, and the average value of the F.F. shows the average in the set flow rate ranges of 15~60%.

[Table 13-1] Flow Rate Measurement

Gas Type	N ₂			ΔQ	
	43°C	45°C	47°C	(43°C)-(45°C)	(47°C)-(45°C)
75%	94.0	93.4	94.0	0.6	0.7
60%	74.9	74.5	75.0	0.3	0.5
45%	56.3	56.1	56.2	0.2	0.1
30%	37.3	37.2	37.3	0.1	0.1
15%	18.2	18.2	18.2	0.0	0.0

(Test specimen F115A, Temperature of chamber: 40°C (fixed))

[Table 13-2] Flow Rate Measurement

[SCCM]

Gas Type	HF			ΔQ	
Heating Temp.	43°C	45°C	47°C	(43°C)–(45°C)	(47°C)–(45°C)
75%	113.4	111.0	110.3	2.4	–0.7
60%	87.5	86.6	86.6	1.0	0.0
45%	65.1	64.7	65.0	0.4	0.2
30%	43.5	43.3	43.5	0.2	0.2
15%	21.5	21.4	21.5	0.1	0.1

(Test specimen F115A, Temperature of chamber: 40°C (fixed))

5 [Table 13-3] Flow Rate Measurement

[SCCM]

Gas Type	N ₂			ΔQ	
Heating Temp.	83°C	85°C	87°C	(83°C)–(85°C)	(87°C)–(85°C)
75%	91.2	90.9	90.9	0.2	–0.1
60%	72.4	72.2	72.1	0.2	–0.1
45%	53.6	53.4	53.3	0.2	–0.1
30%	34.8	34.7	34.6	0.2	–0.1
15%	16.1	16.0	15.9	0.1	–0.1

(Test specimen F115A-HT, Temperature of chamber: 40°C (fixed))

[Table 13-4] Flow Rate Measurement

[SCCM]

Gas Type	HF			ΔQ	
Heating Temp.	83°C	85°C	87°C	(83°C)–(85°C)	(87°C)–(85°C)
75%	106.0	106.1	106.1	–0.1	0.0
60%	84.7	84.8	84.7	–0.1	–0.1
45%	63.0	63.0	62.9	0.1	–0.1
30%	41.1	41.0	40.8	0.1	–0.1
15%	19.0	18.9	18.8	0.1	–0.1

(Test specimen F115A-HT, Temperature of chamber: 40°C (fixed))

[Table 13-5] Results of F.F. Measurement (for high temperature)

Heating Temp.	83°C	85°C	87°C
75%	1.1630	1.1671	1.1683
60%	1.1702	1.1742	1.1749
45%	1.1760	1.1793	1.1791
30%	1.1793	1.1816	1.1813
15%	1.1854	1.1873	1.1883
F.F. Average	1.1777	1.1806	1.1809

Figure 3(a) and (b) are the plotted measurement results of the
5 afore-mentioned Table 13-1 to Table 13-4 inclusive. As apparent from Figure
3(a), in the event that the temperature for heating the test specimen 3 is around
45°C, there is found a tendency that the flow rate error becomes greater as the
set flow rate range rises with respect to the HF gas.

However, it is found that the flow rate error caused by the temperature
10 difference becomes near-zero over the entire area of the set flow rate ranges
when the temperature for heating the test specimen 3 reaches about 85°C.

That is, as found from the results in Figure 3 (a) and Figure 3 (b), it is
desirable that with the flow rate control of the HF gas using the pressure type
flow rate control device, the pressure type flow rate control device 3 is heated to
15 somewhere around 40°C ~ 85°C, or more preferably to the temperature not
lower than 60°C. It is learned that even when the process side pressure on the
downstream side of the orifice is around $10^{-2} \sim 10^2$ Torr, the control of the HF
gas with a practical, high precision can be assured over the flow rate range of

3~300SCCM by the heating temperature as mentioned above.

Figure 4 (a), (b) and (c) show the plotted, actually measured values of the flow rates using F115A, F65A and F20A as the test specimens where the temperature of the test specimen (the pressure type flow rate control device) 3 and temperature conditions for the build-up chamber are parameters.

As apparent from Figure 4 (a) ~ (c), F.F. has a high linearity under the heating conditions ① and ②.

The afore-mentioned test results have revealed the followings:

① Sufficient flow rate control of the HF gas over the flow rate ranges of 3~100SCCM can be achieved by appropriately heating the pressure type flow rate control. (Table 4-1 ~ Table 6-1)

② The pressure type flow rate control device can be heated to 40°C~85°C. 50°C is the guaranteed temperature for the substrate or the base plate (on which electronic components are mounted) inside the test specimen. This means that if the temperature for heating the pressure type flow rate control device is 45°C, it is lower than the guaranteed temperature of 50°C. This indicates that the pressure type flow rate control device with no special heat-resistance can serve sufficiently for the flow rate control of the HF gas. (Table 1 ~ Table 4)

③ When the measurement of the same flow rate was effected using two types of test specimens having different flow rate ranges, the flow rate error of the HF gas is found to be 0.8SCCM at the maximum. (F115A – 42.4% and F65A – 75% Table 8)

④ There is almost no problem about the linearity of the flow rate

measurement characteristics. (Figure 2 (a) ~ (c))

The average value of the measured flow factor F.F is 1.1653, which is almost the same for the all set ranges. (Table 7)

⑤ With regard to the flow rate linearity at a certain set flow rate range, calculation at each build-up pressure interval (20Torr) used for the flow rate calculation reveals that the error of the flow rate linearity at each interval was lower than +0.6% ~ - 0.4%, thus no problem regarding the linearity. (table 11 and Table 12)

⑥ In the event that the temperature for heating the test specimen 3 was set at $45\pm 2^{\circ}\text{C}$, it is found that the flow rate error was less than 2.4SCCM (at the time when heated to $45\sim 47^{\circ}\text{C}$: a flow rate setting of 75%). However, in the event that the heating temperature was set at $85\pm 2^{\circ}\text{C}$ using the test specimen for high temperature, it was found that the flow rate error was less than 0.1SCCM (at the time when heated to $83\sim 85^{\circ}\text{C}$: a flow rate setting of 15%). (Table 13-1~4)

Also, the technical literature study has revealed that the molecular weight of the HF gas is 20g/mol at the time of 76 Torr with the temperature higher than 25°C and 760 Torr with the temperature higher than 80°C , and also that the association heat of 6 HF molecules is 40,800cal/mol. Accordingly, it is assumed that at the time of the flow rate control of the HF gas, the flow rate control can be conducted by heating the gas to the temperature higher than 65°C (with the upper limit setting of 75%) as expressed by the formula.

There is no need to say that the afore-mentioned basic concepts in accordance with the present invention can apply to the flow rate control of the

HF gas using the mass flow control device and the like.

With the present invention, the upper limit for heating the temperature of the pressure type flow rate control device is set at 85°C. This is because when the temperature goes up higher than 85°C, dependency of the molecular
5 association of the HF gas on the pressure/temperature is gone.

With the afore-mentioned mode of carrying out the invention, the case was discussed where the clustering of fluid can be prevented by raising the temperature of the clustering fluid. However, it should be noted that the clustering of the clustering fluid depends not only on the temperature but also on
10 the pressure. Therefore, the clustering of the clustering fluid can be prevented by diluting the clustering fluid to a level lower than a partial pressure by adding the diluting gas.

That is, as illustrated in Figure 5, the fluid (the HF gas) is diluted by adding and mixing in the diluting gas (N₂), thus diluting the fluid (the HF gas) to a level
15 lower than a partial pressure to bring the clustering fluid from a state where a plural number of the gas molecules are assembled to a state where the gas molecules are made monomolecular (with the number of cluster molecules being small) so that the clustering of molecules of the fluid is prevented. By so doing, a stable supply of the clustering fluid such as the HF gas and the like can
20 be achieved without heating.

In Figure 5, 2 designates the supply line for the clustering fluid such as the HF gas and the like, 7 the supply line for the diluting gas such as N₂ gas and the like, 8 the diluting apparatus, 9 the pressure type flow rate control device, and 10 the apparatus using the clustering such as the HF gas and the like.

Effects of the Invention

With the present invention, the clustering fluid such as the HF gas and the like to be flow-rate controlled is passed through the orifice in a state where the pressure type flow rate control device is heated to the temperature not lower than 40, or the clustering fluid such as the HF gas and the like to be flow-rate controlled is passed through an orifice in a state where the pressure of the fluid is made lower than a partial pressure by adding the diluting gas so that the molecules of the fluid are permitted to turn monomolecular. As a result, the molecular association of the clustering fluid or the endothermic amount at the time of dissociation will not be influenced by the pressure of the clustering fluid such as the HF gas and the like. As a result, the flow rate of the clustering fluid such as the HF gas and the like can be controlled using the pressure type flow rate control device with high precision and stability as in the case of the flow rate control of normal gases such as the O₂ gas or the N₂ gas.

In addition, by keeping the heating temperature relatively low like between 40°C ~ 85°C, adverse effects on the electronic components used in the pressure type flow rate control device can be easily avoided. Therefore, the manufacturing costs of the pressure type flow rate control device are not expected to increase.

Furthermore, in the event that the pressure level of the chamber to which the clustering fluid such as the HF gas and the like is supplied is $10^{-2} \sim 10^2$ Torr, the high precision flow rate control of the clustering fluid such as the HF gas and the like can be achieved over the flow rate range of 3~300SCCM. This makes it possible to substantially lower the running costs and equipment costs

compared with the conventional case where a great amount of diluting gas is used.

As explained above, by making use of the flow factor F.F. of the clustering fluid based on the N₂ gas, the present invention can easily achieve the practical
5 effect that the high precision flow rate control of the clustering fluid is made possible using the known and widely used pressure type flow rate control device.

CLAIM

1. A method for the flow rate control of a clustering fluid in which the clustering
5 fluid is supplied from a fluid supply source to desired instruments or apparatuses
using a flow rate control device wherein the association of molecules of the
clustering fluid passing through the said flow rate control device is dissociated by
heating the afore-mentioned flow rate control device so that the clustering fluid
kept in a monomolecular state is supplied by the flow rate control device while
10 subjected to the flow rate control.
2. A method for the flow rate control of a clustering fluid in which the clustering
fluid is supplied from a fluid supply source to desired instruments or apparatuses
using a flow rate control device wherein the association of molecules of the
clustering fluid passing through the said flow rate control device is dissociated by
15 diluting the afore-mentioned clustering fluid to a level lower than a partial
pressure so that the clustering fluid kept in a monomolecular state is supplied by
the flow rate control device while subjected to the flow rate control.
3. A method for the flow rate control of a clustering fluid as claimed in Claim 1
or Claim 2 wherein a flow rate control valve, a heat type flow rate control device
20 or a pressure type flow rate control device is chosen for the flow rate control
device.
4. A method for the flow rate control of a clustering fluid as claimed in Claim 1
wherein the temperature for heating a main body of the flow rate control device
is not lower than 40°C.

5. A method for the flow rate control of a clustering fluid employing a pressure type flow rate control device in which the flow rate Q of the gas passing through an orifice is computed as $Q=KP_1$ (where K is a constant) with the gas passing through the orifice in a state where the ratio P_2/P_1 between the gas pressure P_1 on the upstream side of the orifice and the gas pressure P_2 on the downstream side of the orifice is held at a value not higher than the critical pressure ratio of the gas, wherein the clustering fluid is permitted to pass through the afore-mentioned orifice while the afore-mentioned pressure type flow rate control device is heated to the temperature not lower than 40°C .
6. A method for the flow rate control of a clustering fluid employing a pressure type flow rate control as claimed in Claim 5 wherein a chamber which is connected to the downstream side of the pressure type flow rate control device and receives the clustering fluid is a vacuum device.
7. A method for the flow rate control of a clustering fluid employing a pressure type flow rate control device as claimed in Claim 5 wherein the graduation of the pressure type flow rate control device is determined based on the N_2g gas, and the flow rate factor F.F. of the clustering fluid against the N_2 gas is appropriately chosen according to the temperature of a main body of the pressure type flow rate control device so that the flow rate of the clustering fluid is obtained by multiplying the measured flow rate value of the passing clustering fluid by the afore-mentioned flow rate factor F.F.
8. A method for the flow rate control of a clustering fluid employing a pressure type flow rate control device as claimed in Claim 6 wherein the pressure type flow rate control device is heated to the temperature of $40^{\circ}\text{C}\sim 85^{\circ}\text{C}$, and the

vacuum device which is connected to the downstream side of the pressure type flow rate control device and receives the clustering fluid is a vacuum chamber whose pressure is 10^{-3} Torr $\sim 10^2$ Torr.

9. A method for the flow rate control of a clustering fluid employing a pressure
5 type flow rate control device as claimed in Claim 5 wherein the range of the flow rate control of the clustering fluid to be supplied from the pressure type flow rate control device to the vacuum chamber is 3~300SCCM.

10. A method for the flow rate control of cluster-making fluids as claimed in Claim 1 to Claim 9 inclusive wherein the clustering fluid is hydrogen fluoride gas,
10 water or ozone.

11. A method for the flow rate control of a clustering fluid employing a pressure type flow rate control device as claimed in Claim 7 wherein the temperature of a main body of the pressure type flow rate control device is 45°C while the flow factor F.F. is 1.1665 when the fluid is hydrogen fluoride.

12. A flow rate control device for a clustering fluid which supplies the clustering
15 fluid from a fluid supply source to desired instruments or apparatuses wherein the association of molecules of the clustering fluid passing through the said flow rate control device is dissociated by heating a main body of the afore-mentioned flow rate control device so that the flow rate control device is permitted to control
20 the flow rate of the clustering fluid kept in a monomolecular state.

13. A flow rate control device for a clustering fluid which supplies the clustering fluid from a fluid supply source to desired instruments or apparatuses wherein the association of molecules of the clustering fluid passing through the said flow rate control device is dissociated by diluting the afore-mentioned clustering fluid

to a level lower than a partial pressure so that the flow rate control device is permitted to control the flow rate of the clustering fluid kept in a monomolecular state.

14. A flow rate control device for a clustering fluid as claimed in Claim 12 or
5 Claim 13 wherein a flow rate control valve, a heat type flow rate control device or a pressure type flow rate control device is chosen for the flow rate control device.

15. A flow rate control device for a clustering fluid as claimed in Claim 12 wherein the temperature for heating the main body of a flow rate control device is not lower than 40°C.

10 16. A pressure type flow rate control device for a clustering fluid in which the flow rate Q of a gas passing through an orifice is computed as $Q=KP_1$ (where K is a constant) with the gas passing through the orifice in a state where the ratio P_2/P_1 between the gas pressure P_1 on the upstream side of the orifice and the gas pressure P_2 on the downstream side of the orifice is held at a value not
15 higher than the critical pressure ratio of the gas wherein the fluid to be flow-rate controlled is a clustering fluid, and a heating device is installed in the pressure type flow rate control device so that a main body of the pressure type flow rate control device is heated to the temperature not lower than 40°C by the said heating device.

20 17. A pressure type flow rate control device for a clustering fluid as claimed in Claim 16 wherein the flow factor F.F. of the clustering fluid based on the N₂ gas is appropriately chosen according to the temperature of the main body of the pressure type flow rate control device, and the graduation of the pressure type flow rate control device is determined using the chosen flow factor F.F. value.

18. A flow rate control device for a clustering fluid as claimed in Claim 13 to Claim 17 inclusive wherein the clustering fluid is hydrogen fluoride gas, water or ozone.

19. A pressure type flow rate control device for a clustering fluid as claimed in
5 Claim 17 wherein the temperature of the main body of the pressure type flow rate control device is 45°C while the flow factor F.F. is 1.1665 when the fluid is hydrogen fluoride gas.