which is supplied to a membrane separation unit. The membrane separation method is characterized in that the live cell count (log CFU/mL) of the water to be treated is 3 or more, and in the intermittent addition, a non-addition water supply period and an intermittent addition water supply period are repeated. During the non-addition water supply period, water is supplied without adding the combined chlorine agent. During the intermittent addition water supply period, the combined chlorine agent is added at such a concentration as to peel off a biofilm at an initial stage of biofilm formation in the non-addition water supply period, and water is supplied. The non-addition water supply period continues for 6 to 120 hours, and the intermittent addition water supply period continues for 0.5 to 40 hours. The combined chlorine agent concentration of the water to be treated during the intermittent addition water supply period is a concentration at which a total chlorine concentration becomes 0.5 to 20 mg/L.

[Reference List]

[Patent literature]

[0007] Patent literature 1: Japanese Patent Laid-Open No. 2006-263510

Patent literature 2: Japanese Patent Laid-Open No. 2010-201312

Patent literature 3: pamphlet of International Publication No. WO 2013/005787

[Summary ~~of Invention~~]

[Technical Problem]

[0008] A main object of the present ~~invention~~ disclosure is to provide a reverse osmosis membrane separation method whereby the activity of microorganisms in a biofilm is inhibited and stable water treatment can be continued over a long period of time.

active in a biofilm while adhering to the reverse osmosis membrane.

[0012] Besides, as a result of intensive studies using the incubation model, the present inventor has found that, when the combined chlorine agent containing a sulfamic acid compound (hereinafter, also referred to as "combined chlorine agent") was used in the reverse osmosis membrane containing the biofilm under short-term intermittent addition conditions, the activity of the microorganisms in the biofilm (particularly the activity of biofilm formation by the microorganisms) can be inhibited. At this time, as for the short-term intermittent addition conditions, each of the duration of the intermittent addition water supply period and the duration of the non-addition water supply period (specifically, a period during which these periods are repeated once) was shortened. In this way, the present inventor has completed the present ~~invention~~disclosure.

[0012a] In an aspect, there is provided a membrane separation method in which a membrane separation treatment is performed by an intermittent addition of a combined chlorine agent containing a sulfamic acid compound to water to be treated which is supplied to a reverse osmosis membrane separation unit comprising a reverse osmosis separation membrane,

wherein in the intermittent addition, an intermittent addition water supply period and a non-addition water supply period are repeated,

during the intermittent addition water supply period, the combined chlorine agent is added at such a concentration as to inhibit the activity of microorganisms in a biofilm to the water to be treated and the water is supplied to the reverse osmosis membrane separation unit, and

during the non-addition water supply period, the water is supplied to the reverse osmosis membrane separation unit without adding the combined chlorine agent;

the intermittent addition water supply period continues for 0.25 to 5 hours, wherein the membrane separation method is characterized in that the non-addition water supply period continues for 1 hour or more and less than 6 hours.

[0013] That is, the present disclosure~~invention~~ includes (1) to (9) as follows.

(1)

A membrane separation method in which a membrane separation treatment is performed by an intermittent addition of a combined chlorine agent containing a sulfamic acid compound to water to be treated which is supplied to a reverse osmosis membrane separation unit,

wherein in the intermittent addition, an intermittent addition water supply period and a non-addition water supply period are repeated,

during the intermittent addition water supply period, the combined chlorine agent is added at such a concentration as to inhibit the activity of microorganisms in a biofilm to the water to be treated and the water is supplied to the reverse osmosis membrane separation unit, and

during the non-addition water supply period, the water is supplied to the

(7)

The membrane separation method according to any one of (1) or (6), wherein during the intermittent addition water supply period, a composition ratio of a chlorine-based oxidizing agent to an alkali metal component in the water to be treated is 2×10^{-5} to 1×10^{-3} in terms of Cl/alkali metal (molar ratio).

(8)

The membrane separation method according to any one of (1) or (7), wherein during the intermittent addition water supply period, a ratio of Br and Cl in the water to be treated is 5×10^{-3} or less in terms of Br/Cl (molar ratio).

(9)

The membrane separation method according to any one of (1) or (8), wherein the membrane separation method is performed in a manner that a membrane surface flow velocity in a separation membrane is 4 cm/sec to 15 cm/sec.

[0014] Moreover, although Patent literature 1 discloses that the combined chlorine agent is constantly or intermittently added, in practice, in an example, the combined chlorine agent is constantly added in order to achieve the object. In addition, paragraph [0006] of Patent literature 1 describes that the bactericidal activity of chloramine is as weak as about 1/50 to 1/200 of the bactericidal activity of free chlorine. Thus, in Patent literature 1, it is usually understood that constant addition of a combined chlorine agent formed from a chlorine-based oxidizing agent and a sulfamic acid compound is required in order to prevent contamination of the permeable membrane due to the growth of microorganisms. Therefore, the short-term intermittent addition conditions in the present ~~invention~~disclosure, specifically, the shortening of each of the non-addition water supply period and the intermittent addition water supply period is not realized in Patent literature

1.

[0015] In addition, Patent literature 2 discloses in an example that in a process of continuously adding the combined chlorine agent containing a sulfamic acid compound, the combined chlorine agent is added for 24 hours at a concentration 2 to 10 times higher than a normal concentration once every 30 days. Thus, in Patent literature 2, it is usually understood that constant addition of the combined chlorine agent is required because the combined chlorine agent containing a sulfamic acid compound is added at a normal concentration and then at a high concentration. Therefore, the short-term intermittent addition conditions in the present disclosure~~invention~~, specifically, the shortening of each of the non-addition water supply period and the intermittent addition water supply period is not realized in Patent literature 2.

[0016] Besides, Patent literature 3 discloses a method in which a membrane separation treatment is performed by repeating the non-addition water supply period and the intermittent addition water supply period. During the non-addition water supply period, water is supplied without adding the combined chlorine agent, and during the intermittent addition water supply period, the combined chlorine agent is added at such a concentration as to peel off the biofilm at an initial stage of biofilm formation, and then water is supplied. As described above, in Patent literature 3, the intermittent addition conditions are set focusing on the peeling of the biofilm and the initial stage of biofilm formation, but it is hard to say that the microorganisms are active in a lively manner in the initial stage. That is, in Patent literature 3, it cannot be said that the condition setting is sufficient from the viewpoint of inhibiting the activity of the microorganisms in the biofilm (particularly the activity of biofilm formation by the microorganisms). Thus, the short-term intermittent addition conditions in the present ~~invention~~ disclosure are newly found

from an approach completely different from that in Patent literature 3. Therefore, the short-term intermittent addition conditions in the present ~~invention~~disclosure, specifically, the shortening of each of the non-addition water supply period and the intermittent addition water supply period is not realized in Patent literature 3.

[Advantageous Effects ~~of Invention~~]

[0017] According to the present ~~invention~~disclosure, it is possible to provide a reverse osmosis membrane separation method whereby the activity of microorganisms in a biofilm is inhibited and stable water treatment can be continued over a long period of time.

It should be noted that the effects described herein are not necessarily limited, and any of the effects described in the present specification may be obtained.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0018] FIG. 1 is a flow diagram of a water system showing an example of an embodiment of the present invention in which a combined chlorine agent is added simultaneously or separately in a front stage of a reverse osmosis membrane treatment system.

FIG. 2 shows changes in the activity of microorganisms (A of FIG. 2: protein amount, B of FIG. 2: live cell count, C of FIG. 2: total ATP) when an intermittent addition water supply period continues for 0.5 hour and a non-addition water supply period continues for 0 to 6.5 hours. The horizontal axis indicates a total time (h) of the intermittent addition water supply period and the non-addition water supply period, which is the total time from a moment when an agent is added.

FIG. 3 shows a schematic diagram of an apparatus configuration in a water system test.

[CLAIMS]

[Claim 1]

A membrane separation method in which a membrane separation treatment is performed by an intermittent addition of a combined chlorine agent containing a sulfamic acid compound to water to be treated which is supplied to a reverse osmosis membrane separation unit comprising a reverse osmosis separation membrane,

wherein in the intermittent addition, an intermittent addition water supply period and a non-addition water supply period are repeated,

during the intermittent addition water supply period, the combined chlorine agent is added at such a concentration as to inhibit the activity of microorganisms in a biofilm to the water to be treated and the water is supplied to the reverse osmosis membrane separation unit, and

during the non-addition water supply period, the water is supplied to the reverse osmosis membrane separation unit without adding the combined chlorine agent;

the intermittent addition water supply period continues for 0.25 to 5 hours, ~~and~~

wherein the membrane separation method is characterized in that

the non-addition water supply period continues for 1 hour or more and~~to~~ less than 6 hours.

[Claim 2]

The membrane separation method according to claim 1, wherein when one intermittent addition water supply period and one non-addition water supply period are regarded as one cycle, the one cycle continues for 1.25 to 11 hours.

[Claim 3]

The membrane separation method according to claim 1 or 2, wherein a time ratio of [the non-addition water supply period/the intermittent addition water supply period] is 0.2 to 24.

[Claim 4]

The membrane separation method according to any one of claims 1 to 3, wherein the intermittent addition water supply period continues for 0.5 to 1 hour, and/or the non-addition water supply period continues for 1 to 4 hours.

[Claim 5]

The membrane separation method according to any one of claims 1 to 4, wherein when one intermittent addition water supply period and one non-addition water supply period are regarded as one cycle, an average concentration of the combined chlorine agent in the water to be treated is 0.05 to 1.5 mg/L (as combined chlorine) per the one cycle.

[Claim 6]

The membrane separation method according to any one of claims 1 to 5, wherein the membrane separation method is performed in a state that a salinity concentration in the water to be treated is in a range of 0 to 50000 mg/L.

[Claim 7]

The membrane separation method according to any one of claims 1 to 6, wherein during the intermittent addition water supply period, a composition ratio of a chlorine-based oxidizing agent to an alkali metal component in the water to be treated is 2×10^{-5} to 1×10^{-3} in terms of Cl/alkali metal (molar ratio).

[Claim 8]

The membrane separation method according to any one of claims 1 to 7,

wherein during the intermittent addition water supply period, a ratio of Br and Cl in the water to be treated is 5×10^{-3} or less in terms of Br/Cl (molar ratio).

[Claim 9]

The membrane separation method according to any one of claims 1 to 8, wherein the membrane separation method is performed in a manner that a membrane surface flow velocity in the reverse osmosis ~~a~~ separation membrane is 4 cm/sec to 15 cm/sec.