WHAT IS CLAIMED IS:

1. A method of processing a substrate, comprising:

(a) forming a first film having a first thickness and a carbon concentration that is within a range of 30 to 50 at% on an underlayer in a surface of a substrate by supplying a first process gas to the substrate, wherein the first film contains silicon, carbon, and nitrogen, and the first process gas includes silicon-containing gas and nitrogen-and-carbon-containing gas; and

(b) forming a second film having a second thickness on the first film by supplying a second process gas to the substrate, wherein the second film contains silicon, oxygen, and nitrogen, the second thickness is larger than the first thickness, and the second process gas includes oxidizing gas,

wherein in (b), oxygen atoms, which are derived from the oxidizing gas and diffuse from a surface of the first film toward the underlayer, are absorbed by the first film and the first film is modified.

2. The method according to Claim 1, wherein the carbon concentration is higher than a nitrogen concentration in the first film formed in (a).

3. The method according to Claim 1 or 2, wherein in (a), a silicon carbonitride film is formed as the first film by alternately supplying the silicon-containing gas and the nitrogen-and-carbon-containing gas to the substrate.

4. The method according to any one of Claims 1 to 3, wherein the silicon-containing gas includes halosilane-based gas, and the nitrogen-and-carbon-containing gas includes at least one selected from the group of amine-based gas and organic hydrazine-based gas.

5. The method according to any one of Claims 1 to 4, wherein the first thickness is in a range of 0.05 nm to 0.15 nm.

6. The method according to any one of Claims 1 to 5, wherein the second process gas includes silicon-containing gas, nitrogen-containing gas, and oxygen-containing gas, and

wherein in (b), a silicon oxycarbonitride film or a silicon oxynitride film is formed as the second film by performing a cycle one or more times, the cycle including alternately performing:

supplying the silicon-containing gas to the substrate;

supplying the nitrogen-containing gas to the substrate; and

supplying the oxygen-containing gas to the substrate.

7. The method according to Claim 6, wherein the nitrogen-containing gas includes nitrogen-and-carbon-containing gas, and

wherein in (b), a silicon oxycarbonitride film is formed as the second film.

8. The method according to Claim 7, wherein the silicon-containing gas of the first process gas includes halosilane-based gas, and the nitrogen-and-carbon-containing gas of the first process gas includes at least one selected from the group of amine-based gas and organic hydrazine-based gas.

9. The method according to Claim 6, wherein the nitrogen-containing gas includes nitrogen-and-hydrogen-containing gas, and

wherein in (b), the silicon oxynitride film is formed as the second film.

10. The method according to Claim 9, wherein the silicon-containing gas of the first process gas includes halosilane-based gas, and the nitrogen-and-hydrogen-containing gas includes hydrogen nitride-based gas.

11. The method according to any one of Claims 1 to 10, wherein in (b), at least a portion of carbon atoms contained in the first film is substituted with the oxygen atoms.

12. The method according to any one of Claims 1 to 11, wherein in (b), the first film is modified into a silicon oxycarbonitride film or a silicon oxynitride film.

13. The method according to any one of Claims 1 to 12, wherein in (b), the first film is modified into a silicon oxycarbonitride film or a silicon oxynitride film whose oxygen concentration is higher than a nitrogen concentration.

14. The method according to any one of Claims 1 to 13, wherein the first film is entirely modified into a silicon oxycarbonitride film or a silicon oxynitride film when (b) is completed.

15. The method according to any one of Claims 1 to 14, wherein in (b), the first film is modified into a silicon oxynitride film that does not contain carbon.

16. The method according to any one of Claims 1 to 15, wherein (a) and (b) are performed in a same process chamber.

17. The method according to any one of Claims 1 to 15, further comprising (c) forming a nitride film as the underlayer on the surface of the substrate before performing (a),

wherein at least (c) and (a) are sequentially performed in a first process chamber.

18. The method according to Claim 17, wherein (b) is performed in a second process chamber different from the first process chamber.

19. The method according to any one of Claims 1 to 18, further comprising (d) forming a third film having a third thickness on the second film by supplying the first process gas after performing (b),

wherein the third film contains silicon, carbon, and nitrogen, and the third thickness is smaller than the second thickness.

20. The method according to any one of Claims 1 to 19, wherein the underlayer includes at least one selected from the group of a conductive metal-element-containing film and a nitride film.

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

(a) forming a first film having a first thickness and a carbon concentration that is within a range of 30 to 50 at% on an underlayer in a surface of a substrate by supplying a first process gas to the substrate, wherein the first film contains silicon, carbon, and nitrogen, and the first process gas includes silicon-containing gas and nitrogen-and-carbon-containing gas; and

(b) forming a second film having a second thickness on the first film by supplying a second process gas to the substrate, wherein the second film contains silicon, oxygen, and nitrogen, the second thickness is larger than the first thickness, and the second process gas includes oxidizing gas,

wherein in (b), oxygen atoms, which are derived from the oxidizing gas and diffuse from a surface of the first film toward the underlayer, are absorbed by the first film and the first film is modified.

22. A substrate processing apparatus, comprising:

a process chamber in which a substrate is processed;

a first process gas supply system configured to supply a first process gas, which includes silicon-containing gas and nitrogen-and-carbon-containing gas, to the substrate in the process chamber;

a second process gas supply system configured to supply a second process gas, which includes oxidizing gas, to the substrate in the process chamber; and

a controller configured to be capable of controlling the first process gas supply system and the second process gas supply system to perform a process in the process chamber, the process comprising:

(a) forming a first film having a first thickness and a carbon concentration that is within a range of 30 to 50 at% on an underlayer in a surface of the substrate by supplying the first process gas to the substrate, wherein the first film contains silicon, carbon, and nitrogen; and

(b) forming a second film having a second thickness on the first film by supplying the second process gas to the substrate, wherein the second film contains silicon, oxygen, and nitrogen, and the second thickness is larger than the first thickness,

wherein in (b), oxygen atoms, which are derived from the oxidizing gas and diffuse from a surface of the first film toward the underlayer, are absorbed by the first film and the first film is modified.

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

(a) forming a first film having a first thickness and a carbon concentration that is within a range of 30 to 50 at% on an underlayer in a surface of a substrate by supplying a first process gas to the substrate, wherein the first film contains silicon, carbon, and nitrogen, and the first process gas includes silicon-containing gas and nitrogen-and-carbon-containing gas; and

(b) forming a second film having a second thickness on the first film by supplying a second process gas to the substrate, wherein the second film contains silicon, oxygen, and nitrogen, the second thickness is larger than the first thickness, and the second process gas includes oxidizing gas, and

wherein in (b), oxygen atoms, which are derived from the oxidizing gas and diffuse from a surface of the first film toward the underlayer, are absorbed by the first film and the first film is modified.