CLAIMS

1. A direct synthesis method of hydrogen peroxide from oxygen and hydrogen, comprising supplying a gas phase of hydrogen and oxygen or air of a starting material gas, and an aqueous phase of a reaction catalyst into a flow channel inside the reaction tube by using a reaction tube having an inner wall of a hollow tube coated with a noble-metal thin film, continuously and directly synthesizing hydrogen peroxide from the hydrogen and the oxygen inside the reaction tube under predetermined reaction temperature and reaction pressure conditions, and continuously recovering the synthesized hydrogen peroxide as hydrogen peroxide water.

2. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a reaction tube coated with a thin film constituted by one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is used as the reaction tube.

3. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a reaction tube having a noble-metal thin film coated thereon by electroless plating is used.

4. The direct synthesis method of hydrogen peroxide according to claim 1 or 3, wherein a film thickness of the noble-metal thin film is within a range of 0.5 μm to 2 μm.

5. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 4, wherein any of a corrosion-resistant tube selected from a stainless steel tube, an Inconel tube, a Hastelloy tube, and a titanium tube, or a corrosion-resistant double-wall tube selected from a stainless steel tube, an Inconel tube, and a Hastelloy tube provided with a titanium lining on an inner surface is used as the hollow tube.

6. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 4, wherein a fumed silica tube is used as the hollow tube.

7. The direct synthesis method of hydrogen peroxide according to claim 5, wherein when a thin film constituted by any one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is fabricated by electroless plating inside the reaction tube, an oxide coating film of a metal, which serves as a base, is manufactured by high-temperature

oxidation at a temperature equal to or higher than 600°C, or by supercritical hydroxylation at a temperature equal to or higher than 374°C.

8. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 7, wherein an inner diameter of the reaction tube is from 0.05 mm to 10 mm, or from 0.1 mm to 5 mm.

9. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 8, wherein the reaction temperature is within a range of 0°C to 80°C, or within a range of 5°C to 50°C, and the reaction pressure is within a range of 0.1 MPa to 50 MPa, or within a range of 0.1 MPa to 20 MPa.

10. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 9, wherein the hydrogen peroxide synthesized inside the reaction tube is continuously recovered as hydrogen peroxide water in use of water or an acid aqueous solution.

11. The direct synthesis method of hydrogen peroxide according to claim 10, wherein the water or acid aqueous solution that recovers hydrogen peroxide synthesized inside

the reaction tube includes any one or more ions from a fluoride ion, a chloride ion, a bromide ion, and an iodide ion.

12. The direct synthesis method of hydrogen peroxide according to claim 11, wherein the acidic aqueous solution includes any one or more substances from fluorine, chlorine, bromine, and iodine, and a total concentration of the substance in the acidic aqueous solution is within a range of 0.01 mg/L to 100 mg/L.

13. The direct synthesis method of hydrogen peroxide according to claim 10 or 11, wherein when the hydrogen peroxide synthesized inside the reaction tube is continuously recovered, a gas of the starting material gas phase and a liquid of the aqueous phase are mixed with a mixer, a volume flow rate ratio thereof is adjusted to form a slag flow, in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous, and the synthesized hydrogen peroxide, immediately after synthesized inside the reaction tube, is continuously recovered as hydrogen peroxide water by using the slag flow.

14. The direct synthesis method of hydrogen peroxide according to claim 13, wherein a volume mixing ratio of the gas and the liquid is 5-50 (gas) : 1 (liquid), and the slag flow is formed by making a volume flow rate ratio of the

starting material gas phase and aqueous phase to be recovered of 50 or less of the gas phase per 1 of the aqueous phase, or 10 or less of the gas phase per 1 of the aqueous phase.

15. The direct synthesis method of hydrogen peroxide according to claim 13 or 14, wherein an inner diameter of an outflow portion of the mixer that mixes the gas of the starting material gas phase and the liquid of the aqueous phase, an inner diameter of the reaction tube, and an inner diameter of a reaction tube joint are the same, and a slag flow size is not changed according to a flow channel.

16. The direct synthesis method of hydrogen peroxide according to any one of claims 13 to 15, wherein the slag flow is generated by mixing the gas of the starting material gas phase and the liquid of the aqueous phase with a micromixer while controlling a flow rate and a discharge pressure of the starting material gas phase and the aqueous phase, respectively, by using a mass flow controller for the starting material gas phase and a metering pump for the aqueous phase.

17. The direct synthesis method of hydrogen peroxide according to any one of claims 13 to 16, wherein a stable slag flow is generated by using a T-shaped micromixer as the micromixer.

18. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 13, wherein the volume mixing ratio of hydrogen and oxygen is hydrogen 10% to 33% and oxygen 90% to 67%, and the ratio (oxygen/hydrogen) of oxygen and hydrogen of the starting material gas phase is equal to or greater than 1.6, or the ratio of oxygen and hydrogen is equal to or greater than 2.0.

19. The direct synthesis method of hydrogen peroxide according to any one of claims 10 to 18, wherein the aqueous phase to be recovered from the reaction tube is recirculated to the reaction tube.

20. A device for direct synthesis of hydrogen peroxide for continuous direct synthesis of hydrogen peroxide from oxygen and hydrogen, comprising:

a reaction tube having an inner wall of a hollow tube coated with a noble-metal thin film; means for supplying a gas phase of hydrogen and oxygen or air of a starting material gas into the reaction tube; means for supplying an aqueous phase of a reaction catalyst; and means for continuously recovering synthesized hydrogen peroxide, immediately after synthesized inside the reaction tube in use of water or an acid aqueous solution.

21. The device for direct synthesis of hydrogen peroxide according to claim 20, comprising a reaction tube coated with a thin film constituted by any one or more of palladium, an alloy of palladium and gold, an alloy of palladium and silver, and a palladium porous film as the reaction tube having the inner wall of the tube coated with a noble-metal thin film by electroless plating.

22. The device for direct synthesis of hydrogen peroxide according to claim 20, wherein a reaction tube coated with the noble-metal thin film by electroless plating is used.

23. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 22, wherein any of a corrosion-resistant tube selected from a stainless steel tube, an Inconel tube, a Hastelloy tube, and a titanium tube, or a corrosion-resistant double-wall tube selected from a stainless steel tube, an Inconel tube, and a Hastelloy tube provided with a titanium lining on an inner surface is used as the hollow tube.

24. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 22, wherein a fumed silica tube is used as the hollow tube.

25. The device for direct synthesis of hydrogen peroxide according to claim 23, wherein the reaction tube is used, in which when a thin film constituted by any one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is fabricated by electroless plating inside the reaction tube, an oxide coating film of a metal, which serves as a base, is manufactured by high-temperature oxidation at a temperature equal to or higher than 600°C, or by supercritical hydroxylation at a temperature equal to or higher than 374°C.

26. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 25, comprising a mixer that forms a slag flow in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous in a flow channel entering the reaction tube.

27. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 24, wherein an inner diameter of an outflow portion of the mixer that mixes the starting material gas and water to be recovered, an inner diameter of the reaction tube, and an inner diameter of a reaction tube joint are the same.

28. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 27, comprising means for continuously recovering the hydrogen peroxide, which has been synthesized inside the reaction tube, as hydrogen peroxide water in use of water or an acid aqueous solution.

29. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 28, comprising a mass flow controller that controls the supply of the starting material gas phase and a metering pump that controls the supply of the aqueous phase, wherein

a micromixer that mixes the starting material gas phase and the aqueous phase is provided downstream of the mass flow controller and the metering pump, and a slag flow is generated in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous.

30. The device for direct synthesis of hydrogen peroxide according to any one of claims 26 to 29, wherein a stable slag flow is generated by using a T-shaped micromixer as the micromixer.

31. The device for direct synthesis of hydrogen peroxide according to any one of claims 20 to 29, comprising

recirculation means for recirculating the aqueous phase to be recovered from the reaction tube into the reaction tube.

CLAIMS

1. A direct synthesis method of hydrogen peroxide from oxygen and hydrogen, comprising:

1) supplying a gas phase of hydrogen and oxygen or air of a starting material gas, and an aqueous phase of a reaction catalyst into a flow channel inside the reaction tube by using a reaction tube having an inner wall of a hollow tube coated with a noble-metal thin film, continuously and directly synthesizing hydrogen peroxide from the hydrogen and the oxygen inside the reaction tube under predetermined reaction temperature and reaction pressure conditions, and continuously recovering the synthesized hydrogen peroxide as hydrogen peroxide water; and

2) mixing a gas of the starting material gas phase and a liquid of the aqueous phase with a mixer when the hydrogen peroxide synthesized inside the reaction tube is continuously recovered, adjusting a volume flow rate ratio

thereof to form a slag flow, in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous, and continuously recovering the synthesized hydrogen peroxide, immediately after synthesized inside the reaction tube, as hydrogen peroxide water by using the slag flow.

2. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a reaction tube coated with a thin film constituted by one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is used as the reaction tube.

3. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a reaction tube having a

noble-metal thin film coated thereon by electroless plating is used.

4. The direct synthesis method of hydrogen peroxide according to claim 1 or 3, wherein a film thickness of the noble-metal thin film is within a range of 0.5 μm to 2 μm.

5. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 4, wherein any of a corrosion-resistant tube selected from a stainless steel tube, an Inconel tube, a Hastelloy tube, and a titanium tube, or a corrosion-resistant double-wall tube selected from a stainless steel tube, an Inconel tube, and a Hastelloy tube provided with a titanium lining on an inner surface is used as the hollow tube.

6. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 4, wherein a fumed silica tube is used as the hollow tube.

7. The direct synthesis method of hydrogen peroxide according to claim 5, wherein when a thin film constituted by any one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is fabricated by electroless plating inside the reaction tube, an oxide coating film of a metal, which serves as a base, is manufactured by high-temperature oxidation at a temperature equal to or higher than 600°C, or by supercritical hydroxylation at a temperature equal to or higher than 374°C.

8. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 7, wherein an inner diameter of the reaction tube is from 0.05 mm to 10 mm, or from 0.1 mm to 5 mm.

9. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 8, wherein the reaction temperature is within a range of 0°C to 80°C, or within a range of 5°C to 50°C, and the reaction pressure is within a range of 0.1 MPa to 50 MPa, or within a range of 0.1 MPa to 20 MPa.

10. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 9, wherein the hydrogen peroxide synthesized inside the reaction tube is continuously recovered as hydrogen peroxide water in use of water or an acid aqueous solution.

11. The direct synthesis method of hydrogen peroxide according to claim 10, wherein the water or acid aqueous solution that recovers hydrogen peroxide synthesized inside the reaction tube includes any one or more ions from a fluoride ion, a chloride ion, a bromide ion, and an iodide ion.

12. The direct synthesis method of hydrogen peroxide according to claim 11, wherein the acidic aqueous solution includes any one or more substances from fluorine, chlorine, bromine, and iodine, and a total concentration of the substance in the acidic aqueous solution is within a range of 0.01 mg/L to 100 mg/L.

13. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a volume mixing ratio of the gas and the liquid is 5-50 (gas) : 1 (liquid), and the slag flow in formed by making a volume flow rate ratio of the

starting material gas phase and aqueous phase to be recovered of 50 or less of the gas phase per 1 of the aqueous phase, or 10 or less of the gas phase per 1 of the aqueous phase.

14. The direct synthesis method of hydrogen peroxide according to claim 1, wherein an inner diameter of an outflow portion of the mixer that mixes the gas of the starting material gas phase and the liquid of the aqueous phase, an inner diameter of the reaction tube, and an inner diameter of a reaction tube joint connecting the reaction tube are the same, and a slag flow size is not changed according to a flow channel.

15. The direct synthesis method of hydrogen peroxide according to claim 1, wherein the slag flow is generated by mixing the gas of the starting material gas phase and the liquid of the aqueous phase with a micromixer while

controlling a flow rate and a discharge pressure of the starting material gas phase and the aqueous phase, respectively, by using a mass flow controller for the starting material gas phase and a metering pump for the aqueous phase.

16. The direct synthesis method of hydrogen peroxide according to claim 1, wherein a stable slag flow is generated by using a T-shaped micromixer as the micromixer.

17. The direct synthesis method of hydrogen peroxide according to any one of claims 1 to 12, wherein the volume mixing ratio of hydrogen and oxygen is hydrogen 10% to 33% and oxygen 90% to 67%, and the ratio (oxygen/hydrogen) of oxygen and hydrogen of the starting material gas phase is equal to or greater than 1.6, or the ratio of oxygen and hydrogen is equal to or greater than 2.0.

18. The direct synthesis method of hydrogen peroxide according to any one of claims 10 to 17, wherein the aqueous phase to be recovered from the reaction tube is recirculated to the reaction tube.

19. A device for direct synthesis of hydrogen peroxide for continuous direct synthesis of hydrogen peroxide from oxygen and hydrogen, comprising:

1) a reaction tube having an inner wall of a hollow tube coated with a noble-metal thin film; a reaction tube joint for connecting the reaction tube; means for supplying a gas phase of hydrogen and oxygen or air of a starting material gas into a flow channel inside the reaction tube; means for supplying an aqueous phase of a reaction catalyst; and means for continuously recovering synthesized hydrogen peroxide, immediately after synthesized inside the reaction tube, in use of water or an acid aqueous solution; and

2) a mixer that forms a slag flow in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous by mixing the gas of the starting material gas phase and the liquid of the aqueous phase and adjusting a volume flow rate ratio thereof in a flow channel entering the reaction tube, wherein

3) an inner diameter of an outflow portion of the mixer that mixes the gas of the starting material gas phase and the liquid of the aqueous phase, an inner diameter of the reaction tube, and an inner diameter of the reaction tube joint are the same, and a slag flow size is not changed according to a flow channel.

20. The device for direct synthesis of hydrogen peroxide according to claim 19, comprising a reaction tube coated with a thin film constituted by any one or more of palladium, an alloy of palladium and gold, an alloy of palladium and silver, and a palladium porous film as the

reaction tube having the inner wall of the tube coated with a noble-metal thin film by electroless plating.

21. The device for direct synthesis of hydrogen peroxide according to claim 19, wherein a reaction tube coated with the noble-metal thin film by electroless plating is used.

22. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 21, wherein any of a corrosion-resistant tube selected from a stainless steel tube, an Inconel tube, a Hastelloy tube, and a titanium tube, or a corrosion-resistant double-wall tube selected from a stainless steel tube, an Inconel tube, and a Hastelloy tube provided with a titanium lining on an inner surface is used as the hollow tube.

23. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 21, wherein a fumed silica tube is used as the hollow tube.

24. The device for direct synthesis of hydrogen peroxide according to claim 22, wherein the reaction tube is used, in which when a thin film constituted by any one or more of palladium, an alloy of palladium and gold, a film having gold nanoparticles precipitated on a palladium thin film surface, an alloy of palladium and silver, and a palladium porous film is fabricated by electroless plating inside the reaction tube, an oxide coating film of a metal, which serves as a base, is manufactured by high-temperature oxidation at a temperature equal to or higher than 600°C, or by supercritical hydroxylation at a temperature equal to or higher than 374°C.

25. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 24, comprising means for continuously recovering the hydrogen peroxide, which has been synthesized inside the reaction tube, as hydrogen peroxide water in use of water or an acid aqueous solution.

26. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 25, comprising a mass flow controller that controls the supply of the starting material gas phase and a metering pump that controls the supply of the aqueous phase, wherein a micromixer that mixes the starting material gas phase and the aqueous phase is provided downstream of the mass flow controller and the metering pump, and a slag flow is generated in which phase states of the starting material gas phase and the aqueous phase are caused to be alternately continuous.

27. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 26, wherein a stable slag flow is generated by using a T-shaped micromixer as the micromixer.

28. The device for direct synthesis of hydrogen peroxide according to any one of claims 19 to 26, comprising recirculation means for recirculating the aqueous phase to be recovered from the reaction tube into the reaction tube.