## [CLAIMS]

[Claim 1]

A method of manufacturing a semiconductor device, comprising:

modifying a surface of a substrate comprising a high aspect ratio structure whose aspect ratio is 20 or more ~~in~~to form an impurity-containing layer conformally at an inner surface of the high aspect ratio structure by performing:

(a) supplying an impurity-containing gas containing an impurity and a dilution gas into a process chamber in which the substrate is accommodated;

(b) plasma-exciting the impurity-containing gas and the dilution gas; and

(c) supplying an active species containing the impurity generated by plasma-exciting the impurity-containing gas and the dilution gas to the substrate,

wherein a flow rate ratio of the impurity-containing gas to the dilution gas is controlled in (a) such that a partial pressure of the impurity-containing gas in the process chamber is set to a predetermined partial pressure less than a partial pressure at which the impurity-containing gas forms deposits containing a polymer in the process chamber.

[Claim 2]

The method of claim 1, wherein the predetermined partial pressure is equal to or less than 0.01 Pa.

[Claim 3]

The method of claim 1, wherein the predetermined partial pressure is equal to or less than 0.002 Pa.

[Claim 4]

The method of claim 1, wherein the dilution gas comprises at least one selected from the group of a hydrogen-containing gas and a rare gas.

[Claim 5]

The method of claim 1, wherein the impurity comprises at least one selected from the group of boron, arsenic, phosphorus and gallium.

[Claim 6]

The method of claim 1, wherein the impurity-containing gas comprises at least one selected from the group of diborane gas, boron trichloride gas and boron trifluoride gas.

[Claim 7]

The method of claim 1, wherein the surface of the substrate comprises at least one selected from the group of a silicon-containing film and a silicon-containing base of the substrate.

[Claim 8]

The method of claim 7, wherein the surface of the substrate comprises at least one selected from the group of a silicon oxide film, a silicon nitride film and a silicon oxynitride film.

[Claim 9]

The method of claim 1, wherein a step coverage of the impurity-containing layer formed on the surface of the substrate is controlled by adjusting a magnitude of a high frequency power supplied to plasma-excite the impurity-containing gas and the dilution gas in (b).

[Claim 10]

The method of claim 9, wherein the step coverage of the impurity-containing layer is adjusted to be increased by reducing the magnitude of the high frequency power in (b).

[Claim 11]

The method of claim 1, further comprising

modifying a surface of the impurity-containing layer into an oxide layer after the impurity-containing layer is formed on the surface of the substrate, by performing:

(d) plasma-exciting an oxygen-containing gas; and

(e) supplying an active species containing oxygen generated by plasma-exciting the oxygen-containing gas to the surface of the substrate.

[Claim 12]

The method of claim 11, wherein ~~a high aspect ratio structure whose aspect ratio is 20 or more is provided on the surface of the substrate, and~~ the oxide layer is conformally formed with respect to ~~an~~ the inner surface of the high aspect ratio structure.

[Claim 13]

The method of claim 12, wherein a step coverage of the oxide layer formed on the inner surface of the high aspect ratio structure is equal to or greater than 70 %.

[Claim 14]

A substrate processing apparatus comprising:

a process chamber in which a substrate is accommodated;

a gas supplier configured to supply an impurity-containing gas containing an impurity and a dilution gas to the process chamber;

an exhauster configured to exhaust the process chamber;

a plasma generator configured to plasma-excite the impurity-containing gas and the dilution gas supplied to the process chamber; and

a controller configured to be capable of controlling the gas supplier, the exhauster and the plasma generator to perform:

modifying a surface of the substrate comprising a high aspect ratio structure whose aspect ratio is 20 or more ~~in~~to form an impurity-containing layer conformally at an inner surface of the high aspect ratio structure by performing:

(a) supplying the impurity-containing gas and the dilution gas into the process chamber;

(b) plasma-exciting the impurity-containing gas and the dilution gas; and

(c) supplying an active species containing the impurity generated by plasma-exciting the impurity-containing gas and the dilution gas to the substrate,

wherein the controller is further configured to be capable of controlling a flow rate ratio of the impurity-containing gas to the dilution gas in (a) such that a partial pressure of the impurity-containing gas in the process chamber is set to a predetermined partial pressure less than a partial pressure at which the impurity-containing gas forms deposits containing a polymer in the process chamber.

[Claim 15]

A program that causes, by a computer, a substrate processing apparatus to perform:

modifying a surface of a substrate comprising a high aspect ratio structure whose aspect ratio is 20 or more ~~in~~to form an impurity-containing layer conformally at an inner surface of the high aspect ratio structure by performing:

(a) supplying an impurity-containing gas containing an impurity and a dilution gas into a process chamber in which the substrate is accommodated;

(b) plasma-exciting the impurity-containing gas and the dilution gas; and

(c) supplying an active species containing the impurity generated by plasma-exciting the impurity-containing gas and the dilution gas to the substrate,

wherein a flow rate ratio of the impurity-containing gas to the dilution gas is controlled in (a) such that a partial pressure of the impurity-containing gas in the process chamber is set to a predetermined partial pressure less than a partial pressure at which the impurity-containing gas forms deposits containing a polymer in the process chamber.