## [CLAIMS]

[Claim 1]

A method of manufacturing a semiconductor device, comprising modifying ~~an~~ metal oxide film formed on a substrate by performing:

(a) supplying a reactive species containing an element of a rare gas generated by converting a gas containing the rare gas into a plasma state to the metal oxide film containing an impurity, metal and oxygen for the rare gas to break a bond between the impurity and the oxygen or the metal and a bond between the metal and the oxygen; and

(b) after (a), supplying a reactive species containing oxygen generated by converting an oxygen-containing gas different from the gas containing the rare gas into a plasma state to the metal oxide film so as to form a crystal structure of the metal oxide film.

[Claim 2]

The method of claim 1, wherein the oxygen-containing gas comprises the rare gas and oxygen, and

the reactive species containing the element of the rare gas and the reactive species containing oxygen generated by converting the oxygen-containing gas into the plasma state are supplied to the metal oxide film in (b) so as to prevent the oxygen in the metal oxide film from being inactivated.

[Claim 3]

The method of claim 2~~1~~, wherein a partial pressure ratio of the rare gas in the gas containing the rare gas is greater than that of the rare gas in the oxygen-containing gas.

[Claim 4]

The method of claim 3, wherein the partial pressure ratio of the rare gas in the

oxygen-containing gas is equal to or less than 50 %.

[Claim 5]

The method of claim 1~~4~~, wherein the oxygen-containing gas is free of the rare gas.

[Claim 6]

The method of claim 3, wherein the gas containing the rare gas comprises the rare gas alone.

[Claim 7]

The method of claim 1, wherein the oxygen-containing gas comprises oxygen and hydrogen, and

a~~the~~ reactive species containing oxygen and hydrogen generated by converting the oxygen-containing gas into the plasma state is supplied to the metal oxide film in (b).

[Claim 8]

The method of claim 1, wherein the oxygen-containing gas comprises the rare gas, oxygen and hydrogen, and

the reactive species containing the element of the rare gas and a~~the~~ reactive species containing oxygen and hydrogen generated by converting the oxygen-containing gas into the plasma state are supplied to the metal oxide film in (b).

[Claim 9]

The method of claim 8, wherein a partial pressure ratio of the rare gas in the gas containing the rare gas is greater than that of the rare gas in the oxygen-containing gas.

[Claim 10]

The method of claim 9, wherein the partial pressure ratio of the rare gas in the oxygen-containing gas is equal to or less than 50 %.

[Claim 11]

The method of claim 9, wherein the gas containing the rare gas comprises the rare gas alone.

[Claim 12]

The method of any one of claims 1 through 11, wherein the rare gas comprises helium gas.

~~[Claim 13]~~

~~The method of any one of claims 1 through 11, wherein the oxide film comprises a metal oxide film.~~

[Claim ~~14~~13]

The method of any one of claims 1 through 11, wherein stopping a supply of the gas containing the rare gas and removing a residual gas on the substrate are~~is~~ pe~~re~~formed after (a).

[Claim ~~15~~14]

The method of claim 1, wherein the rare gas is supplied to a process chamber in which the substrate is accommodated in (a) and a gas containing oxygen is supplied to the process chamber in (b) while the rare gas is continuously supplied in (b) after (a) is completed, and

the reactive species containing the element of the rare gas and the reactive species containing oxygen are generated by plasma-exciting a mixed gas of the rare gas and the gas containing oxygen so as to prevent the oxygen in the metal oxide film from being inactivated, wherein the mixed gas serves as the oxygen-containing gas.

[Claim ~~16~~15]

A substrate processing apparatus, comprising:

a process chamber in which a substrate with a metal oxide film containing an impurity, metal and oxygen formed thereon is accommodated;

a rare-gas supplier capable of supplying a gas containing a rare gas into the process chamber;

an oxygen-containing gas supplier capable of supplying an oxygen-containing gas different from the gas containing the rare gas into the process chamber;

a plasma generator capable of plasma-exciting the gas containing the rare gas and the oxygen-containing gas supplied into the process chamber; and

a controller capable of performing:

(a) controlling the rare-gas supplier and the plasma generator to supply the gas containing the rare gas into the process chamber in which the substrate is accommodated and to plasma-excite the gas containing the rare gas supplied into the process chamber for the rare gas to break a bond between the impurity and the oxygen or the metal and a bond between the metal and the oxygen; and

(b) after (a), controlling the oxygen-containing gas supplier and the plasma generator to supply the oxygen-containing gas into the process chamber in which the substrate is accommodated and to plasma-excite the oxygen-containing gas supplied into the process chamber so as to form a crystal structure of the metal oxide film.

[Claim ~~17~~16]

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

modifying a~~n~~ metal oxide film formed on a substrate by performing:

(a) supplying a reactive species containing an element of a rare gas generated by converting a gas containing the rare gas into a plasma state to the metal oxide film containing an impurity, metal and oxygen for the rare gas so as to break a bond between the impurity and the oxygen or the metal and a bond between the metal and the oxygen; and

(b) after (a), supplying a reactive species containing oxygen generated by converting an oxygen-containing gas different from the gas containing the rare gas into a plasma state to the metal oxide film so as to form a crystal structure of the metal oxide film.

[Claim 17]

The method of claim 1, wherein the impurity comprises at least one selected from the group consisting of hydrogen, carbon, nitrogen, chlorine, silicon and fluorine.

[Claim 18]

A method of manufacturing a semiconductor device, comprising:

(a) supplying a reactive species, which contains an element of a rare gas generated by converting a gas containing the rare gas into a plasma state, to a metal oxide film formed on a substrate and containing an impurity, metal and oxygen; and

(b) after (a), modifying the metal oxide film by supplying a reactive species containing oxygen generated by converting an oxygen-containing gas different from the gas containing the rare gas into a plasma state to the metal oxide film so as to reduce a leakage current.