METHOD OF PROCESSING SUBSTRATE, METHOD OF MANUFACTURING SEMICONDUCTOR DEVICE, SUBSTRATE PROCESSING APPARATUS, AND PROGRAM

TECHNICAL FIELD

[0001] The present disclosure relates to a method of processing a substrate, a method of manufacturing a semiconductor device, a substrate processing apparatus, and a program.

BACKGROUND

[0002] A film may be formed on a substrate including a recess formed on the surface of the substrate by supplying a process gas to the substrate (*see*, for example, Patent Document 1).

[0003] [Prior Art Document]

Patent Document

[0004] Patent Document 1: International Publication Pamphlet No. WO/2022/196339

SUMMARY

[0005] The present disclosure provides a technique capable of improving step coverage of a film formed on a substrate.

[0006] According to one embodiment of the present disclosure, there is provided a technology including, supplying a process gas to a substrate from at least one storage in which the process gas is stored, wherein, in the act of supplying the process gas, an amount of gas in the at least one storage at an end time point of the act of supplying the process gas is set to be equal to or greater than a predetermined amount.

[0007] Some embodiments of the present disclosure provide a technique capable of improving a step coverage of a film formed on a substrate.

BRIEF DESCRIPTION OF DRAWINGS

[0008] FIG. 1 is a vertical cross-sectional view schematically illustrating a substrate processing apparatus according to an embodiment of the present disclosure.

FIG. 2A is a diagram illustrating a first process gas supply system according to an embodiment of the present disclosure, FIG. 2B is a diagram illustrating a second process gas supply system according to an embodiment of the present disclosure, and FIG. 2C is a diagram illustrating an exhaust system according to an embodiment of the present disclosure.

FIG. 3 is a schematic configuration diagram of a controller of the substrate processing apparatus according to an embodiment of the present disclosure, in which a control system of the controller is illustrated as a block diagram.

FIG. 4 is a diagram illustrating a substrate processing sequence according to an embodiment of the present disclosure.

FIG. 5 is a flowchart illustrating details of a first process gas supply process in FIG. 4.

FIG. 6 is an image diagram of an interior of a tank in the first process gas supply process in FIG. 4.

DETAILED DESCRIPTION

[0009] One embodiment of the present disclosure will be described mainly with reference to FIGS. 1 to 6. Drawings used in the following description are schematic, and the dimensional relationships, ratios, and the like of various elements shown in the drawings may not match actual ones. Further, the dimensional relationships, ratios, and the like of various elements among plural drawings may not match one another.

[0010] (1) Configuration of Substrate Processing Apparatus

A configuration of a substrate processing apparatus 10 will now be described with reference to FIG. 1.

[0011] The substrate processing apparatus 10 includes a reaction tube storage chamber 206b and includes, inside the reaction tube storage chamber 206b, a cylindrical reaction tube 210

extending in a vertical direction, a heater 211 serving as a heating part (furnace body) provided around an outer periphery of the reaction tube 210, a gas supply structure 212 serving as a gas supplier, and a gas exhaust structure 213 serving as a gas exhauster. The gas supplier may include an upstream gas guide 214 and nozzles 223 and 224 described later. Further, the gas exhauster may include a downstream gas guide 215 described later. A compartment of the reaction tube 210 in which a substrate S is processed is referred to as a process chamber 201. Further, the process chamber 201 may also be referred to as a process space inside which the substrate S is arranged.

[0012] The gas supply structure 212 is installed upstream of a gas flow direction of the reaction tube 210, and a gas is supplied from the gas supply structure 212 into the reaction tube 210. The gas is supplied to the substrate S in a horizontal direction. The gas exhaust structure 213 is installed downstream of the gas flow direction of the reaction tube 210, and the gas inside the reaction tube 210 is discharged from the gas exhaust structure 213. The gas supply structure 212, an interior of the reaction tube 210, and the gas exhaust structure 213 are in fluid communication with one another in the horizontal direction.

[0013] The upstream gas guide 214 is installed on an upstream side of the reaction tube 210 between the reaction tube 210 and the gas supply structure 212 to regulate a flow of the gas supplied from the gas supply structure 212. Further, the downstream gas guide 215 is installed on a downstream side of the reaction tube 210 between the reaction tube 210 and the gas exhaust structure 213 to regulate a flow of the gas discharged from the reaction tube 210. A lower end of the reaction tube 210 is supported by a manifold 216.

[0014] The reaction tube 210, the upstream gas guide 214, and the downstream gas guide 215 are formed in a continuous structure and are made of, for example, materials such as quartz or SiC. The reaction tube 210, the upstream gas guide 214, and the downstream gas guide 215 are constituted by heat-transmissive members that transmit heat radiated from the heater 211. The heat from the heater 211 heats the substrate S and the gas.

[0015] The gas supply structure 212 includes a distributor 225 connected to a gas supply pipe

251 and a gas supply pipe 261 and configured to distribute a gas supplied from each gas supply pipe. A plurality of nozzles 223 and 224 is installed downstream of the distributor 225. The gas supply pipe 251 and the gas supply pipe 261 supply different types of gases as described later. The nozzles 223 and 224 are arranged in a vertical relationship or in a lateral relationship. In the embodiment of the present disclosure, the gas supply pipe 251 and the gas supply pipe 261 are collectively referred to as a gas supply pipe 221. Each nozzle is also referred to as a gas discharger. The distributor 225 is configured to allow each gas to be supplied from the gas supply pipe 251 to the nozzle 223 and from the gas supply pipe 261 to the nozzle 224.

[0016] The upstream gas guide 214 includes a housing 227 and a partition plate 226. The partition plates 226 extend in the horizontal direction and are formed in continuous structures without holes. Herein, the term "horizontal direction" refers to a sidewall direction of the housing 227. A plurality of partition plates 226 is arranged in the vertical direction. The partition plates 226 are fixed to a sidewall of the housing 227 and are configured to prevent the gas from moving beyond the partition plates 226 to a lower or upper adjacent region.

[0017] Each partition plate 226 is installed at a position corresponding to each substrate S. The nozzles 223 and 224 are installed between the partition plates 226 and between the partition plates 226 and the housing 227. A flow of the gas discharged from the nozzles 223 and 224 is regulated by the partition plates 226 and the gas is then supplied to a surface of the substrate S. In other words, the gas is supplied from a lateral direction of the substrate S when viewed from the substrate S.

[0018] In a state in which substrates S are supported by a substrate support 300, the downstream gas guide 215 is configured so that a ceiling portion of the downstream gas guide 215 is higher than the uppermost substrate S and a bottom portion of the downstream gas guide 215 is lower than the lowermost substrate S on the substrate support 300.

[0019] The downstream gas guide 215 includes a housing 231 and partition plates 232. The partition plates 232 extend in the horizontal direction and are formed in continuous structures without holes. Here, the term "horizontal direction" refers to a sidewall direction of the housing

231. Furthermore, a plurality of partition plates 232 is arranged in the vertical direction. The partition plates 232 are fixed to a sidewall of the housing 231 and are configured to prevent the gas from moving beyond the partition plates 232 to a lower or upper adjacent region.

[0020] Each partition plate 232 is installed at a position corresponding to each substrate S and a position corresponding to each partition plate 226. It is desirable that the corresponding partition plates 226 and 232 are provided at an equal height. Furthermore, when processing the substrates S, it is desirable that the heights of the substrates S are aligned with the heights of the partition plates 226 and 232. As a result, a flow in the vertical direction across the partition plates 226, the substrates S, and the partitions 232 is suppressed, thereby uniformizing a pressure difference of gases on the respective substrates S. Accordingly, uniform processing can be performed for each substrate S.

[0021] By installing the partition plates 226 and 232 in the positional relationship described above, pressure loss in the vertical direction can be made uniform at upstream and downstream sides of each substrate S. In other words, a horizontal gas flow across the partition plate 226, the substrate S, and the partition plate 232, as indicated by arrows in the drawing, in which a vertical flow is suppressed, can be reliably formed. Accordingly, it is possible to reduce the pressure difference of gases on respective substrates S. This enables uniform processing for each substrate S.

[0022] The gas exhaust structure 213 is provided downstream of the downstream gas guide 215. The gas exhaust structure 213 mainly includes a housing 241 and an exhaust pipe connector 242. The housing 231 and the housing 241 are configured so that heights of ceiling portions of the housing 231 and the housing 241 and heights of bottom portions of the housing 231 and the housing 241 are continuous, respectively. An exhaust hole 244 is formed in a downstream side of the housing 241 and in a lower or horizontal direction of the housing 241 and is configured to discharge the gas which passes through the downstream gas guide 215. The gas exhaust structure 213 is a lateral exhaust structure installed in a lateral direction of the reaction tube 210 to discharge the gas in the lateral direction of the substrate S.

[0023] A transfer chamber 217 is provided below the reaction tube 210 via the manifold 216. In the transfer chamber 217, the substrate S is mounted (placed) on the substrate support (hereinafter, sometimes simply referred to as "boat") 300 by a vacuum transfer robot via a substrate load port, or the substrate S is discharged from the substrate support 300 by the vacuum transfer robot.

[0024] The substrate support 300, a partition plate support 310, and a vertical driver 400 that drives the substrate support 300 and partition plate support 310 (collectively referred to as "substrate holder") in the vertical direction and the rotational direction can be stored in the transfer chamber 217. FIG. 1 illustrates a state in which the substrate support 300 is raised by the vertical driver 400 and is stored in the reaction tube 210.

[0025] The vertical driver 400 includes a rotational driver 430 that rotates the substrate support 300 and the partition plate support 310 together and a boat elevator 420 that drives the substrate support 300 vertically relative to the partition plate support 310. The rotational driver 430 and the boat elevator 420 are fixed to a base flange 401 which serves as a lid supported on a base plate 402 by a side plate 403. A vacuum sealing O-ring 446 is provided on an upper surface of the base flange 401. As illustrated in FIG. 1, by being driven by a vertical drive motor 410, the upper surface of the base flange 401 is raised to a position at which the upper surface of the base flange 401 is pressed against the transfer chamber 217, so that the interior of the reaction tube 210 can be hermetically sealed. A vacuum bellows 443 is connected between a support 440 and a support 441 fixed to the substrate support 300.

[0026] A substrate support part is constituted by at least the substrate support 300 configured to support the substrate S and is stored inside the reaction tube 210. Further, the substrate support part is configured to move the substrate S by the vacuum transfer robot via the substrate load port (not illustrated) into the transfer chamber 217 or transfer the moved substrate S to the interior of the reaction tube 210 to form of a thin film on the surface of the substrate S. The substrate load port is installed, for example, at a sidewall of the transfer chamber 217. In addition, the substrate support part may include the partition plate support 310.

[0027] A plurality of substrates S is mounted in the substrate support 300 at a predetermined interval in the vertical direction. Here, the partition plates 314, disposed immediately below the substrates S, are arranged either above or below the substrates S or above and below the substrates S. The partition plates 314 separate spaces between each substrate S. The predetermined interval between the substrates S mounted in the substrate support 300 is the same as a vertical interval between the partition plates 314 fixed to the partition plate support 310. Further, a diameter of the partition plate 314 is configured to be larger than a diameter of the substrate S.

[0028] Although an example of five substrates S supported in the substrate support 300 is illustrated here, the present disclosure is not limited thereto. For example, the substrate support 300 may be configured to be capable of supporting approximately 5 to 50 substrates S. In addition, the partition plate 314 is also referred to as a separator. Further, notation of a numerical range such as "5 to 50 substrates" in the present disclosure implies that a lower limit value and an upper limit value are included in that range. Thus, for example, "5 to 50 substrates" indicates "5 substrates or more and 50 substrates or less". The same applies to other numerical ranges.

[0029] Next, details of a gas supply system and an exhaust system will be described with reference to FIGS. 2A to 2C.

[0030] As illustrated in FIG. 2A, a first process gas source 252, a mass flow controller (MFC) 253 serving as a flow rate controller (flow rate control part), a valve 275 serving as an opening/closing valve, a tank 259 serving as a storage that stores a gas inside the tank 259, a pressure sensor 301 serving as a pressure meter (pressure measurement part) that measures a pressure inside the tank 259, and a valve 254 are installed at the gas supply pipe 251 in this order from the upstream direction. Here, the pressure sensor 301 can be regarded as an example of a measurement portion that measures the amount of gas in the tank 259 or as a detector that detects when the amount of gas in the tank 259 reaches a specific level.

[0031] The first process gas source 252 is a gas source for supplying a first process gas as a

process gas. The first process gas is a raw material gas and is one of process gases. As the first process gas source 252, a vaporizer or the like may be used. A first process gas supply system 250 (also referred to as "raw material gas supply system") mainly includes the gas supply pipe 251, the MFC 253, the valve 275, the tank 259, the pressure sensor 301, and the valve 254. The first process gas source 252 may be included in the first process gas supply system 250.

[0032] A gas exhaust pipe 302 for discharging the gas inside the tank 259 to an outside of the process chamber 201 without supplying the gas inside the tank 259 to the process chamber 201 is connected between the pressure sensor 301 and the valve 254 in the gas supply pipe 251. A valve 303 is installed at the gas exhaust pipe 302. A first process gas exhaust system (also referred to as a raw material gas exhaust system) mainly includes the gas exhaust pipe 302 and the valve 303. The first process gas exhaust system may be included in the first process gas supply system 250.

[0033] A gas supply pipe 255 is connected between the valve 275 and the tank 259 in the gas supply pipe 251. An inert gas source 256, an MFC 257, and a valve 258 are installed at the gas supply pipe 255 in this order from the upstream direction. An inert gas is supplied from the inert gas source 256. A first inert gas supply system mainly includes the gas supply pipe 255, the MFC 257, and the valve 258. The inert gas supplied from the inert gas source 256 acts as a purge gas that purges a gas remaining in the reaction tube 210 in a substrate processing process. The inert gas source 256 may be included in the first inert gas supply system. The first inert gas supply system may be included in the first process gas supply system 250.

[0034] As illustrated in FIG. 2B, a second process gas source 262, an MFC 263, and a valve 264 are installed at the gas supply pipe 261 in this order from the upstream direction. The second process gas source 262 is a gas source for supplying a second process gas as the process gas. The second process gas is a reactive gas or a modifying gas that reacts with the raw material gas and is one of the process gases. A second process gas supply system 260 (also referred to as "reactive gas supply system") mainly includes the gas supply pipe 261, the MFC 263, and valve 264. The second process gas source 262 may be included in the second process gas supply

system 260.

[0035] A gas supply pipe 265 is connected to a downstream side of the valve 264 in the gas supply pipe 261. An inert gas source 266, an MFC 267, and a valve 268 are installed at the gas supply pipe 265 in this order from the upstream direction. An inert gas is supplied from the inert gas source 266. A second inert gas supply system mainly includes the gas supply pipe 265, the MFC 267, and the valve 268. The inert gas supplied from the inert gas source 266 acts as a purge gas that purges the gas remaining in the reaction tube 210 in the substrate processing process. The inert gas source 266 may be included in the second inert gas supply system. The second inert gas supply system may be included in the second process gas supply system 260.

[0036] Next, an exhaust system will be described with reference to FIG. 2C. An exhaust system 280 configured to exhaust an atmosphere of the reaction tube 210 includes an exhaust pipe 281 that is in communication with the reaction tube 210 and is connected to the housing 241 via the exhaust pipe connector 242.

[0037] As illustrated in FIG. 2C, a vacuum pump 284 serving as a vacuum exhauster is connected to the exhaust pipe 281 via a valve 282 and an automatic pressure controller (APC) valve 283 serving as a pressure regulator (pressure regulation part) and is configured to be vacuum-exhausted such that an internal pressure of the reaction tube 210 reaches a predetermined pressure (degree of vacuum). The exhaust pipe 281, the valve 282, and the APC valve 283 are collectively referred to as the exhaust system 280. The exhaust system 280 is also referred to as a process chamber exhaust system. In addition, the vacuum pump 284 may be included in the exhaust system 280.

[0038] Next, a controller, which is a control part (control means), will be described with reference to FIG. 3. The substrate processing apparatus 10 includes a controller 600 configured to control an operation of each component of the substrate processing apparatus 10.

[0039] The controller 600 is schematically illustrated in FIG. 3. The controller 600 is configured as a computer including a central processing unit (CPU) 601, a random access memory (RAM) 602, a memory 603 as a memory part, and an input/output (I/O) port 604. The

RAM 602, the memory 603, and the I/O port 604 are configured to be capable of exchanging data with the CPU 601 via an internal bus 605.

[0040] The memory 603 is constituted by, for example, a flash memory, a hard disk drive (HDD), and the like. A control program that controls operations of the substrate processing apparatus 10, a process recipe in which substrate processing procedures or conditions are written, or the like are readably stored in the memory 603.

[0041] The process recipe is combined so that each procedure in the substrate processing process, which will be described later, is executed by the controller 600 so as to obtain an expected result. The process recipe functions as a program. Hereinafter, the process recipe or the control program may be generally and simply referred to as a "program." The term "program" used herein may indicate a case in which the process recipe alone is included, a case in which the control program alone is included, or a case in which both the process recipe and the control program are included. The RAM 602 is configured as a memory area (work area) in which programs, data, or the like read by the CPU 601 is temporarily stored.

[0042] The I/O port 604 is connected to the above-described vertical driver 400, heater 211, APC valve 283, vacuum pump 284, MFCs 253, 257, 263, and 267, valves 254, 258, 264, 268, 275, 282, and 303, pressure sensor 301, rotational driver 430, and the like.

[0043] The CPU 601 is configured to read and execute the control program from the memory 603 and to read the process recipe from the memory 603 in response to an input of an operation command from an input/output device 681, or the like. Then, the CPU 601 is configured to be capable of controlling, according to contents of the read process recipe, an operation of raising or lowering the substrate support 300 by the vertical driver 400, a heating operation by the heater 211, an opening/closing operation of the APC valve 283, an operation of starting or stopping the vacuum pump 284, a flow rate regulation operation of various gases by the MFCs 253, 257, 263, and 267, an opening/closing operation of the valves 254, 258, 264, 268, 275, 282, and 303, an operation of measuring the pressure inside the tank 259 by the pressure sensor 301, an operation of rotating the substrate support 300 and adjusting a rotational speed of the substrate support 300

using the rotational driver 430, and the like. The MFCs 253, 257, 263, and 267, the valves 254, 258, 264, 268, 275, 282, and 303, the APC valve 283, and the vacuum pump 284 can be used as a gas controller configured to be capable of controlling the supply and exhaust of gas into the process chamber 201.

[0044] The controller 600 according to the embodiment of the present disclosure may be configured by installing the above-described program in the computer using an external memory (for example, a magnetic disk such as a hard disk, an optical disc such as a DVD, a magneto-optical disc such as an MO, and a semiconductor memory such as a USB memory) 682 that stores the above-described program. Further, a means for supplying the program to the computer is not limited to supplying the program via the external memory 682. For example, the program may be supplied using a communication means such as the Internet or a dedicated line, without using the external memory 682. The memory 603 and the external memory 682 are configured as non-transitory computer-readable recording media that stores the program. Hereinafter, the memory 603 and the external memory 682 may be generally and simply referred to as a "recording medium." The term "recording medium" used herein may indicate a case in which the memory 603 alone is included, a case in which the external memory 682 alone is included, or a case in which both the memory 603 and the external memory 682 are included.

[0045] (2) Substrate Processing Process

Next, as a process of manufacturing a semiconductor device, a process of forming a thin film on the substrate S using the substrate processing apparatus 10 of the above-described configuration will be described with reference to FIGS. 4 to 6. In the following description, operations of each component constituting the substrate processing apparatus 10 are controlled by the controller 600.

[0046] Here, a film formation processing of forming a film in a recess such as a trench or a hole of the substrate S using a first process gas and a second process gas will now be described.

[0047] The term "substrate" used herein may refer to "a substrate itself "or "a stacked body of a substrate and certain layers or films formed on a surface of the substrate." The phrase "a

surface of a substrate" used herein may refer to "a surface of the substrate itself" or "a surface of certain layer or the like formed on the substrate." The expression "a certain layer is formed on a substrate" used herein may mean that "a certain layer is formed directly on a surface of a substrate itself" or that "a certain layer is formed directly on a layer or the like formed on the substrate." The term "wafer" used herein may be synonymous with the term "substrate."

[0048] (Substrate Loading Process S1)

A plurality of substrates S is charged onto the substrate support 300 in the transfer chamber 217 (Wafer Charging). Then, the substrate support 300 supporting the substrates S is raised by the vertical driver 400, so that the substrates S are loaded into the reaction tube 210 (Boat Loading), and the lower end of the reaction tube 210 is sealed by the base flange 401 via the O-ring 446. At this time, the surfaces of the substrates S are positioned so as to be aligned with the heights of the partition plates 226 and the partition plates 232.

[0049] (Pressure/Temperature Regulation Process S2)

The interior of the reaction tube 210 is vacuum-exhausted by the vacuum pump 284 so as to reach a desired pressure (Pressure Regulation). In addition, the heater 211 heats the process chamber 201 to a desired temperature (Temperature Regulation). Further, the rotational driver 430 starts to rotate the substrates S. These various operations are performed continuously at least until the processing of the substrates S is completed.

[0050] (Film Processing Process S3)

Next, the film processing process S3 is performed by executing a first process gas supply process S31, a purge gas supply process S32, a second process gas supply process S33, and a purge gas supply process S34 a predetermined number of times (n times, where n is an integer of 1, 2, or greater).

[0051] [First Process Gas Supply Process S31]

Next, the first process gas is flash-supplied to the process chamber 201 in which the substrates S are arranged. Here, the term "flash-supply" means that a large flow rate of gas is supplied into the reaction tube 210 in a short period of time.

[0052] Next, a storage process S311 and a supply process S312 are executed a predetermined number of times (p times, where p is an integer of 1, 2, or greater) to perform the first process gas supply process S31. FIG. 5 is a flowchart illustrating details of the first process gas supply process S31, and FIG. 6 is an image diagram illustrating the first process gas supply process S31. In FIG. 6, the amount of the first process gas stored in the tank 259 is schematically represented by a height of a gray rectangle in the tank 259. Further, FIG. 6 illustrates the amount of the first process gas stored in the tank 259, sequentially from the left, at the start of the storage process S311; at the end of the storage process S311 and at the start of the supply process S312; at the end of the supply process S312 and at the start of the storage process S311; and at the end of the storage process S311 and at the start of the supply process S312. Hereinafter, as illustrated in FIG. 6, a case in which a predetermined amount X or more of the first process gas is stored in the tank 259 at the start of the first process gas supply process S31 will be described by way of example.

[0053] <Storage Process S311>

In this step, the valve 275 is opened in a state in which the valves 254, 258, 303, 264, and 268 are closed, and the valve 275 is closed when a predetermined time passes. At this time, the first process gas, a flow rate of which is regulated by the MFC 253, is filled into the tank 259 in which the predetermined amount X or more of the first process gas is stored. The valve 275 is closed when a predetermined time passes.

[0054] At this time, the flow rate of the first process gas supplied to the tank 259 and/or the time for which the first process gas is/are continuously supplied to the tank 259 is controlled according to a remaining amount of the first process gas in the tank 259. In other words, the amount of the first process gas stored in the tank 259 in this step can be varied in each cycle of the first process gas supply process S31. The amount of the first process gas stored in the tank 259 in this step can be regulated by varying the flow rate of the first process gas supplied to the tank 259 (hereinafter referred to as the storage flow rate) and/or the time for which the first process gas is continuously supplied to the tank 259 (hereinafter referred to as the storage time).

Therefore, the amount of the first process gas stored in the tank 259 at the start of the supply process S312 can be kept constant between cycles, even when the remaining amount of the first process gas in the tank 259 at the end of the supply process S312 described later is varied between cycles.

[0055] In this step, any one of the storage flow rate and the storage time may be kept constant throughout a plurality of cycles, and the other may be varied at least once during the plurality of cycles. That is, according to the remaining amount of the first process gas in the tank 259 at the end of the supply process S312, the storage flow rate may be controlled by the MFC 253, or the storage time may be controlled by opening and closing the valve 275. Alternatively, both the storage flow rate controlled by the MFC 253 and the storage time controlled by opening and closing the valve 275 may be controlled. By keeping the storage flow rate constant throughout the plurality of cycles and varying the storage time at least once during the plurality of cycles, the amount of the first process gas stored in the tank 259 is easily controlled even when the pressure inside the tank 259 is high. By keeping the storage time constant throughout the plurality of cycles and varying the storage flow rate at least once during the plurality of cycles, variations in processing between lots caused by changes in the storage time is reduced.

[0056] <Supply Process S312>

The first process gas stored in the tank 259 is flash-supplied to the substrates S. That is, the valve 254 is opened in a state in which the valves 258, 275, 303, 264, and 268 are closed, and the valve 254 is closed when a predetermined time passes. At this time, the first process gas of high pressure stored in the tank 259 is flash-supplied to the substrates S via the gas supply pipe 251. The first process gas is supplied in large quantities from the gas supply structure 212 through the upstream gas guide 214 into the reaction tube 210 in a short period of time and then is exposed. Thereafter, the first process gas is exhausted through a space on the substrates S, the downstream gas guide 215, the gas exhaust structure 213, and the exhaust pipe 281. At this time, the valve 282 and the APC valve 283 are opened.

[0057] The amount of the first process gas remaining in the tank 259 at an end time point (a time

point when the valve 254 is closed) of this step is set to be equal to or greater than the predetermined amount X. In other words, in this step, the valve 254 is closed at a time point when the amount of the first process gas in the tank 259 reaches a target amount Z equal to or greater than the predetermined amount X. The predetermined amount X can be set to any value between 0.1% to 99% of the amount of the first process gas (a predetermined amount Y) stored in the tank 259 at the start of the supply process S312.

[0058] Here, setting the predetermined amount X to 10% to 90% of the predetermined amount Y, controlling a step coverage and a film formation rate to become desirable values becomes easier. If the predetermined amount X is less than 10% of the predetermined amount Y, the first process gas is supplied to the substrate S at a low flow rate, which may result in a deterioration of the step coverage. In addition, as will be described later, the first process gas undergoes an increased temperature drop while expanding in a process close to adiabatic, which may result in a deterioration of the film formation rate. If the predetermined amount X exceeds 90% of the predetermined amount Y, the amount of the first process gas supplied to the substrate S is reduced, which may result in a deterioration of the film formation rate. Here, setting the predetermined amount X to 20% to 85% of the predetermined amount Y is desirable, since this makes it easier to control the step coverage and the film formation rate to desirable values. Setting the predetermined amount X to 30% to 80% of the predetermined amount Y is further desirable, since this makes it much easier to control the step coverage and the film formation rate.

[0059] As described above, the first process gas may be flash-supplied to the substrate S under desired conditions. Specifically, the first process gas can be flash-supplied so that at least one selected from the group of the flow velocity, a residence time, a decomposition rate, and a partial pressure of a first process gas inside the process chamber 201 falls within a desired range. In other words, by leaving the predetermined amount X or more of the first process gas to remain in the tank 259 at the end time point of this step, for example, in this step, the flow velocity of the first process gas can be set to be equal to or higher than a predetermined flow velocity and the

residence time of the first process gas in the process chamber 201 can be shortened.

[0060] Here, the predetermined amount X remaining in the tank 259 at the end time point of this step is set so that the decomposition rate of the first process gas in the process chamber 201 in which the substrates S are present in this step, is equal to or less than a predetermined decomposition rate. If the decomposition rate of the first process gas supplied to the substrate S becomes higher than the predetermined decomposition rate, the step coverage may be easily deteriorated. By preventing the decomposition rate of the first process gas supplied to the substrate S from becoming higher than the predetermined decomposition rate, the step coverage can be improved.

[0061] In other words, the predetermined amount X is set so that the flow velocity of the first process gas in the process chamber 201 in this step is equal to or higher than the predetermined flow velocity. This makes it possible to prevent the decomposition rate of the first process gas supplied to the substrate S from becoming higher than the predetermined decomposition rate. This makes it possible to improve the step coverage.

[0062] When the first process gas is supplied, reaction by-products such as hydrogen chloride (HCl) that inhibit the adsorption of the first process gas onto the surface of the substrate S may be generated. Therefore, while at least a portion of adsorption sites on the surface of the substrate S is adsorbed by an element contained in the first process gas, the reaction by-products such as HCl are adsorbed onto the adsorption sites on the substrate S and inhibit the adsorption of the element contained in the first process gas. In this step, the flow velocity of the first process gas in the process chamber 201 is set to be equal to or higher than the predetermined flow velocity, and the first process gas is flash-supplied. Since a large amount of the first process gas is supplied to the substrate S at the start of the supply for a short period of time, the adsorption sites by the reaction by-products onto the substrate S are reduced and an amount of adsorption by the element contained in the first process gas can be increased. This can improve a step coverage performance and improve the film formation rate.

[0063] Here, the flow velocity of the first process gas refers to a number indicating an index of a

distance traveled per unit time by the first process gas supplied into the process chamber 201. Alternatively, in the process chamber 201, the flow velocity of the first process gas may refer to a number indicating an index of a distance traveled per unit time by the first process gas. Alternatively, the flow velocity of the first process gas may refer to a number indicating an index of a distance traveled per unit time by a gas in a processing space. Alternatively, the flow velocity of the first process gas may refer to an average flow velocity of the first process gas on the substrate S. In addition, the flow velocity of the first process gas may refer to an average flow velocity from when the first process gas travels from one location (or region) until the first process gas reaches another location (or region), for example, an average flow velocity from when the first process gas is discharged from a gas supplier such as the nozzle 223 into the process chamber 201 until the first process gas reaches the exhaust hole 244. Alternatively, the average flow velocity of the first process gas in the processing space may be used as the flow velocity of the first process gas. Alternatively, the average flow velocity of the first process gas in the process chamber 201 may be used as the flow velocity of the first process gas. Alternatively, the flow velocity of the first process gas at a certain point on the substrate S, in the processing space, or in the process chamber 201 may be used as the flow velocity of the first process gas.

[0064] In addition, by setting the flow velocity of the first process gas in the process chamber 201 to be equal to or higher than the predetermined flow velocity, a residence time of the first process gas in the process chamber 201 is equal to or less than a predetermined residence time. That is, the predetermined amount X may be set so that the residence time of the first process gas in the space in which the substrates S are present in this step is equal to or less than the predetermined residence time. This makes it possible to prevent the decomposition rate of the first process gas supplied to the substrate S from becoming higher than the predetermined decomposition rate.

[0065] When forming a film in a recess (or a groove, a trench, or a hole) formed on the substrate S, as a reactivity of the gas increases, adsorption onto an opening side of the recess becomes

easier while adsorption onto a deeper side of the recess becomes difficult. Therefore, when forming the film in the recess using, as the first process gas, a gas that generates a substance with high reactivity upon decomposition, by setting the residence time of the first process gas to be equal to or less than the predetermined residence time and/or by setting the flow velocity of the first process gas to be equal to or higher than the predetermined flow velocity, the decomposition rate of the first process gas may be lowered and the step coverage may be improved and enhanced.

[0066] Here, the residence time of the first process gas refers to a number indicating an index of the time from when the first process gas is supplied into the process chamber 201 until the first process gas is exhausted from the process chamber 201. The residence time of the first process gas may also refer to a number indicating an index of the time from when the first process gas that reaches a process space until the first process gas leaves the process space. Alternatively, the residence time of the first process gas may refer to the time from when the first process gas reaches the substrate S until the first process gas leaves the substrate S. In addition, the residence time of the first process gas may refer to, for example, the time from when the first process gas reaches the process space until the first process gas leaves the process space. Alternatively, the residence time of the first process gas may refer to the time from when the first process gas is discharged from the gas supplier such as the nozzle 223 until the first process gas reaches the exhaust hole 244. Alternatively, the residence time of the first process gas may refer to the time from when the supply of the first process gas is started to when the supply of the first process gas is ended. For instance, the residence time of the first process gas may refer to the time from when a predetermined component of the substrate processing apparatus 10 starts or ends a predetermined operation to when the predetermined component starts or ends the predetermined operation again, for example, the time from when the valve 254 downstream of the tank 259 is opened to when the valve 254 is closed.

[0067] The predetermined amount X may be set so that a partial pressure of the first process gas in the space in which the substrates S are present in this step is equal to or higher than a

predetermined partial pressure. This allows the first process gas to reach even the deeper side of the recess more easily, thereby improving the step coverage. Furthermore, a probability of collision of the first process gas with the surface of the substrate S per unit time and unit area is increased. This may improve the film formation rate, which is a thickness of the film formed on the surface of the substrate S per unit time.

[0068] The predetermined amount X may be set so that a temperature inside the tank 259 (for example, a temperature of the tank 259 itself or a temperature of the first process gas in the tank 259) in this step is equal to or higher than a predetermined temperature. This may improve the film formation rate.

[0069] When the first process gas filled in the tank 259 is supplied from the tank 259 to the process chamber 201, the first process gas in the tank 259 expands in a short period of time. At this time, since the first process gas in the tank 259 undergoes a process close to adiabatic expansion, a temperature of the first process gas released from the tank 259 drops, which causes the temperatures in the tank 259, the substrate S, and the process chamber 201 to also drop. Furthermore, due to these temperature drops, a reaction between the substrate, the first process gas, and/or the second process gas becomes less likely to proceed (a reactivity of the first process gas and/or the second process gas decreases), which may result in a deterioration in the film formation rate.

[0070] A case in which the predetermined amount X is set so that the temperature of the tank 259 or the temperature of the first process gas in the tank 259 in this step is equal to or higher than the predetermined temperature is considered. In this case, the temperature drop of the first process gas can be reduced, and a decrease in the reactivity of the first process gas and/or the second process gas can be suppressed. Thus, the film formation rate may be improved.

[0071] The predetermined amount X may also be set so that the first process gas present in the tank 259 in this step is not liquefied or solidified. As a result, a generation of particles may be suppressed.

[0072] When the first process gas filled in the tank 259 is supplied from the tank 259 to the

process chamber 201, the first process gas is supplied to the substrate S through a process close to adiabatic expansion, which causes the temperature of the first process gas or the tank 259 to drop. At the same time, the pressure inside the tank 259 is also lowered. As a result, the temperature and pressure in the tank 259 may reach a temperature and pressure at which the first process gas is liquefied or solidified. The liquefied or solidified first process gas may cause the generation of particles.

[0073] In this step, in a case where the predetermined amount X is set so as not to liquefy or solidify the first process gas present in the tank 259, the temperature drop caused by the adiabatic expansion of the first process gas can be reduced. This makes liquefaction or solidification of the first process gas less likely to occur. Thus, the generation of the particles may be suppressed.

[0074] In other words, the amount of the first process gas in the tank 259 at an end time point of the supply process S312 can be set so that the first process gas in the tank 259 is not liquefied or solidified between a start time point and the end time point of the supply process S312. Accordingly, even in a case in which, for example, a raw material gas that is easily liquefied or solidified by a decrease in pressure and/or temperature is flash-supplied, the raw material gas in the tank 259 can be suppressed from being liquefied or solidified, thereby suppressing the generation of the particles. In other words, the technique of the present disclosure can be suitably applied even in a case in which a process gas obtained by vaporizing a liquid or solid substance at room temperature and normal pressure is flash-supplied.

[0075] In this step, the valve 258 may be opened to supply an inert gas into the gas supply pipe 251 via the gas supply pipe 255. In addition, in order to prevent the first process gas from entering the gas supply pipe 261, the valves 268 and 264 may be opened to supply the inert gas into the gas supply pipe 261.

[0076] <Performing Predetermined Number of Times, S313>

A cycle of the above-described storage process S311 and supply process S312 is performed a predetermined number of times (p times, where p is an integer of 1, 2, or greater).

Consequently, a first layer of a desired thickness can be formed.

[0077] In the flash-supply, the first process gas stored in the tank 259 is supplied to the process chamber 201 while expanding in a process close to adiabatic expansion. Therefore, by performing the flash-supply, the temperature of the first process gas and a temperature of the first process gas supply system 250 including the tank 259 are dropped. Here, the technique of the present disclosure may control a magnitude of the pressure drop in the tank 259 by the amount of the first process gas in the tank 259 at the end of the flash-supply. Therefore, the technique of the present disclosure may control the magnitude of the temperature drop in the tank 259 caused by the adiabatic expansion of the first process gas accompanied by the flash-supply.

[0078] The amount of the first process gas in the tank 259 may be controlled by the pressure in the tank 259. That is, the amount of the first process gas in the tank 259 may be controlled by the pressure in the tank 259 which is measured by the pressure sensor 301. Here, the control of the pressure in the tank 259 can be regarded as the control of the amount of the first process gas in the tank 259. Therefore, it can be considered that the supply process S312 ends after the measurement portion (for example, the pressure sensor 301) detects that the amount of gas in the tank 259 is equal to or greater than the predetermined amount.

[0079] That is, the supply process S312 may be performed so that the pressure in the tank 259 at the end time point of the supply process S312 is equal to or higher than a predetermined pressure. Even in this case, the decomposition rate of the first process gas supplied to the substrate S can be prevented from becoming higher than the predetermined decomposition rate. Here, the predetermined pressure can be set to any value between 0.1% and 99% of the pressure in the tank 259 at the start of the supply process S312.

[0080] Here, setting the predetermined pressure to 10% to 90% of the pressure in the tank 259 at the end time point of the supply process S312 is desirable, since this makes it easier to control the step coverage and the film formation rate to desirable values. In a case where the predetermined pressure is less than 10% of the pressure in the tank 259, the step coverage may

be deteriorated due to the first process gas being supplied to the substrate S at a low flow rate. In addition, as described later, the temperature drop of the first process gas may increase due to the first process gas undergoing a process close to an adiabatic expansion, and the film formation rate may be deteriorated. In a case where the predetermined pressure exceeds 90% of the pressure in the tank 259, the amount of the first process gas supplied to the substrate S is reduced, and the film formation rate may be deteriorated. Here, setting the predetermined pressure to 20% to 85% of the pressure in the tank 259 at the end time point of the supply process S312 is desirable, since this makes it easier to control the step coverage and the film formation rate to desired values. Setting the predetermined pressure to 30% to 80% of the pressure in the tank 259 at the end time point of the supply process S312 is further desirable, since this makes it much easier to control the step coverage and the film formation rate.

[0081] [Purge Gas Supply Process S32]

In this step, a purge gas is supplied to the process chamber 201 in which the substrates S are placed. That is, after the first layer is formed on the substrates S, the valves 254, 258, and 268 are opened in a state in which the valves 275, 303, and 264 are closed, and the inert gas is supplied as the purge gas into the gas supply pipes 251 and 261 via the gas supply pipes 255 and 265. At the same time, the interior of the process chamber 201 is vacuum-exhausted by the vacuum pump 284 and is then purged while the valve 282 and the APC valve 283 of the exhaust pipe 281 are opened.

[0082] [Second Process Gas Supply Process S33]

In this step, the valve 264 is opened in a state in which the valves 254, 258, 275, 303, and 268 are closed, and the valve 264 is closed when a predetermined time passes. At this time, the second process gas, the flow rate of which is regulated by the MFC 263, is supplied to the substrate S via the gas supply pipe 261. The second process gas is supplied from the gas supply structure 212 via the upstream gas guide 214 into the reaction tube 210 and then is exposed. Thereafter, the second process gas is exhausted via the space on the substrates S, the downstream gas guide 215, the gas exhaust structure 213, and the exhaust pipe 281. At this time, the valve

282 and the APC valve 283 are open. Here, the valve 268 may be opened to allow the inert gas to flow through the gas supply pipe 261 via the gas supply pipe 265. In addition, in order to prevent the second process gas from entering the gas supply pipe 251, the valves 258 and 254 may be opened to allow the inert gas to flow through the gas supply pipe 251. Consequently, the first layer on the substrate S can be modified into a second layer.

[0083] [Purge Gas Supply Process S34]

In this step, the purge gas is supplied to the process chamber 201 in which the substrates S are placed, by the same processing procedure as in the purge gas supply process S32 described above. That is, after the first layer on the substrate S is modified into the second layer, the valves 254, 258, and 268 are opened in a state in which the valves 275, 303, and 264 are closed and, the inert gas as the purge gas are supplied into the gas supply pipes 251 and 261 via the gas supply pipes 255 and 265. At the same time, the interior of the process chamber 201 is vacuum-exhausted by the vacuum pump 284 and is then purged while the valve 282 and the APC valve 283 of the exhaust pipe 281 are opened.

[0084] [Performing Predetermined Number of Times, S35]

A cycle of the above-described first process gas supply process S31, purge gas supply process S32, second process gas supply process S33, and purge gas supply process S33 is performed a predetermined number of times (n times, where n is an integer of 1, 2, or greater). As a result, a film of a predetermined thickness containing an element derived from the first process gas and an element derived from the second process gas is formed on the substrate S including a recess. As a result, a film with improved step coverage performance and enhanced film formation rate can be formed on the substrate S including the recess.

[0085] (Atmospheric Pressure Restoration Process S4)

After the film processing on the substrate S is completed, the inert gas is supplied into the process chamber 201 and exhausted through the exhaust pipe 281. The inert gas acts as the purge gas, and the interior of the process chamber 201 is purged with the inert gas. As a result, gases or reaction by-products remaining in the process chamber 201 are removed from the

process chamber 201. Thereafter, an atmosphere of the process chamber 201 is substituted with the inert gas, and the pressure inside the process chamber 201 is restored to an atmospheric pressure (Restoration to Atmospheric Pressure).

[0086] (Substrate Unloading Process S5)

The base flange 401 is lowered by the vertical driver 400 to open the lower end of the reaction tube 210. Then, the processed substrate S is unloaded to the transfer chamber 217 while being supported by the substrate support 300 (Boat Unloading). The processed substrate S is discharged from the substrate support 300 in the transfer chamber 217 (Wafer Discharging).

[0087] (3) Pre-evaluation Process

Next, a pre-evaluation process performed before the above-described substrate processing process will be described.

[0088] In the pre-evaluation process, the time from the start time point of the supply process S312 of the first process gas supply process S31 to a time point when the amount of the first process gas in the tank 259 becomes the predetermined amount X is obtained using a predetermined method. Then, data obtained in this process is used in the above-described substrate processing process. Examples of the predetermined method include obtaining data from substrate processing or experiments performed in advance, making predictions based on the obtained data, and performing numerical calculations including analysis using computational fluid dynamics (CFD), machine learning, deep learning or the like.

[0089] Then, a current amount of the first process gas in the tank 259 is identified based on the pressure in the tank 259 measured by the pressure sensor 301. The valve 275, the MFC 253, and the like are controlled so that the supply process S312 is ended before or at a time point when the time acquired by the pre-evaluation process elapses from the start time point of the supply process S312. This eliminates the need to perform control such as ending the flash-supply at a time point when the amount of the first process gas in the tank 259 measured by the pressure sensor 301 is equal to or greater than the predetermined amount X. In other words, control by the controller 600 can be simplified.

[0090] In the above description, the gas flow is expressed as being formed horizontally, but the a main stream of gas formed in the horizontal direction overall is sufficient, and as long as the uniform processing of a plurality of substrates is not affected, the gas flow may be diffused in the vertical direction. Further, it is to be noted that expressions "approximately the same", "the same", "equivalent", and the like used in the above description are intended to encompass the substantially same.

[0091] (Other Embodiments)

While the embodiments of the present disclosure are specifically described above, the present disclosure is not limited thereto and various modifications may be made without departing from the spirit thereof.

[0092] In the above-described embodiments, the first process gas using the raw material gas and the second process gas using the reactive gas are described. However, the present disclosure is not limited thereto, and the reactive gas may be used as the first process gas and the raw material gas may be used as the second process gas. Further, a cycle in which the first process gas supply process is performed after the second process gas supply process may be performed a predetermined number of times. That is, the reactive gas may be supplied after the raw material gas is flash-supplied, and the reactive gas may be flash-supplied after the raw material gas is supplied. Furthermore, both the raw material gas and the reactive gas may be flash-supplied. That is, the reactive gas may be flash-supplied after the raw material gas is flash-supplied. The above-described effects may still be obtained in these cases.

[0093] As the raw material gas, for example, predetermined element-containing gases containing predetermined elements such as tungsten (W), titanium (Ti), molybdenum (Mo), tantalum (Ta), cobalt (Co), yttrium (Y), ruthenium (Ru), hafnium (Hf), zirconium (Zr), aluminum (Al), silicon (Si), and the like can be used. As the raw material gas, one or more of these gases can be used. As the material gas, a gas obtained by exciting these gases by plasma or the like may be used.

[0094] As the predetermined element-containing gases, for example, halogen-based predetermined element-containing gases containing a predetermined element and a halogen

element can be used. As the halogen-based predetermined element-containing gases, for example, gases containing tungsten hexachloride (WCl_6), tungsten hexafluoride (WF_6), titanium tetrachloride ($TiCl_4$), titanium tetrafluoride (TiF_4), molybdenum pentachloride ($MoCl_5$), molybdenum pentafluoride (MoF_5), molybdenum dioxide dichloride (MoO_2Cl_2), molybdenum oxide tetrachloride ($MoOCl_4$), tantalum pentachloride ($TaCl_5$), tantalum pentafluoride (TaF_5), cobalt difluoride (CoF_2), cobalt dichloride ($CoCl_2$), yttrium trifluoride (YF_3), yttrium trichloride (YCl_3), ruthenium trichloride ($RuCl_3$), ruthenium trifluoride (RuF_3), hafnium tetrachloride ($HfCl_4$), hafnium tetrafluoride (HfF_4), zirconium tetrachloride ($ZrCl_4$), zirconium tetrafluoride (ZrF_4), aluminum trichloride ($AlCl_3$), aluminum trifluoride (AlF_3), dichlorosilane (SiH_2Cl_2), 1,2-dichlorodisilane ($Si_2H_4Cl_2$), 1,1,1-trichlorodisilane ($Si_2H_3Cl_3$), 1,1,2-trichlorodisilane ($Si_2H_3Cl_3$), pentachlorodisilane (Si_2HCl_5), hexachlorodisilane (Si_2Cl_6), tetrafluorosilane (SiF_4), and the like. In addition, as the predetermined element-containing gases, for example, gases containing monosilane (SiH_4), disilane (Si_2H_6), trisilane (Si_3H_8), tetrasilane (Si_4H_{10}), and the like can be used. One or more of these gases can be used as the predetermined element-containing gases.

[0095] Further, as the predetermined element-containing gases, for example, organic predetermined element-containing gases containing a predetermined element and an organic ligand can be used. As the organic predetermined element-containing gases, for example, gases containing hexadimethylaminoditungsten ($W_2[N(CH_3)_2]_6$), bis(tert-butylimino)bis(dimethylamino)tungsten ($(t\text{-}C_4H_9NH)_2W=(Nt\text{-}C_4H_9)_2$), tetrakisethylmethylaminotitanium ($Ti[N(C_2H_5)(CH_3)]_4$), bisethylcyclopentadienylruthenium ($Ru(EtCp)_2$), biscyclopentadienylruthenium ($Ru(Cp)_2$), tetrakisethylmethylaminohafnium ($Hf[N(MeE)_t]_4$), tetrakisdiethylaminohafnium ($Hf[N(Et)_2]_4$), tetrakisdimethylaminohafnium ($Hf[N(Me)_2]_4$), trisdimethylaminocyclopentadienylhafnium ($(Cp)Hf[N(Me)_2]_3$), tetrakisethylmethylaminozirconium ($Zr[N(Me)Cp]_4$), tetrakisdiethylaminozirconium ($Zr[N(Et)_2]_4$), tetrakisdimethylaminozirconium ($Zr[N(Me)_2]_4$), trisdimethylaminocyclopentadienylzirconium ($(Cp)Zr[N(Me)_2]_3$), trimethylaluminum ($Al(CH_3)_3$), trisdimethylaminosilane ($Si[N(CH_3)_2]_3H$), and the like can be used. One or more of

these gases can be used as the predetermined element-containing gases.

[0096] The reactive gas may be, for example, gases containing a reducing gas, an oxidizing gas, a nitriding gas, a sulfiding gas, a selenizing gas, a tellurizing gas, and the like. As the reactive gas, one or more of these gases can be used. As the reactive gas, a gas obtained by exciting these gases using plasma or the like may be used. For example, in a case where one of the first process gas and the second process gas is a predetermined element-containing gas and the other is the reducing gas, a film containing a predetermined element alone can be formed on the substrate S. In addition, in a case where one of the first process gas and the second process gas is the predetermined element-containing gas and the other is any one of the oxidizing gas, the nitriding gas, the sulfiding gas, the selenizing gas, and the tellurizing gas, an oxide film of the predetermined element, a nitride film of the predetermined element, a sulfide film of the predetermined element, a selenide film of the predetermined element, or a telluride film of the predetermined element can be formed on the wafer.

[0097] As the reducing gas, for example, one or more of gases including hydrogen (H_2) gas, deuterium (D_2) gas, borane (BH_3) gas, diborane (B_2H_6) gas, carbon monoxide (CO) gas, ammonia (NH_3) gas, monosilane (SiH_4) gas, disilane (Si_2H_6) gas, trisilane (Si_3H_8) gas, monogermane (GeH_4) gas, digermane (Ge_2H_6), and the like can be used. In addition, as the reactive gas, for example, an oxidizing gas of an oxygen (O)-containing gas can be used. As the oxidizing gas, for example, one or more of gases including oxygen (O_2), ozone (O_3), water vapor (H_2O), a mixed gas of H_2 and O_2, hydrogen peroxide (H_2O_2), nitrous oxide (N_2O), and the like can be used. As the nitriding gas, for example, one or more of nitrogen-hydrogen-based gases such as ammonia (NH_3) gas, diazene (N_2H_2) gas, hydrazine (N_2H_4) gas, and N_3H_8 gas can be used. As the sulfiding gas, for example, gases containing sulfane (H_2S), disulfane (H_2S_2), diammonium sulfide ($(NH_4)_2S$), dimethyl sulfide ($(CH_3)_2S$), and the like can be used. As the sulfiding gas, for example, one or more of these gases can be used. As the selenizing gas, for example, gases containing hydrogen selenide (H_2Se), hydrogen diselenide (H_2Se_2), dimethyl selenide ($(CH_3)_2Se$), and the like can be used. As the selenizing gas, for example, one or more

of these gases can be used. As the tellurizing gas, for example, gases containing hydrogen telluride (H_2Te), dihydrogen ditelluride (H_2Te_2), dimethyltelluride ($(CH_3)_2Te$), or and the like case be used. As the tellurizing gas, one or more of these gases can be used.

[0098] As the inert gas, for example, noble gases such as helium (He), argon (Ar), neon (Ne) gases, xenon (Xe) gas, nitrogen (N_2), and the like can be used. As the inert gas, one or more of these gases can be used.

[0099] In the above-described embodiments, the case in which one tank 259 is used to flash-supply the first process gas is described, but the present disclosure is not limited thereto. That is, a plurality of tanks may be used to flash-supply the first process gas to the substrate S. By using the plurality of tanks, while the first process gas is being stored in one tank, the first process gas can be flash-supplied from another tank. In the embodiment of the present disclosure as well, the same effects as in the above-mentioned embodiments are obtained. In the embodiment of the present disclosure, throughput can be improved. In addition, the first process gas may be flash-supplied to the substrate S at least partially from the plurality of tanks at the same time. In this embodiment of the present disclosure, the same effects as in the above-described embodiments are obtained. In this embodiment of the present disclosure, the flow velocity of the first process gas in the process chamber 201 can be easily increased. This makes it easier to improve step coverage.

[0100] Further, in a case where the first process gas is flash-supplied using two or more tanks 259, the flash-supply of the first process gas from the two or more tanks 259 to the substrate S may be performed at least partially simultaneously. In this embodiment of the present disclosure, the same effects as in the above-described embodiments are obtained. In this embodiment of the present disclosure, the amount of the first process gas supplied from one tank can be reduced. This makes it easier to supply a sufficient amount of the first process gas even when the predetermined amount X is set to a large amount. Therefore, it is easier to further improve the step coverage.

[0101] Although a case in which the tank 259 is provided in the above-described first process

gas supply system 250 is described, the present disclosure is not limited thereto. That is, the second process gas supply system 260 may be provided with a tank and the second process gas may be flash-supplied. In addition, each of the first process gas supply system 250 and the second process gas supply system 260 may be provided with a tank and each of the first process gas and the second process gas may be flash-supplied. In this embodiment of the present disclosure as well, the same effects as in the above-described embodiments are obtained.

[0102] In the above-described embodiments, the amount of the first process gas remaining in the tank 259 at the end time point of the supply process S312 (the time point when the valve 254 is closed) is set to be equal to or greater than the predetermined amount X. However, the present disclosure is not limited thereto. For example, the amount of a gas containing the first process gas remaining in the tank 259 at the end time point of the supply process S312 (the time point when the valve 254 is closed) may be set to be equal to or greater than the predetermined amount X. Here, as the gas containing the first process gas, for example, a first process gas-containing gas in which the first process gas and a gas (for example, N_2 gas as the inert gas) different from the first process gas are mixed can be used. In addition, for example, the amount of the gas different from the first process gas remaining in the tank 259 at the end time point of the supply process S312 (the time point when the valve 254 is closed) may be set to be equal to or greater than the predetermined amount X. In these embodiments, the same effects as in the above-described embodiments are obtained.

[0103] In the above-described embodiments, although a case in which a film is formed on the substrate S using the first process gas and the second process gas in the film forming process performed by the substrate processing apparatus is described by way of example, the present disclosure is not limited thereto. That is, other types of gases may be used as the process gases used in the film processing process to form different types of thin films. Furthermore, this embodiment of the present disclosure can be applied even when three or more types of process gases are used.

[0104] In the above-described embodiments, the film formation process is described by way of

example as the film processing process, but the present disclosure is not limited thereto. That is, this embodiment of the present disclosure is applicable to film formation processes other than the film formation process exemplified in the above-described embodiments.

[0105] In the above-described embodiments, examples in which a film is formed using a batch-type substrate processing apparatus capable of processing a plurality of substrates at a time are described. The present disclosure is not limited to the above-described embodiments and may be suitably applied to, for example, a case in which a film is formed using a single wafer-type substrate processing apparatus capable of processing a single substrate or several substrates at a time. Further, in the above-described embodiments, examples in which a film is formed using a substrate processing apparatus including a hot-wall-type process furnace are described. The present disclosure is not limited to the above-described embodiments and may also be suitably applied to, for example, a case in which a film is formed using a substrate processing apparatus including a cold-wall-type process furnace. Even when these substrate processing apparatuses are used, it is possible to perform each process using the same processing procedures and processing conditions as in the above-described embodiments or modifications, and the same effects as in the above-described embodiments or modifications are obtained.

[0106] The above-described embodiments or modifications may be used in proper combinations. Processing procedures and processing conditions used in this case may be the same as, for example, the processing procedures and processing conditions of the above-described embodiments or modifications.

EXPLANATION OF REFERENCE NUMERALS

[0107] S: substrate,259: tank (storage)

WHAT IS CLAIMED IS:

1. A method of processing a substrate, comprising:

supplying a process gas to a substrate from at least one storage in which the process gas is stored,

wherein, in the act of supplying the process gas, an amount of a gas in the at least one storage at an end time point of the act of supplying the process gas is set to be equal to or greater than a predetermined amount.

2. The method of Claim 1, wherein, in the act of supplying the process gas, an amount of a gas including the process gas in the at least one storage at the end time point of the act of supplying the process gas is set to be equal to or greater than the predetermined amount.

3. The method of Claim 1 or 2, wherein the predetermined amount is set such that a decomposition rate of the process gas in a space in which the substrate is present is equal to or less than a predetermined decomposition rate in the act of supplying the process gas.

4. The method of any one of Claim 1 to 3, wherein the predetermined amount is set such that a flow velocity of the process gas in a space in which the substrate is present is equal to or higher than a predetermined flow velocity in the act of supplying the process gas.

5. The method of any one of Claims 1 to 4, wherein the predetermined amount is set such that a residence time of the process gas in a space in which the substrate is present is equal to or less than a predetermined residence time in the act of supplying the process gas.

6. The method of any one of Claims 1 to 5, wherein the predetermined amount is set such that a partial pressure of the process gas in a space in which the substrate is present is equal to or

higher than a predetermined partial pressure in the act of supplying the process gas.

7. The method of any one of Claims 1 to 6, wherein the at least one storage includes a plurality of storages, and

wherein the act of supplying the process gas includes supplying the process gas to the substrate from the plurality of storages.

8. The method of any one of Claims 1 to 7, wherein the act of supplying the process gas is ended after detecting that the amount of the gas in the at least one storage is equal to or greater than the predetermined amount.

9. The method of any one of Claims 1 to 8, further comprising:

storing the process gas in the at least one storage,

wherein a cycle including the act of storing the process gas and the act of supplying the process gas is performed a predetermined number of times.

10. The method of Claim 9, wherein the predetermined number of times include a plurality of times, and

wherein the act of storing the process gas includes varying at least one selected from the group of a flow rate of the process gas supplied to the at least one storage and a time for which the process gas is continuously supplied to the at least one storage at least once while the cycle is performed the plurality of times, according to an amount of the process gas in the at least one storage.

11. The method of Claim 10, wherein the act of storing the process gas includes keeping the flow rate of the process gas supplied to the at least one storage constant throughout the plurality of cycles and varying the time for which the process gas is continuously supplied to the at least

one storage at least once during the plurality of cycles.

12. The method of Claim 10, wherein the act of storing the process gas includes keeping the time for which the process gas is continuously supplied to the at least one storage constant while the cycle is performed the plurality of times and varying the flow rate of the process gas supplied to the at least one storage at least once while the cycle is performed the plurality of times.

13. A method of manufacturing a semiconductor device, comprising:

supplying a process gas to a substrate from a storage in which the process gas is stored,

wherein, in the act of supplying the process gas, an amount of gas in the storage at an end time point of the act of supplying the process gas is set to be equal to or greater than a predetermined amount.

14. A substrate processing apparatus, comprising:

a process chamber in which a substrate is arranged;

a storage configured to store a process gas therein;

a gas controller configured to be capable of controlling a supply of the process gas to the substrate from the storage; and

a controller configured to be capable of controlling the gas controller to perform:

the supply of the process gas to the substrate from the storage,

wherein an amount of a gas in the storage at an end time point of the supply of the process gas is set to be equal to or greater than a predetermined amount.

15. A program that causes, by a computer, a substrate processing apparatus to perform a process comprising:

supplying a process gas to a substrate from a storage in which the process gas is stored,

wherein, in the act of supplying the process gas, an amount of a gas in the storage at an

end time point of the act of supplying the process gas is set to be equal to or greater than a predetermined amount.