WHAT IS CLAIMED IS:

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

(a) forming NH termination on a surface of a substrate by supplying a first reactant containing N and H to the substrate;

(b) forming a first SiN layer having SiCl termination formed on its surface by supplying $SiCl_4$ as a precursor to the substrate to react the NH termination formed on the surface of the substrate with the $SiCl_4$;

(c) forming a second SiN layer having NH termination formed on its surface by supplying a second reactant containing N and H to the substrate to react the SiCl termination formed on the surface of the first SiN layer with the second reactant; and

(d) forming a SiN film on the substrate by performing a cycle a predetermined number of times under a condition where the $SiCl_4$ is not gas-phase decomposed after performing (a), the cycle including non-simultaneously performing (b) and (c),

wherein a supply time of the $SiCl_4$ in (b) is set longer than a supply time of the second reactant in (c).

2. The method according to Claim 1, wherein the cycle is performed the predetermined number of times under a condition in which the $SiCl_4$ is not pyrolyzed.

3. The method according to Claim 1, wherein the cycle is performed the predetermined number of times under a condition in which the $SiCl_4$ does not generate an intermediate.

4. The method according to Claim 1, wherein the cycle is performed the predetermined number of times under a condition in which no gas-phase reaction occurs.

5. The method according to Claim 1, wherein (b) is performed under a condition in which

an adsorptive substitution reaction occurs between the NH termination formed on the surface of the substrate and the $SiCl_4$, and

wherein (c) is performed under a condition in which an adsorptive substitution reaction occurs between the SiCl termination formed on the surface of the first SiN layer and the second reactant.

6. The method according to Claim 1, wherein (b) is performed under a condition in which Si-N bonds are formed by bonding of Si constituting the $SiCl_4$ and N constituting the NH termination formed on the surface of the substrate, and at that time, Si-Cl bonds which are not converted into the Si-N bonds among Si-Cl bonds contained in the $SiCl_4$ are held without being broken.

7. The method according to Claim 1, wherein (b) is performed under a condition in which at least a portion of Si-Cl bonds in the $SiCl_4$ and at least a portion of N-H bonds in the NH termination formed on the surface of the substrate are broken, and Si-N bonds are formed by bonding of Si after the at least a portion of Si-Cl bonds in the $SiCl_4$ are broken and N after the at least a portion of N-H bonds in the NH termination formed on the surface of the substrate are broken, and at that time, Si-Cl bonds which are not converted into the Si-N bonds among the Si-Cl bonds in the $SiCl_4$ are held without being broken.

8. The method according to Claim 1, wherein (b) is performed under a condition in which Si constituting the $SiCl_4$ is bonded to N constituting the NH termination formed on the surface of the substrate in a state in which Cl is bonded to each of three bonds among four bonds of Si constituting the $SiCl_4$.

9. The method according to Claim 1, wherein (b) is performed under a condition in which Si after at least a portion of Si-Cl bonds in the $SiCl_4$ are broken is bonded to N after at least a portion of N-H bonds in the NH termination formed on the surface of the substrate are broken in

a state in which Cl is bonded to each of three bonds among four bonds of Si constituting the $SiCl_4$.

10. The method according to Claim 1, wherein a supply time of the first reactant in (a) is set longer than a supply time of the second reactant in (c).

11. The method according to Claim 1, wherein a supply time of the first reactant in (a) is set longer than a supply time of the $SiCl_4$ in (b).

12. The method according to Claim 1, wherein a supply time of the $SiCl_4$ in (b) is set such that an amount of an adsorptive substitution reaction occurring between the NH termination formed on the surface of the substrate and the $SiCl_4$ in a central portion of the substrate becomes substantially equal to an amount of an adsorptive substitution reaction occurring between the NH termination formed on the surface of the substrate and the $SiCl_4$ in an outer peripheral portion of the substrate.

13. The method according to Claim 1, wherein a supply time of the $SiCl_4$ in (b) is set such that a thickness of the first SiN layer formed in a central portion of the substrate becomes substantially equal to a thickness of the first SiN layer formed in an outer peripheral portion of the substrate.

14. The method according to Claim 1, wherein in (a), the first reactant is supplied to the substrate from a side of the substrate, and

wherein, in (b), the $SiCl_4$ is supplied to the substrate from the side of the substrate, and

wherein, in (c), the second reactant is supplied to the substrate from the side of the substrate.

15. The method according to Claim 1, wherein a pattern is formed on the surface of the substrate, the pattern including a recess, and

wherein the recess is a trench or a hole.

16. A substrate processing apparatus, comprising:

a process chamber in which a substrate is processed;

a reactant supply system configured to supply a first reactant containing N and H and a second reactant containing N and H to the substrate in the process chamber;

a precursor supply system configured to supply $SiCl_4$ as a precursor to the substrate in the process chamber;

a heater configured to heat the substrate in the process chamber; and

a controller configured to control the reactant supply system, the precursor supply system, and the heater so as to perform a process in the process chamber, the process comprising:

(a) forming NH termination on a surface of the substrate by supplying the first reactant to the substrate;

(b) forming a first SiN layer having SiCl termination formed on its surface by supplying the $SiCl_4$ to the substrate to react the NH termination formed on the surface of the substrate with the $SiCl_4$;

(c) forming a second SiN layer having NH termination formed on its surface by supplying the second reactant to the substrate to react the SiCl termination formed on the surface of the first SiN layer with the second reactant; and

(d) forming a SiN film on the substrate by performing a cycle a predetermined number of times under a condition where the $SiCl_4$ is not gas-phase decomposed after performing (a), the cycle including non-simultaneously performing (b) and (c),

wherein a supply time of the $SiCl_4$ in (b) is set longer than a supply time of the second reactant in (c).

17. A program stored in a computer-readable recording medium that causes, by a computer, a substrate processing apparatus to perform a process in a process chamber of the substrate processing apparatus, the process comprising:

(a) forming NH termination on a surface of a substrate by supplying a first reactant containing N and H to the substrate;

(b) forming a first SiN layer having SiCl termination formed on its surface by supplying $SiCl_4$ as a precursor to the substrate to react the NH termination formed on the surface of the substrate with the $SiCl_4$;

(c) forming a second SiN layer having NH termination formed on its surface by supplying a second reactant containing N and H to the substrate to react the SiCl termination formed on the surface of the first SiN layer with the second reactant; and

(d) forming a SiN film on the substrate by performing a cycle a predetermined number of times under a condition where the $SiCl_4$ is not gas-phase decomposed after performing (a), the cycle including non-simultaneously performing (b) and (c),

wherein a supply time of the $SiCl_4$ in (b) is set longer than a supply time of the second reactant in (c).