distribution (double exponential distribution.

$$F(X) = \exp(-\exp(-(x - \lambda)/\alpha)$$

where, α and λ are scale parameter and location parameter, respectively, both of which determine the distribution, but corresponds to the minimal value. The method to estimate the maximum penetration depth of pitting using the Gumbel distribution is outlined as follows, using an analysis example for bottom plates of petroleum storage tanks.

The petroleum storage tank investigated was a 30,000-kL capacity conical roof type reservoir tank, which contains heavy oil. After 10 years of service, the tank was overhauled and a considerable degree of corrosion development was identified on the bottom plate. The pitting corrosion was formed locally and it is necessary to determine whether the pitting depth exceeds the bottom plate thickness and generates leakage, but it is not always easy to inspect the whole area of the wide bottom plate. The extreme value analysis method was used to accurately estimate pitting depth with respect to several small sections selected at random in place of inspecting the whole area. The same method was used to obtain one maximum depth for each selected section and to estimate the maximum pit depth which may possibly have occurred over the entire bottom plate area of the petroleum tank from the distribution of these small numbers of maximum pit depth. This type of technique is naturally applicable to a wide variety of equipment including heat exchanger piping and heat transfer tubes.

For details of the analysis method, see references. Software for extreme-value statistics is commercially available (EVAN: edited by Japan Society of Corrosion Engineering , Maruzen), but can run only on the PC-9801 (NEC) or equivalent personal computers. For other computer models, original software must be developed in accordance with the information available from the technical publications listed in references[63,64] or the program must be transferred.

3.2.4 Corrosion Failure Analysis Equipment

According to conventional experience, in plants operating under a corrosive environment, it is not too much to say that unexpected corrosion does occur even if careful selection of materials is carried out, and MSF seawater desalting plants are no exception. This may, to some extent, be attributed to a lack of knowledge on the part of plant designers concerning corrosion protection, and it is often attributed in part to the reasons why MSF plants must be designed with economy in mind due to their nature as industrial plants. Once a corrosion failure occurs, economical losses can be substantial. The causes must therefore be thoroughly located and preventive measures must be established.

When corrosion accidents occur, corrosion protection engineers must take the following actions:

(1) Investigation of operating conditions,
(2) Analysis of causes of corrosion accident,
(3) Experiment to reproduce the corrosion,
(4) Investigation of preventive measures.

Of these, action (2) which concerns the process of analyzing the causes of the corrosion accident is the most important and at each stage, various experimental and analytical apparatuses are required. This section describes the typical analytical apparatuses which are required when the causes of a corrosion accident must be identified using the samples taken from a plant where the corrosion accident occurred.

(1) Various Observation Equipment

Diagnosis of corrosion trouble begins with detailed observation of the plant which has generated the corrosion accident. Observation must be carried out both microscopically and microscopically.

1) Stereophotographic equipment: color photos.
2) Video microscope: observation of enlarged surface.
3) Metallographic microscope: observation of corrosion propagation features and microstructure.
4) Scanning electron microscope (SEM)* : detailed observation of surface and fractured surface.
5) Transmission electron microscope (TEM)* : observation of microstructure of metal.
6) Ancillary equipment: Ancillary equipment required for the above observation (equipment for cutting, polishing, etching, vapor deposition, etc.)
7) Corrosion measuring equipment: micrometers, dial depth gauges, and electronic balances, etc.

Of the above equipment, those underlined are recommended as necessary for the minimum requirements.
* Equipment attached with EDS (energy dispersive spectroscopy) for simultaneous composition analysis is considered convenient.

(2) Various Analytical Equipment

In determining corrosion causes, various analysis results of the chemistry of the working environment and applied materials, as well as the analysis of the chemical compositions of corrosion products and surface deposits, together with an analysis of the state, frequently provide useful information. In particular, not only principal components of the process fluid but also impurities in the chemistry of the working environment (ex. Cl−ion) often have serious effects on corrosion.

1) ICP analyzer
2) ICP−MS analyzer
3) Fluorescent X−ray analyzer
4) Emission spectrograph
5) Atomic absorption−photometer
6) Gas chromatography
7) Ion chromatography
8) Liquid chromatography
9) Infrared spectrophotometer (including Fourier transformation)
10) Laser Raman spectrophotometer
11) Nuclear magnetic resonance equipment
12) X−ray micro analyzer (EPMA)
13) X−ray diffractometer (XRD)
14) Auger electron spectrophotometer (AES)
15) X−ray photo−electric spectrophotometer (XPS)
16) Secondary ion mass analyzer (SIMS)
17) Differential thermal analyzer
18) Image processor

The underlined equipment is recommended as a minimum requirement.

(3) Corrosion Test Equipment on Laboratory Level

To identify causes of corrosion accident and investigate preventive measures, reproduction experiments are frequently carried out, but for the corrosion test equipment required for these reproduction experiments, it is recommended that suitable equipment be selected or that modification of the equipment listed in Section 2.1 be carried out, depending on the purposes.

3.3 Summary of Results

Test equipment to evaluate the corrosion resistance of materials for MSF seawater desalting plants, in particular, metallic materials, were roughly classified into

(1) basic corrosion test equipment on the laboratory level,

(2) corrosion monitoring equipment, and
(3) large-scale corrosion test equipment on the production line level.

The current technical level was summarized by a literature search, and the corrosion-testing and analytical equipment recommended for installation in SWCC laboratories were proposed. It is essential for SWCC with abundant operating experience of seawater desalting plants over many years to establish a testing and research system which enables data accumulation that will contribute to the estimation of life as well as to proposing measures to extend the life. This should be carried out on basis of detailed investigation results of the corrosion features of long-operated seawater desalting plants.

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4.3 R-1 Sterilization

RO DESALINATION
LITERATURE SURVEY NO. 3, R-1

STERILIZATION

JULY 1992

By

Sugimoto Yukinobu
JAPAN INTERNATIONAL COOPERATION AGENCY

And

Hassan Munshi
SALINE WATER CONVERSION CORPORATION

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1. Introduction

For the past several decades, the MSF flash process has been the dominant desalination method used for obtaining drinking water by sea water desalination. However, recently the RO process is playing more and more important role in sea water desalination because of simplicity and less energy consumption more. In the RO process, one of the problems encountered with RO sea water desalination is the occurrence of membrane bacterial fouling. When the RO membranes are fouled by bacteria, in general, the flux decreases and the salt rejection rate also falls. An example of this is the acetyl cellulose type membranes. They are attacked by various kinds of micro-organisms. Certain membranes, even if they are not attacked, are slimed by the proliferation of micro-organisms which cause membrane blockage due to their adhesion to the membrane surfaces. Moveover, this process occurs quite suddenly and over a short period of time thus it becomes essential to sterilize the feed. Bacterial fouling can be stopped or reduced by feed disinjection by chlorine, the SBS(Sodium Bisulphate) and the copper sulphate. Each of those disinfection methods have good and bad points. As it is difficult to discriminate among them, in the Middle East region, the most suitable method is selected by comparative examination of the technological details, tinged with price considerations.

In the present survey, the outline, principle equipment and effect of the above-mentioned processes will be reviewed including the economical viewpoint.

2. Methods

This study is limited to the literature survey. The experimental study is not to be executed. The literature retrieval was based on JOIS, DIALOG and the open materials.

3. Results

3.1 *Identification of Types and Number of Bacteria in Water:*

Introduction :

Organic bio-fouling becomes more recognized as a major potential problem in RO plants, so, more efforts will be needed to measure, analyze and monitor these parameters which should lead to the development of both practical and effective operational monitoring and control strategies which are generally lacking at the recent time.

Bio-fouling is a problem of bacterial/algal/fungal/yeast growth by attachment on to suitable subtract. Surfaces like filter media and RO membranes whereas organic fouling can be described on the deposition on these surfaces of the waste and/or decay products of biological activity within the system.

Microorganisms-what are they?

- Microscopic in size;
- Unicellular To Filamentous;
- Attached or free-living
- Types
    1. Bacteria [E.coli]
    2. Fungi [Yeast]
    3. Algal [Diatoms]

1. Bacteria :

- Procaryotic
- Basic shape
    - Rod (Bacillus)
    - Spherical (Coccus)
    - Spiral

- Oxygen Requirements:
  - Aerobic
  - Anaerobic
  - Facultative

- Resistant Forms
  - Capsules (Glycocalyx)
  - Spores
  - Cysts

2. **Fungi**

I Filamentous (Mold)
  - Eucaryotic
  - Non-photosynthetic
    (Lack Chlorophyll)
  - Coenocytic or Acellular
  - Prefer growth on a substrate in Aquatic environments
  - Reproduction (propagules)
    - Hyphal Fragmentation
    - Spores

II. Yeast
  - Eucaryotic
  - Nonphotosynthetic
  - Typically Unicellular
  - Avoid shape
  - Growth on media similar to bacteria
  - Reproduce by budding

3. **Algae**

I. Blue-Green "Algae"
  - Procaryotic
  - Unicellular, Colonial Aggregates or filamentous

- Photosynthetic (Chlorophyll) or Hetrotrophic
- Reproduction by fission
- Associated with Algal ((Blooms))

II. Diatoms
- Eucaryotic
- Unicellular, colonial aggregates or filamentous
- Photosynthetic (Chlorophyll) or Hetrotrophic
- Silicon dioxide trustrule
- Sexual reproduction
- Associated with Algal ((Blooms))

The importance of monitoring and control of microbiological activity concentration, growth and reproduction rate of microorganism in seawater RO plants using chlorine may not be fully appreciated. For seawater RO plants, various factors which are site dependent and which affect biological activity must be recognized.

The degree of microbiological activity present in the raw seawater will determine the pretreatment requirements necessary for microbiological control in seawater RO plants. The concentration and types of microorganism depend on the source of the seawater. Generally, water from seawells has significantly lower biological activity than surface seawater. Furthermore, deep surface seawater usually has less activity than shallow shore line sea water. In addition, the biological activity can very considerably from one site to another even at a single site from season to season.

Biological activity is dependent on site specific factors such as :
- Temperature
- pH
- Organic and inorganic nutrients
- Oxygen availability
- Sunlight
- Pollution and/or surface water runoff.

Temperature is one of the most important factors that influences microbial activity. Although microoganisms are present and can grow at temperatures ranging from 0°C to 100°C, each microorganism has an optimum temperature for growth and reproduction, i.e., each microorganism will only grow within a certain temperature range. The temperature of the seawater significantly affects the types of microorganisms which are present in seawater and their activity (growth and reproduction rate).

Microorganisms are found and can grow in both acidic and alkaline waters. Most of the major groups of bacteria found in seawater are most active at neutral to slightly alkaline conditions (pH-7-9). In general, these microorganisms cannot tolerate extreme acidic ($pH<2$) or extreme alkaline ($pH>11$) conditions.

The biological activity is also dependent on the concentration of both organic and inorganic nutrients. Open ocean seawater with low concentration of organics can support growth but the microorganisms tend to aggregate and attach themselves to surfaces rather than grow freely. By contrast the shore line waters, subject to land run off and pollution, are usually rich in organics, and consequently, bacteria tend to be more abundant and grow freely. Phytoplankton also are abundant. The bacterial activity will especially be enhanced because of the elevated concentration of dissolved oxygen and particulate organics.

Some types of microorganisms (like phytoplankton) can thrive on certain inorganic compounds ammonia, nitrates and phosphates. Thus, the biological activity in seawater can be significantly increased when these inorganic nutrients, usually associated with pollution and surface water, are present.

The majority of active microorganisms in surface seawater require oxygen for survival and growth and, therefore, are aerobic. Because many bacteria can grow with or without oxygen (facultative anaerobes), the depletion of oxygen from seawater does not ensure reduced biological activity.

Sunlight is necessary for the growth and activity of phytoplankton and photosynthetic bacteria in seawater. Significant seasonal increases in phytoplankton activity (i.e. blooms) are related to the increased intensity of sunlight, availability of nutrients, and stability of the surface seawater layers. In temperate water of North America these blooms occur in early Spring (March) and again in the fall (September).

Microbiological Analyses

Microbiological analyses are used to determine the type and the concentration or number of microorganisms in the seawater. Although numerous Microbiological techniques are available for the analysis of water and waste water, it is critical that the appropriate ones be used for seawater.

Samples for Microbiological analysis must be collected in clean, sterile containers. These containers can be either glass or plastic bottles or polyethylene bags.

The Microscopic and culturing techniques recommended for the RO biomonitoring program are :

- Acridine orange direct count;
- INT direct microscopic technique;
- Standard serial dilution plating;
- Membrane filtration;
- Anaerobe enumeration with API broth.

Best Method for Microbiological Analyses

The best method is direct microscopy using either INT or acridine orange. The major drawback of cultural techniques is that they take too long (i.e. upto 9 weeks in most cases) for the final results.

Chlorination

Surface seawater is chlorinated at the intake structure. Whether the chlorine is added as a gas, a solution NaOC or $Ca(OCl)_2$ or from an electrolytic chlorine generation, the chemical reaction that occur in the seawater are the same. When chlorine is added to seawater, most of the HOCl will react with the high (60-80mg/L) concentration of bromide ion in the seawater to form HOBr. This reaction occurs fairly rapidly. For the sake of simplicity the term "chlorine" will be used to discuss the various reactions but in actuality "chlorine" is present mainly in the form of HOBr with some HOCl.

Chlorine will rapidly react with such species as F^{++}, NO^-_2, H_2S and some organic compounds. The amount of chlorine consumed by these reactions is called the chlorine demand. If ammonia or other nitrogenous material is present, chloramines will form which will decompose as more chlorine is added. Eventually, a break point will be reached where free chlorine residuals (HOBr + HOCl) will start to form. All of these reactions are illustrated in Figure 1. Thus, when chlorine is added to seawater, HOCl, HOBr, chloramines and bromamines may be present, if ammonia or other nitrogenous material is not present, the chloramines and bromamines will not form. But chlorine demand will still be present. Seawater that is relatively clear will usually have a chlorine demand of less than 2 to 3 mg/L, and a chlorine break point will not be observed.

Since free chlorine (HOCl + HOBr) is the main disinfecting agent, sufficient chlorine must be added to exceed the chlorine demand and, if present, the chlorination break point. The degree of disinfection is dependent on the :

- Types of organism present,
- Temperature,
- pH
- Contact time - and
- Concentration of free chlorine

Hypochlorous (HOCl) and hypobromous (HOBr) acid are in equilibrium with hypochlorite and hypobromite:

$$HOCl \qquad H^+ + OCl^+$$
$$HOBr \qquad H^+ + OBr^-$$

Dechlorination

Since some of the RO membranes are affected by chlorine, the chlorine must be completely removed before the seawater enters the permeators. Based on economics, $NaHSO_3$ is the best dechlorination agent reacts instantaneously and quantitatively with both HOCl and HOBr.

$$NaHSO_3 + HOCl \text{ ----> } NaHSO_4 + HCl$$
$$NaHSO_3 + HOBr \text{ ----> } NaHSO_4 + HBr$$

If for any reason, the free chlorine is not completely removed before it enters the permeator, the chlorine will react with the polymer causing its degradation. The reaction will decrease the RO product flow and overall RO performance. The loss in product flow from chlorine exposure is irreversible.

Chloramines and bromamines if present, may not be completely removed by the additional of $NaHSO_3$.

$NaHSO_3$ Shock Treatment

The efficiency of $NaHSO_3$ as a biocide against marine microorganisms, depends on exposure time, concentration of the $NaHSO_3$ and the types of microorganisms present. In general, the higher the concentration and the longer, the exposure time, the greater will be the degree of disinfection using a 30-minute exposure time, the effect of $NaHSO_3$ concentration on the degree of disinfection for seawater is given in Table 3.1.1.

Table 3.1.1

Effect of $NaHSO_3$ Concentration on Degree of Disinfection for seawater

Concentration of $NaHSO_3$ (mg/L)	% Kill[a,b]
200	96
500	99
750	99.9
1000	99.9

a = Exposure time = 30 minutes
b = Serial dilution plating technique with Marine Agar 22/6 used to measure concentration of microorganisms in Atlantic ocean seawater.

In the laboratory studies have demonstrated that even hardy, aerobic marine bacteria isolated from the high TDS (13,000 mg/L) brackish water can be significantly inhibited by $NaHSO_3$ (Table 3.1.2).

Table 3.1.2

Disinfection ability of $NaHSO_3$ for Hardy Aerobic Marine Bacteria

Concentration of $NaHSO_3$ (mg/L)	Exposure time (Hrs)	% Kill[a,b]
500	0.5	17
	1.0	31
	2.0	45
	4.0	74
	0.5	54
750	1.0	57
	2.0	63
	4.0	75

a = Bacteria isolated from 13000 mg/L TDS brackish water.
b = Serial dilution.

In the Ras Abu Jarjour RO Plant Bahrain, they found out that:

1. The series of experiments and trials in the plant have shown positive results such as improved micronguard filter and also RO membrane performance by preventing aerobic bacteria contamination entering the feed water from the SHMP [sodium hexameta phosphate] tanks and dosing line.

2. Optimum concentration of SBS to be added to the SHMP tanks is found to be 0.25% and this concentration is found to control bacteria and does not affect SHMP reversion on the phosphate.

3. Quite recently, RO performance started again to decline even though addition of SBS into SHMP was continued. At the moment, this is understood as to reduce peeling of algae from the GRP pipes causing photosynthetic activity within. The GRP pipes were painted in the past and now are due for repairs. It should also be considered that even passage of a small amount of light through the GRP pipe gradually increases biological growth in the pipe system.

Identification of Foulants in Ras Abu Jarjur RO plant : Sulfate reducing bacteria (SRB)

The plant draws its feed water from 15 deep wells. The type of water is never sterile Bacterial species tend to utilize whatever nutrient is available in order to survive. If oxygen is not available, oxygen- rich anions like SO_4 are utilized as an energy source. SRB reduce it anaerobically to sulfides. Thus giving the characteristic rotten egg smell.

Numerous tests were done to identify this bacteria group. A method, number 38, prepared according to the American Petroleum Institute (API) proved effective in culturing SRB.

The raw feed water proved positive for sulfate reducing bacteria at a population of 1-10 Cells/ml.

Algae Growth due to Light Intrusion

The piping materials used mostly in plant processes are made of GRP. From tests conducted, it was found that this material was transparent enough to let ordinary light pass through.

The plant water contains enough nutrients which together with sunlight, allows photosynthetic activity to produce massive algal growth, found on the inner walls of the collector, pipes, dual media filter beds and other areas where transparent GRP materials are used.

This led to the conclusion that if the situation was left unattended, serious problems could happen, Algae and their associated bacteria grow rapidly and they quickly clog the filters.

Embedded Bacteria in the Permeators

For the complete removal of embedded biological foulants in RO permeators, proper cleaning procedures were required to be established considering the particular nature of the fouling materials.

Aerobic Bacteria from SHMP

Sodium hexameta phosphate (SHMP) a well established antiscalant in the water industry is dosed at 7 mg/L after carbon filters to prevent sulfate scaling. In R.A.J RO plant the

SHMP was dissolved in the dechlorinated service water in different concentrations. The results are shown in Figure 3.1.1.

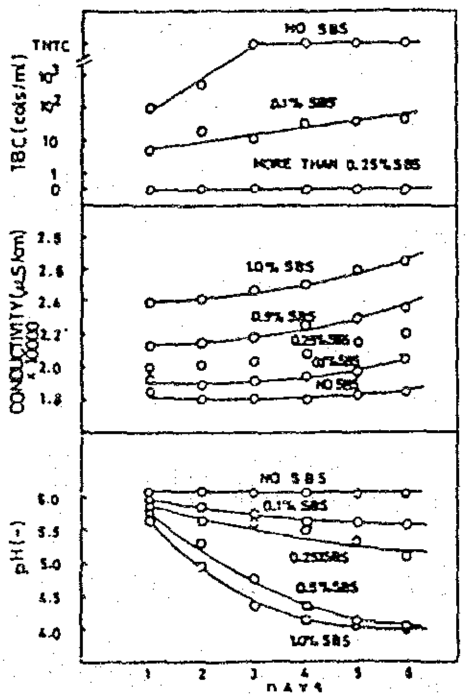


Figure 3.1.1 Behavior of SHMP in Service Water

Laboratory studies were continued to confirm SBS effect to sterilize SHMP in different concentrations and investigate any unknown side effects which might occur in SHMP tanks.

Various concentrations of SBS were added upto 7% SHMP solution to simulate SHMP tanks and analysed for free phosphate, TBC, pH and Ec during 6 days. The results are shown in the Figure 3.1.2.

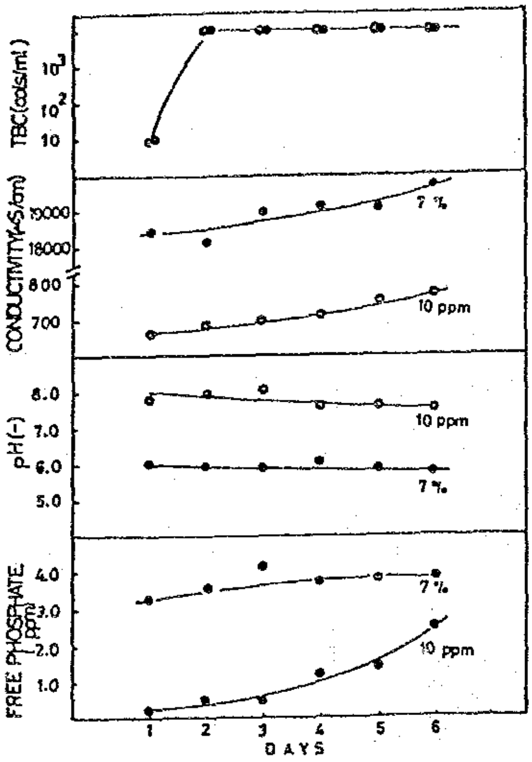


Figure 3.1.2 **Effect of SBS in SHMP Solution**

- Low concentration, less than 0.1% if SBS could not sterilize SHMP solution.

- Minimum 0.2% of SBS is found to be required to keep SHMP in a sterilized condition.

- High concentration of SBS reduced pH within a few days it reached to pH 4 within 5 to 6 days and conductivity is also found to increase rapidly. It is supposed that hydrolysis of SHMP occurred more rapidly in higher concentration of SBS.

3.2 Sterilization with Chlorine

3.2.1 Overview

(1) Sterilization effect of chlorine

Before seawater RO process was developed and industrialized, chlorine has been the major method for sterilization of drinking water taken from rivers and lakes. Chlorine treatment method should not be regarded as the last line of defense against diseases contained in water, but as a perfect solution for sterilization[6]. In case of hypochlorite, if 0.1 ppm is added to well water, it will change from rH28.2 to rH34.5 at pH7.4. However, this is considered effective in killing algae and germs which cause disease[7]. The residual chlorine concentration is considered as an important control value for daily water supply at water purification plants in Japan and elsewhere.

(2) Advantages of chlorine

The positive aspects of chlorine is that it is highly effective against germs and microorganisms, and sterilizes very quickly. Also, each of the various chlorine injection equipment and the large plant facilities can be purchased on a commercial basis, and injection can be made continuous[8].

(3) Disadvantages of chlorine

On the other hand, as chlorine is toxic, it is troublesome to handle. In regards to algae, its effectiveness drops in comparison to copper sulfate (approx. 1/5 effectiveness). Contacting with substance that is easily oxidized, or exposed to sunlight, it disappears in a short period[9]. Also, the free chlorine reacts with organic matter in fluid under treatment, and is widely known to produce the cancerating substance, trihalomethane[10,71]. There is also need to take precautions on the concentration and the method of use.

(4) Chlorine resistance property of reverse osmosis membrane.

Sterilization with chlorine causes deterioration of membrane and degradation of the performance[11] of all reverse osmosis membranes, except for RO membranes made of acetate cellulose acetate. Consequently, when chlorine is used, for RO membranes other than cellulose acetate, there is a need of dechlorination of pretreated raw-water before entering into the membrane[12]. Also, even with the cellulose acetate membrane, there is a need to pay close attention to the acetyl value, and the chlorine concentration[13].

There are many reports on sterilization of many seawater RO desalination plants using the membrane, studying their pretreatment processes[69] and comparing their performance. Regardless whether the disinfection are continuous additives or intermittent additives, these conclusion of these reports is that sterilization by chlorine is the most cost-effective and is the most effective sterilization method.

3.2.2 Principles

(1) Physical and chemical properties of chlorine[14]

State: Normal state is yellow green gas
Odor: A stimulating odor
Toxicity: Toxic
Weight: 2.5 times weight of air
Gas pressure: Approx. 6 atm at 0°C
Solubility: Dissolves slightly in water, maintains yellow color. At 9°C, 1% dissolves.
Crystals: In a saturated water solution under 9°C, light yellow hydrocholride crystals form.
Handling State: Can be handled as liquid chlorine, bleaching powder, or sodium hypochlorite.

(2) Functions as a Disinfectant

As chlorine is highly effective against germs and other organisms, it is used as a disinfectant. In particular, it is widely used for sterilization in water systems. Chlorine treatment is an important and vital process in the water purification process.

Chlorine is also used for the oxidization of iron, manganese, and hydrogen sulfide in raw water and also for the removal of the oxides. It is often used for deodorizing and decomposing organic matter[15]. Negative aspects have also been reported[17]. When chlorine is used for the raw water containing hydrogen sulfide, serious colloid fouling will occur in the RO membrane. Also as it is highly oxidizing, when there is humic acid present, depending on the conditions of the decomposed organic matter, the chlorine degraded humic acid becomes a rich nutrient source for bacteria. Furthermore, it has also been reported that when seawater is sterilized with chlorine, haloform such as brominated methane is produced, which is hazardous to health[72]. Also, chlorine has other peculiar properties which means that it has to be used with care[18].

(3) Chemical equlibrium of chlorine aqueous solution

Chlorine dissolved in water chemically reacts with water, becomes hypochlorous acid (HOCl) and hydrochloric acid. The hypochlorous acid is furthermore decomposed into an hypochlorous acid ion (ClO^-) and hydrogen ion.

These reactions are reversible, and depending on the pH and the temperature of the water, its state changes as shown in Fig. 3.1[19]. When pH is 5 and under, the chlorine exists as a chlorine molecule. However, when the pH is 6 and under, a hypochlorite acid ion appears. Furthermore, from around 7 pH, the hypochlorous acid rapidly decreases, and when pH is 7.5 and under, the hypochlorous acid ion gradually becomes the strongest. When the pH is near 10, all become hypochlorous acid ions[20]. This characteristic curve fluctuates with the parameter of the temperature; with increase in temperature, decreases the amount of hypochlorous acid.

(4) Definition of free chlorine and its sterilization property

Normally, chlorine sterilization is conducted in the neutral range, therefore treated water contains both hypochlorous acid and hypochlorous acid ions. Chlorine which exists in water as both hypochlorous acid and as a hypochlorous acid ion, is called free chlorine[21]. This free chlorine has the strongest sterilizing property and oxidizing property(see Fig.3.2.1).

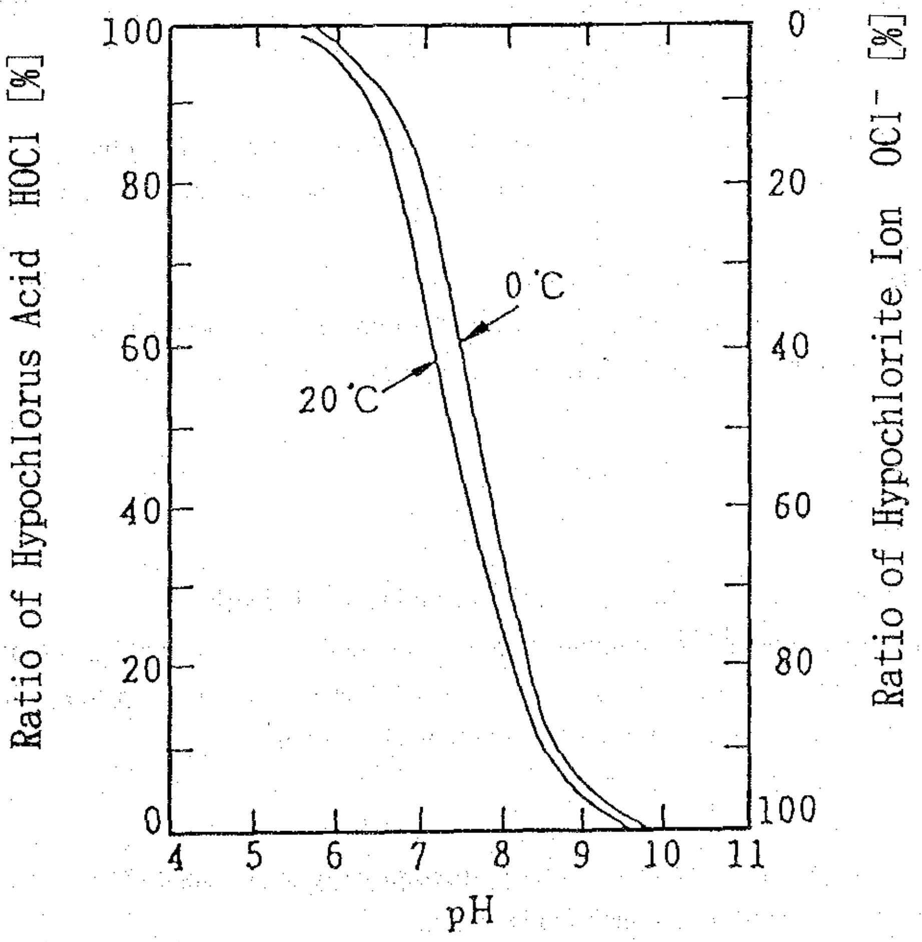


Fig. 3.2.1 Relationship of ratio of hypochlorus acid and hypochlorite ion for various pH[19]

(5) Definition of bonded chlorine and its sterilization property

If ammonium compounds are in water or added to water, the residual chlorine combines with these compounds to become chloramine. It also reacts forming bonds with organic nitrogen compounds to form a chlorine derivative compounds. In this way, chlorine in the water which combines with ammonia and organic nitrogen compounds is called bonded chlorine. Bonded chlorine has sterilization property and oxidizing property even if much weaker than those of free chlorine[22]. Also according to another report[23], though its sterilization property is not as strong as free chlorine, it does possess sterlization powers. From the point of suppressing aftergrowth of bacteria in treated water, it is much superior to chlorine.

Chloramine is created in reaction with ammonia. The produced substance changes according to the following composition proportions.

$2NH_3 + Cl_2 = NH_2Cl + NH_4Cl$: Monochloramine
$3NH_3 + 2Cl_2 = NH_2Cl + 2NH_4Cl$: Dichloramine
$4NH_3 + 3Cl_2 = NCl + 3NH_4Cl$: Trichloramine

Prior to the above reaction, chlorine first of all changes to hypochlorous acid. Under conditions with rapid mixing and a lot of ammonium ions, the following reaction occurs[24]:

$pH > 8.5$: Only monochloramine is produced.
$8.5 > pH > 5$: A mixture of monochloramine and dichloramine. The proportion of components is determined by the pH value. In the case of pH 7, ratio is 1:1.
$5 > pH > 4.4$: Only dichloramine is produced.
$4.4 > pH$: Trichloramine.

(6) Relationship between the amount of added chlorine and residual chlorine[25].

The relationship between the amount of chlorine added into water, and the amount of residual chlorine detected is shown in Fig. 3.2.2. The relation indicates three patterns Type I, Type II, and Type III depending on the properties of the water treated.

Type I

When chlorine in treated water is not used, theoretically the residual chlorine must be detected as increasing in proportion to the added amount.

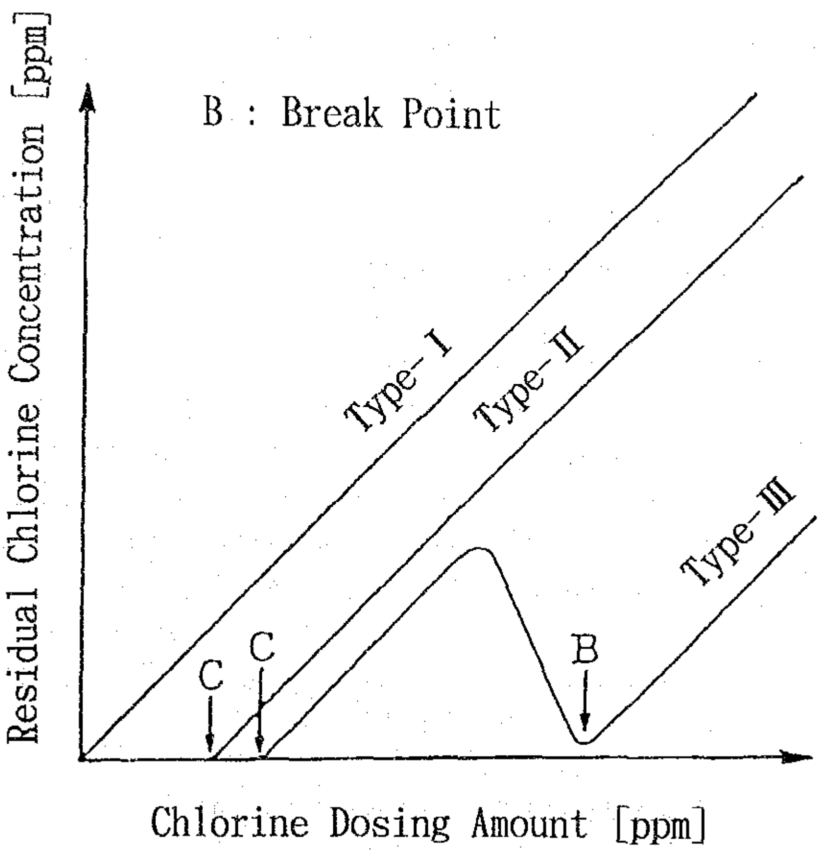


Fig. 3.2.2 Relationship of chlorine dosing amount and residual chlorine concentration[25]

Type II

The above Type I is the ideal case. Actual treated water shows the Type II pattern. Up to the point of amount C, in the induction period, the residual chlorine does not appear. However, when point C is exceeded, it is detected in proportion to the added amount. The reason for this behavior is because there are substances in the water which consume the chlorine. The substances which consume chlorine are ammonia (nitrous acids), iron, manganese, hydrogen sulfide, organic substances, and living organisms.

Type III

Originally, residual chlorine is produced in the same pattern as Type II, and it increases at the beginning. However, when a certain level is reached, it rapidly drops close to zero. However, if more chlorine is added, the residual chlorine will increase in proportion to the added amount. This is a phenomenon that can be seen in water which contains a slightly large amount of ammonia compounds and organic nitrogen compounds. The first peak, point P, is the bonded residual chlorine which has combined with the contained compounds. The chlorine which increases gradually after point B is the free chlorine.

(7) Chlorine consumption volume and chlorine demand volume[26]

Before we discuss the actual amount of chlorine required for achieving sterilization, the following terms will be defined.

Chlorine consumption volume: The chlorine consumption volume is the amount of chlorine added up to point C where the chlorine begins to be residual.

Chlorine demand volume: The difference between the amount of chlorine added into water to be treated and the amount of chlorine residual in the water after a stipulated period of time (in the case of city water, 1 hour). In reference to Type II and Type III, the chlorine demand volume is the amount of chlorine added up to point C in Type II where free residual chlorine begins to be detected, and up to point B in Type III.

Type III Point B: This is particularly called the break point.
The method of adding chlorine beyond the break point until free residual chlorine is detected, is called chlorine treatment at break point. The amount of chlorine required for break point chlorine treatment is quite large, approximately ten times the amount of ammonium nitrogen. However, its oxidizing property and sterilization property is great, and it is highly effective for sterilizing, oxidizing and deodorizing contaminated water[27]. This break point is the practical sterilization point, and with further dechlorination treatment, water with no color, odor, and taste is obtainable. It is reported that knowing the break point addition volume is useful[29].

3.2.3 Effect of Chlorine Sterilization

(1) Conditions for chlorine treatment and sterilization property

Sterilization property of chlorine greatly changes according to the chemical structure of the residual chlorine, the water temperature, and the pH. Appropriate parameters must therefore be selected.

1) Comparison of free chlorine and bonded chlorine

Required amount to achieve same sterilization power: bonded chlorine = 25 times amount of free chlorine

Required contact time to produce the same effect with the same volume: bonded chlorine = 100 times that of free chlorine.

2) Effect of water pH on free residual chlorine

pH 6.0–8.0: 0.2 ppm, complete sterilization with 10 minutes of contact.
pH 8.0–9.0: 0.4 ppm+, 10 minutes contact required.
pH 9.0–10.0: 0.8 ppm+, 10 minutes contact required.
pH 10.0+ : Add 1.0 ppm+ or 4 hours contact required.

(2) Standard chlorine ratio for various living organisms[30].

Table 3.2.1 shows the standard chlorine addition rates for various living organisms. In actuality this is a standard, and can be increased or decreased depending on the existence of decomposable organic material, ammonia, oxidizable inorganic matter in the water to be treated and whether it is exposed to sunlight. As these components consume chlorine, there is a need to increase the chlorine accordingly. The amount is estimated from the chlorine demand volume of the water to be treated.

(3) Determining chlorine addition volume[31]

The chlorine addition volume must be the total of 1)–3), as follows:

1) Chlorine demand volume of water.
2) Amount of chlorine consumed by facilities in contact with water.
3) Amount of chlorine required for sterilization and algae extermination.

Item 1) can be determined by experimentation, and item 3) can be determined from Table 3.1.1. However, as item 2) is difficult to determine by prior experimentation and calculation, it can only be estimated. In actuality, the added amount is adjusted according to the amount of residual chlorine.

(4) Chlorine–resistance of RO membrane, and methods of using chlorine

In seawater desalination plants which use RO membranes, and when chlorine is used as the disinfectant, the amount to add, the concentration and the method of application cannot be

Table 3.2.1 Standard dosing rate as sterilization[31]

Organisms		Chlorine(Cl_2) 〔ppm〕
sulfur bacteria	*Beggiatoa*	0.50
	Thiothrix	0.50 ～ 1.00
Iron bacteria	*Crenothrix*	0.50
	Gallionella	0.25
	Sphaerotilus	0.25
Fungi	*Leptomitus*	—
	Saprolegnia	—
Cyanophyceae	*Anabaena*	0.50 ～ 1.00
	Aphanizomenon	0.50 ～ 1.00
	Oscillatoria	1.10
	Phormidium	3.00
	Polycystis	1.00
Diatoms	*Achnanthes*	2.00 ～ 3.00
	Asterionella	0.50
	Attheya	
	Cyclotella	1.00
	Fragilaria	2.00
	Melosira	0.50 ～ 2.00
	Navicula	
	Nitzschia	
	Rhizosolenia	
	Stephanodiscus	
	Synedra	
	Tabellaria	
Green algae	*Ankistrodesmus*	
	Chlamydomonas	
	Closterium	
	Coccomyxa	2.50 ～ 3.00
	Cosmarium	1.50 ～ 2.00
	Draparnaldia	
	Eudorina	
	Gloeocystis	
	Hydrodictyon	
	Microspora	
	Palmella	2.50 ～ 3.00
	Scenedesmus	
	Sphaerocystis	
	Spirogyra	0.70 ～ 1.50
	Staurastrum	
	Tetraspora	1.00 ～ 1.50
	Ulothrix	
	Volvox	0.30 ～ 1.00
	Zygnema	
Golden-brown algae	*Dinobryon*	0.30 ～ 1.00
	Mallomonas	
	Synura	0.30 ～ 1.00
	Uroglenopsis	0.30 ～ 1.00
	Ceratium	0.30 ～ 1.00
	Peridinium	
Crustacea	*Cyclops*	0.30 ～ 1.00
	Daphnia	1.00 ～ 3.00
Oligochaeta	*Nais*	1.00
Insecta	*Chironomus*	15.0 ～ 50.0

determined simply for particular bacteria or other microorganisms to be treated. It must be determined according to adequate evaluation of the chlorine-resistance properties and oxidation resistance properties of the the RO membrane being used, and the fouling of the membrane caused by residual microorganisms.

1) Chlorine-resistance of cellulose acetate membrane

Fouling of the membrane by microorganisms can be prevented by chlorine treatment. However, continuous addition of a larg amount of chlorine, and continuous exposure to high chlorine atmosphere must be avoided. Because the physical strength of the membrane, and the permeation characteristics of the solute[32] will be damaged. Also, in regards to the amount of chlorine added, in practice a definite answer cannot be given. However, there is literature giving data from experiments which have investigated the chlorine resistance of membranes[75](see Table 3.2.2, Table 3.2.3).

Table 3.2.2 Durability of cellulose acetate membrane for chlorination(42-51ppm, 800psi)[32]

Days of exposure	Salt rejection(%)	Membrane Coefficient (10^{-5}g/cm^2·sec·atm)
Intitial control	97.6	1.52
0.17	97.2	1.48
1	97.2	1.60
2	96.7	1.74
5	97.5	1.80
7	97.3	1.93
9	96.3	1.90
12	95.4	2.03
Final control	97.5	1.63

Table 3.2.3 Durabitity of cellulose acetate membrane for chlorination (42–51ppm, 800psi) (10ppm, 800psi)[75]

Days of exposure	Salt rejection(%)	Membrane Coefficient (10^{-5}g/cm^2·sec·atm)
1	97.1	1.11
2	97.2	1.12
3	97.3	1.05
6	97.1	1.24
8	97.0	1.07
10	97.2	1.25
13	96.9	1.20
15	97.0	1.27
Control1	97.1	1.17
Control2	97.8	1.08

A cellulose acetate membrane was saturated with 42–51 ppm of a chlorine atmosphere for 1–12 days, and the measurements of the membrane properties during this period are shown in Table 3.2.2. The properties of the membrane saturated in a 42 – 51 ppm chlorine water solution drops, the desalination rate drops, and conversely the solute and salt membrane permeation volume becomes double that of the control membrane, and clearly the solute elimination rate declines.

For membranes saturated in 10 ppm of chlorine solution, there was not change in the membrane permeation properties for the solute, however the permeation volume of water increased about 10 per cent.

Another report[32] states concerning this kind of chlorine effect, that it was observed that even 2mg/liter of chlorine negatively affected the water permeation properties and solute permeation properties of membranes. On the other hand, there is a report that states that chlorine of 2mg/l shows no particular effect, whereas when this reaches 4mg/l, membrane properties decline. The conclusion is that though there is no clarified limit value, it is estimated if 2–1 mg/l is added for cellulose acetate membranes, there is extremely little negative affect on the membrane characteristics.

2) Chlorine resistance of aromatic polyamide membrane

As the chlorine resistance of aromatic polyamides is extremely low, addition of chlorine must be avoided. If chlorine was added at the previous stage, and residual chlorine exists, it must be supplied to a membrane module after activated charcoal filtration or reducing agent has been added to eliminate the chlorine. Another report by a researcher at a polyamide membrane manufacturer states that though the RO membrane has originally excellent performance, its chlorine resistance is extremely low, and therefore the chlorine needs to be removed with sodium sulfate[69].

3) Dissolved chlorine gas permeation properties of cellulose acetate membrane

In the case of cellulose acetate membrane, it was stated under section 1) that if the chlorine is kept below a stipulated concentration level, the properties of the membrane can be maintained. However the same monogram which reported this[75] further states as follows. When the pH is low for cellulose acetate membranes, the dissolved chlorine adequately passes through the membrane. Sterilization treatment can therefore be accomplished easily from the water production side of the membrane or from the face of the desalination system. Fig.3.2.3 shows how the dissolved chlorine gas permeability of the cellulose acetate membrane changes with the solution pH.

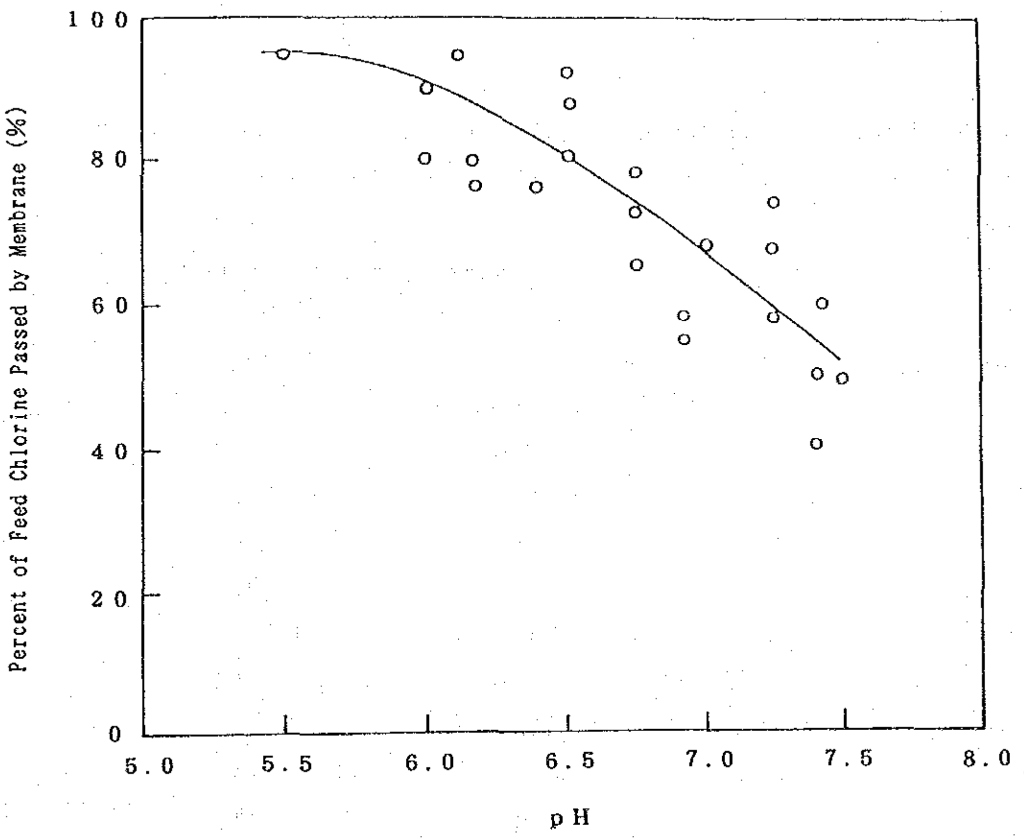


Fig. 3.2.3 Effect of pH on the transport of chlorine through cellulose acetate membrane[75]

3.2.4 Method of Using Chlorine for Seawater Desalination Plants

(1) Effect of Raw Water Components

The fundamental principles of sterilization and algae extermination using chlorine are as discussed above. However, when chlorine is used, on an actual industrial plant scale, the components of the raw water include; water, suspended matter, dissolved salt, general inorganic substance, organic substance, living organisms, germs, microalgae. Furthermore, the components differ according to where the raw water is drawn, the season, temperature, and how long the area has been industrialized. Therefore, not only the aspect of sterilization, but also overall plant facilities, piping system, especially RO membrane fouling, corrosion and deterioration must be taken into consideration. And these need to be looked at as factors causing major problems such as decreased performance, plant stoppages, equipment replacement, increased costs, etc.[34]. Typical seawater composition of Middle East and the Pacific Ocean is shown in Table 3.2.4[35].

Table 3.2.4 Typical seawater composition of Middle East and the Pacific Ocean[35]

Composition	the Gulf	Red Sea	the Pacific
E.C.(μs/cm)	62,700	55,300	45,600
TDS(ppm)	50,600	42,500	34,500
T.H.(mg/l)	8,600	7,280	6,300
pH(–)	8.0	8.0	8.0
Na^+(mg/l)	15,100	12,800	10,560
K^+(mg/l)	568	498	380
Ca^{2+}(mg/l)	520	448	400
Mg^{2+}(mg/l)	1,770	1,500	1,270
B(mg/l)	6.3	5.6	4.4
Cl^-(mg/l)	26,400	22,300	19,000
SO_4^{2+}(mg/l)	3,580	3,030	2,650
Br^-(mg/l)	89	76	65

(2) Example of Industrial Water Production Plant Using Chlorine Components

Japan which is located in the area where the Asian Monsoons occur, has a large rainfall per unit area which is approximately double the world average. In most part of the country, drinking water is taken from rivers, lakes, and wells. However, looking at the rainfall on a per capita basis, this is approximately one sixth the world average. Especially in the Tokyo metropolitan and Kanto area where there is a large population, the rainfall per capita is low at approximately one third of the above, and is smaller in comparison to Saudi Arabia. Therefore water resources cannot be said to be abundant[36](see Fig.3.2.4).

Historically, most of the desalination plants for supplying drinking water used to be constructed on small islands. However with large industrial complexes being constructed in different areas in the past 30 years, desalination of seater/brackish water has also been undertaken on the mainland.

1) The Water Re-Use Promotion Center, Chigasaki Seaside Research Facility

The plant plays a great role in the Japanese development of practical technology for RO seawater desalination. There is a lot of literature in Japan presenting the research data of this plant, and there are also literature from overseas which refer to the plant. Using this plant as the starting point for organizing information, information obtained, limited to issues related to sterilization using chlorine, is given below. Fig. 3.2.5 shows the plant is producing 800m^3 of water per day.

a) When cross-linked polyether RO membrane is used.

<1> Water Intake

Intake pipe extends 200m from plant on land, and 600m out into the ocean, with water being taken a total of 800m away from the plant.

<2> Chlorine Addition

Part of the seawater feed is sucked up into the chlorine generating equipment, and chlorine is produced using an electrolytic process. Seawater with concentration of several ppm's of chlorine in the form of sodium hypochlorite is then supplied back to the water intake, sterilization is accomplished to prevent growth of microorganisms and algae in the piping.

<3> Flocculant

Ferric chloride ($FeCl_3$) is added, and flock is formed.

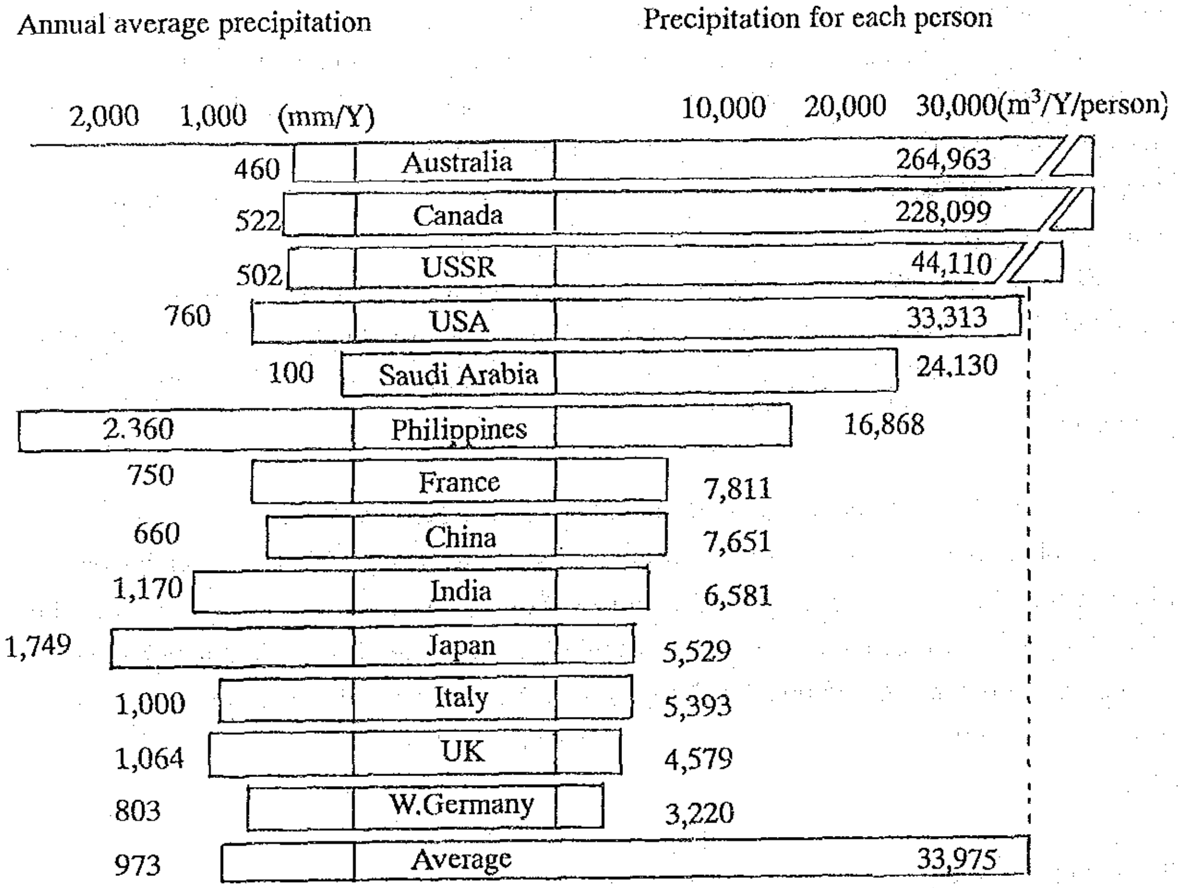


Fig. 3.2.4 World precipitation[36]

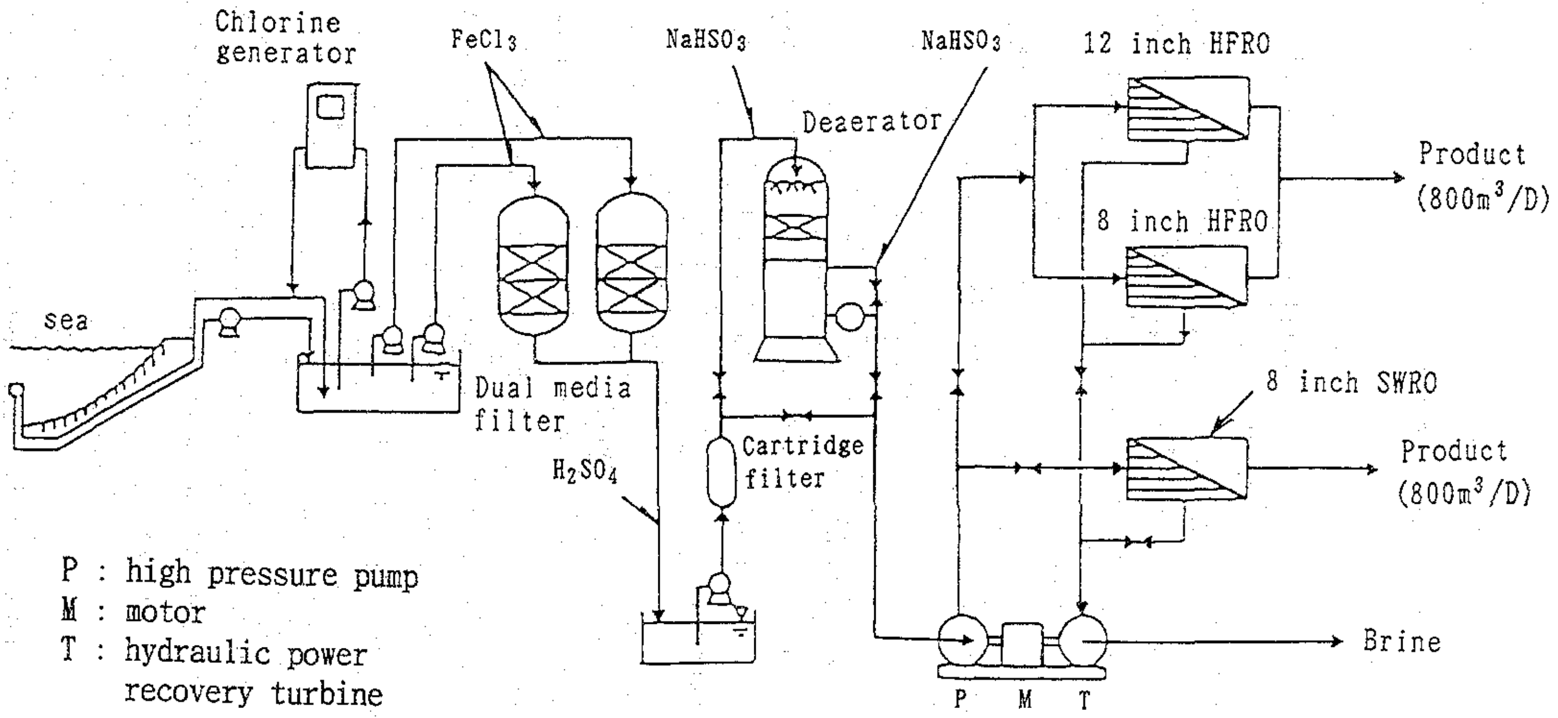


Fig. 3.2.5 Flow diagram of seawater RO desalination plant[37]

<4> Filtration

A multi-layer filter with a series of two towers is used. Sulfuric acid is added to the filtered water, the pH is adjusted, and sent to the deaeration tower.

<5> Sodium bisulfite and deaeration.

After sodium bisulfite is added to remove residual chlorine and dissolved oxygen, deaeration is accomplished immediately at the pressure reduction and deaeration device. This is because the cross-linked polyether RO membrane is oxidized and degraded with residual chlorine of strong oxidizing property and dissolved oxygen. Addition of sodium bisulfite and the deaeration process are imperative as a pretreatment process before raw water is passed through the RO membrane. From the aspect of operating an industrial plant, it is said that the necessity of this treatment is a factor in determining which RO membrane to use[39]. Even in the case when cross-linked aramid RO membrane is used, pretreatment similar to that for the cross-linked polyether RO membrane is imperative for removing residual chlorine[40].

<6> Desalination

The treated seawater whose Fouling Index (FI) is 1.9-3.8, is made to pass through the spiral-wound type membrane module as the seawater which has undergone adequate pretreatment. Next, the seawater is processed through a 10 μm cartridge filter, and is pumped with a high pressure pump to the RO equipment.

<7> Types of Raw Water and Operation Performance

The seawater near the plant and the seawater adjusted to assume the high temperature, highly concentrated seawater found in the Middle East area are used as raw water(see Table 3.2.5). Even with the seawater of the Middle East area standard, it was proved that with single stage RO membrane treatment, the quality of the water product is indicated by a total dissolved solid matter concentration of 210-220 mg/liter[41].

b) When cellulose triacetate is used

In addition to the test of the Toray Industries cross-linked polyether membrane (product name PEC-1000) and the cellulose triacetate hollow fiber membrane (product name HOLLOSEP) made by the Toyobo Co. was tested.

<1> Process Configuration

The process for cross-linked polyether membranes shown in Table 3.2.5 was used. The difference between cross-linked polyether and cellulose triacetate is the treatment of the free chlorine and oxygen residual in the filtrate water which accumulates in the filtrate water tank.

Table 3.2.5 Performance; Operating Results

	Chigasaki seawater	Adjusted seawater for the Middle East
FEED WATER		
pH	6.4–6.9	---
T.D.S.	34000–36000	50000–55000
Temperature	---	30–40°C
PURODUCT		
Quantity	738–813m³/D	738–813m³/D
Recovery	38–42%	21–25%
E.C.	440–460μS/cm	---
T.D.S.	----	210–220mg/l

In the case of cellulose triacetate, by not adding $NaHSO_3$ directly after the filtrate water tank and not reducing the deaeration pressure mid downstream at the deaeration tower, the water is directly pressured into the RO hollow fiber type membrane after passing through the cartridge filter. In particular, the free chlorine is passed through the membrane as is, and after this, the dechlorination agent sodium bisulfite is added into the collected water to remove the chlorine[42].

<2> Membrane Fouling

The cellulose acetate membrane hydrolyzes under acidic or alkali conditions, and in highly reactive substances such as chlorine, deacetylation occurs[43]. However its chlorine resistance is superior to cross-linked aramid membranes, and cross-linked polyether membranes. This was confirmed at the same plant. As long as the chlorine concentration is under a certain level, fouling is minimal[44]. In regards to this value, according to the explanation of the manufacturer of cellulose triacetate, Toyobo Co., it has an adequate chlorine resistance to a chlorine concentration 0.1–0.5 ppm, and will at least clear the required chlorine concentration standard of 0.1 ppm for drinking water. According to another report[45], it is desirable for the upper limit to be under 0.8 ppm.

<3> Microorganisms fouling and decline in performance

Cellulose Triacetate membranes which are resistant to chlorine and oxygen, experience fouling and from microorganisms when the chlorine concentration is under a certain level[46]. By comparison, the cross-linked aramid membrane has a strong resistance feabiofouling.

<4> Chlorine Sterilization

For this reason, chlorine added at the water intake is not removed as free chlorine. It passes through the membrane pores, stunts growth of microorganisms and suppresses formation of slime. Dechlorination is accomplished by adding sodium sulfite in the finally collected water. When operation is shut down, even if flow of water is stopped, by adding free chlorine inside the RO membrane equipment, bacteria will not attach. When the plant is being operated, it is relatively hard for bacteria to become attached[48].

<5> Requires on deaeration for the reduction of O_2, Cl_2, Overall

The cellulose triacetate membrane requires no operations for reduction and deaeration, overall operations are simple. The Toyobo Co. cellulose triacetate membrane was incorporated for the 56,800 tons/day desalination plant in Jeddah, Saudi Arabia[49] for the above reason. For the above reason the Toyobo Co. cellulose tyriacetate membrane was incorporated in the 56,800 tons/day.

c) Other reports concerning fouling resistance of membranes

We have already mentioned that cross-linked polyether membranes, and SU-700 series of cross-linked aramid which may be sterilized with chlorine, this is reported in the catalog by Toray Industries and describes it as dechlorination and deoxidation. However, there is a title "SU-700 series Cross-linked Aramid Composite Membrane which makes possible chlorine sterilization," in the catalog published by Toray Industries,and explains it as, "being able to use chlorine, and hydrogen peroxide disinfectants."

Also, another report[50] points out that the decomposition of cellulose acetate membrane by microorganisms is not caused only by the action of microorganisms: first of all deacetylation occurs as a chemical reaction of residual chlorine, and after this the microorganisms decomposes the cellulose. In this way chlorine sterilization does suppress slime formation, however, the residual chlorine causes the membrane to be decomposed by the action of the microorganisms. Therefore caution is required, and chlorine sterilization limits were pinpointed.

Furthermore, in another report[73], though the aromatic polyamide (made by DuPont) causes Hydrolysis of the amide bond, the usable pH range is broader than cellulose acetate. However, it is recognized that cellulose acetate is superior to aromatic polyamides in dealing with free chlorine, as shown in Table 3.2.6,

Table 3.2.6 Limits of operation of RO membranes[51]

Membranes Material	Cellulose acetate	Polyamide(Du Pont)
pH & temperature	pH 2 to 8 at up to 15°C pH 3 to 6 at up to 30°C	pH 4 to 11 at up to 40°C
Free chlorine	Up to 1 mg.l continuous 50mg/l shock dose	0.1 mg/l pH<8 0.25mg/l pH>8

(2) Hakatajima Seawater Desalination Unit[51]

The flow diagram shown in Fig. 3.2.6 is the sea water desalination facility constructed on Hakatajima Island for the purpose of producing drinking water. Operation started in March 1981.

Water Production: 350 m^3/day

Production Quality (chlorine ions): 90–180 ppm

Concentration of Completely Dissolved Solid Particles: 200 ppm

RO Membrane: Aromatic
Polyamide
Hollow Fiber Type

1) Water Intake

Raw seawater is taken 80m offshore at mid–level.

2) Addition of Chlorine

Sodium hypochlorite is fed from plant to water intake. Through this, activity of bacteria and algae in the seawater is terminated. Concepts are the same as the Chigasaki Seaside Research Facility discussed above.

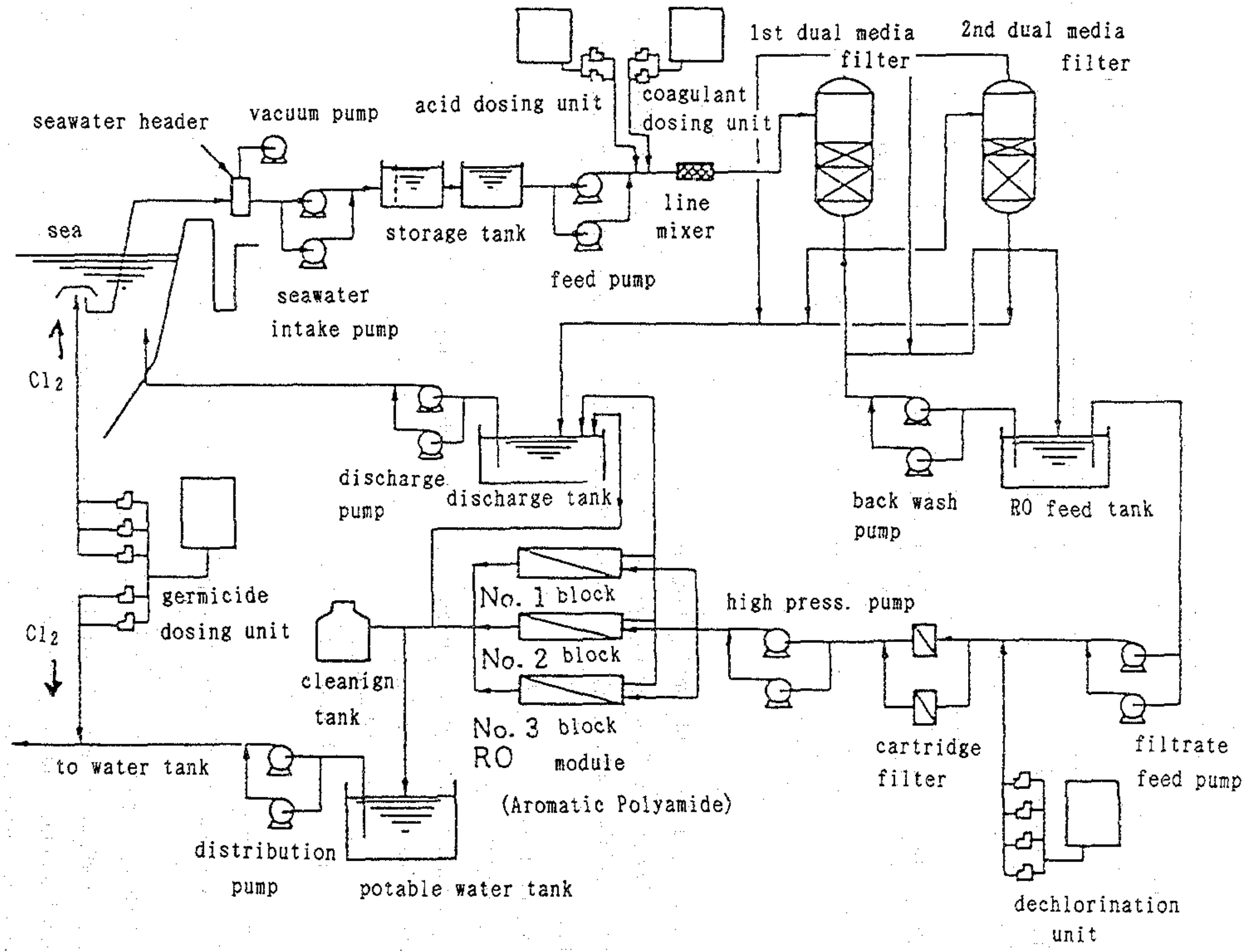


Fig. 3.2.6 Flow diagram of 300m^3/D Hakatajima(Japan), RO desalination plant[51]

3) Pretreatment Equipment

The pretreatment facility is a typical pretreatment facility, composed of sedimentation basin, seawater storage basin, first filter, second filter, RO raw water pool.

4) Raw Seawater Treatment Method

Raw seawater containing chlorine is sent from the seawater storage basin, via the sedimentation basin, by the filter's raw water feeding pump to the filter. However prior to this, sulfuric acid, flocculant, and aluminum sulfate are added to adjust the pH level. The Site Density Index (SDI) of the raw water filtrated by the linear double stage filter is sufficiently below 3. The seawater fed to the RO raw water pool is further pumped through a safety filter and pressured into the RO membrane device. The residual chlorine is retained until immediately prior to the RO membrane device to prevent microorganisms fouling. And then reducing agent, and sodium bisulfite is injected to remove chlorine and thus avoid the fouling of the aromatic polyamide membrane by chlorine.

5) Posttreatment with Chlorine

After collecting the water in the water purification pool, chlorine is further added as a post treatment. Drinking water quality standard value is accordingly secured, and the water is stored.

The above was a description of a typical pretreatment and chlorine treatment example at a plant using the aromatic polyamide hollow fiber membrane module.

(3) Watanaki Village Simple Seawater Desalination Plant[52]

As Watanaki village in Okinawa prefecture is located on a small island, it has no rivers to speak of and the village has relied on the past on rainwater only. In 1987, a 360 million yen simple water supply facility was completed. The facility produces drinking water with the reverse osmosis method using 8 inch spiral-wound type polysulfone composite membranes. It is automatically operated and consists of the latest equipment that supplies an abundance of pure delicious water.

1) Summary of Facility

Table 3.2.7 shows Outline of the Facility

Table 3.2.7 Outline of the facility[52]

Capacity	$240m^3/D$	
Type	RO single stage desalination	
Operating Pressure	$45-55kg/cm^2$	
Recovery	45%	
Membrane	Polysulfone derivative 8-inch spiral-wound type synthetic composite membrane	
Intake	seawater	deep beach well
	brackish	existing well
Intake capacity	seawater: $490m^3/D$, brackish:$90m^3/D$	
Power	105KVA	

2) Quality of Purified Water

Table 3.2.8 Quality of drinking-water[52]

Constituent or Characteristics	Test Results	Japanese Guideline value
Chloride	65.3mg/l	200mg/l
Total dissolved solids	210mg/l	500mg/l
pH	6.9	5.8-8.6
Hardness(as $CaCO_3$)	42.0mg/l	300mg/l
Organic compounds($KMnO_4$)	not detected	10mg/l
Escherichia coli	not detected	not detectable
Odour	nil	not offensive

3) Flow Diagram of Seawater Desalination Plant

Flow diagram of the plant is shown in Fig.3.2.7.

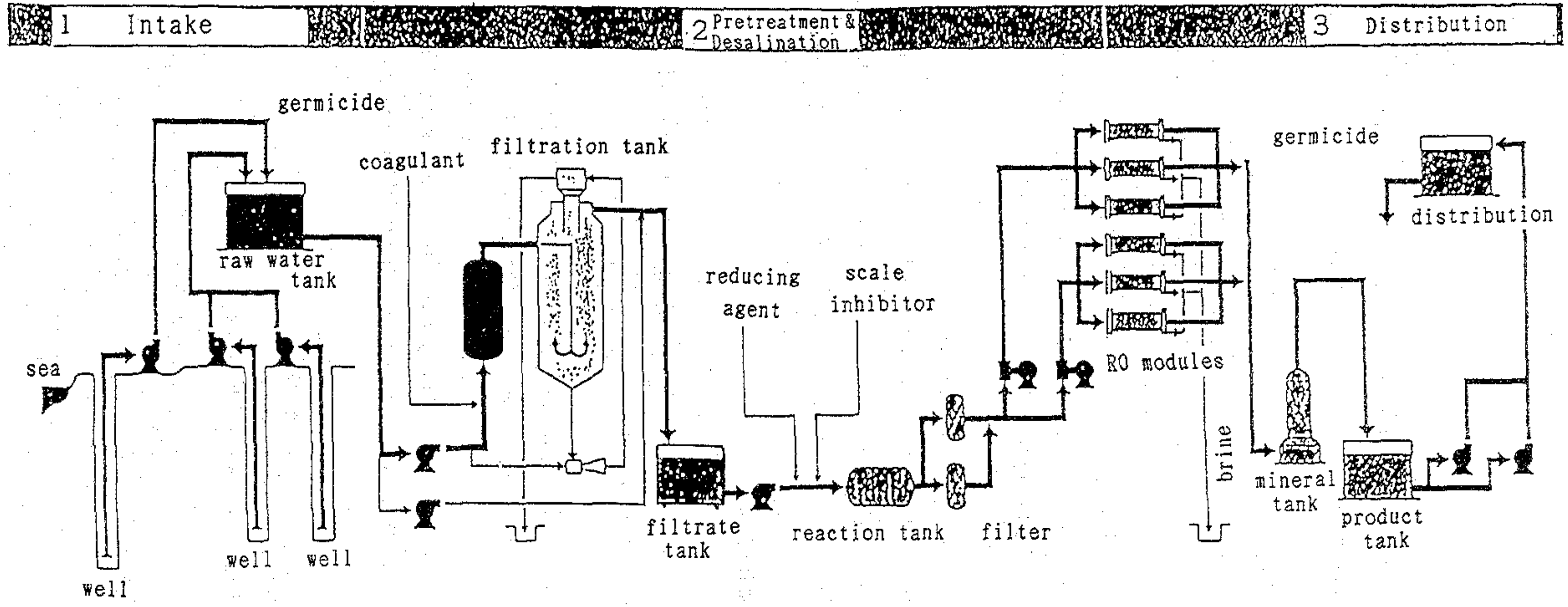


Fig. 3.2.7 Flow diagram of seawater desalination plant[52]

<1> Addition of chlorine

In the seawater storage basin where seawater and brackish water is stored, sodium hypochlorite is added as the disinfectant for sterilization.

<2> Flocculation and filtration.

Before entering the flocculation tank, a flocculant is added in the raw seawater, and after filtration in the sand filtration tank, the water is stored in the filtrate water tank.

<3> Dechlorination and Deoxidation:

Next reducing agent is added, and dechlorination and deoxidation is conducted in the reaction tank. To prevent formation of scale, scale suppressing agent is added at the same time.

<4> Seawater Desalination by RO membrane

The RO membrane is a polysulfone composite membrane for single stage desalination. A feature that can be cited is that free chlorine and dissolved chlorine are removed from the raw seawater prior to entering the RO membrane to prevent membrane fouling.

<5> Chlorine for Posttreatment

The permeate is passed through a mineral tank to add taste. Furthermore, by adding sodium hypochlorite it is disinfected, and thus attaining the water quality level, it is stored in the clean water reservoir as purified water.

(4) Mitsubishi Reverse Osmosis Seawater Desalination Plant Facility[53]

Mitsubishi Heavy Industries is under contract by the Nagasaki Holland Village in Nagasaki Prefecture for construction of a desalination plant to be completed in March 1992. The RO membrane is the cellulose acetate hollow fiber type module. This project is reported as being an interesting example of a plant using sodium hypochlorite as the disinfectant.

a) Specifications

Table 3.2.9 shows the plant specifications

Table 3.2.9 Plant specification[53]

Type	RO single-stage seawater desalination
Product Capacity	1,000m^3/D(500m^3/D* 2 unit
Membrane	Hollow Fiber(CA?)
Product Quality	T.D.S.: 500mg/l Chloride: 200mg/l pH: 5.8-8.6
Completion	March 1992

b) Flow Diagram

Fig. 3.2.8 shows the plant's flow diagram. The RO water production equipment is composed of the water intake pretreatment facilities, RO facilities, and the post treatment facilities.

1) Addition of Chlorine

It is explained that Sodium hypochlorite is added in the pretreatment facilities for sterilization. Whether it is added at the intake port or added in the raw water tank is not clear. However, as problems with algae in the raw water conveying pipes can be anticipated, addition at the intake port is assumed.

2) Flocculation and Filtration

Next the flocculant is added, and after the flock is formed, water is filtered at the multi-stage filter. Next by adding acid, the pH is adjusted, and after passing through the safety filter, it is conveyed to the RO facility.

3) Seawater Desalination by RO Membrane

Furthermore, the filtrate is pressurized by the RO membrane's high pressue pump to approximately 50-70 kg/cm^2, and supplied to the cellulose acetate hollow fiber type RO membrane. Approximately 40 per cent of the supplied water passes through the membrane and becomes desalinated water. The chlorine added at the water intake pretreatment facility is not treated for removing chlorine and sent to the celllulose acetate membrane. The technological concept for this is similar to the method of using cellulose triacetate at Chigasaki Seaside Research Facility, discussed under section 3.2.4(2).

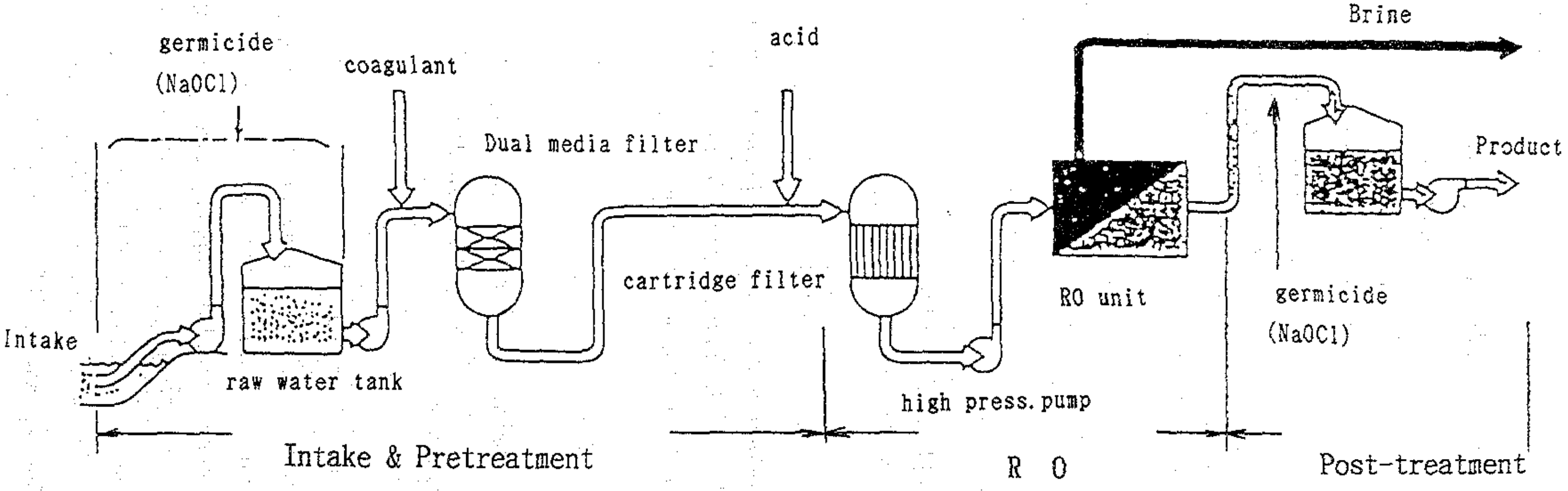


Fig. 3.2.8 Flow diagram of Mitsubishi RO seawater desalination plant[53]

4) Chlorine for Post Treatment

At the post treatment facility, the permeate is added with alkali and disinfectant, to become drinking water. The raw seawater has already undergone sterilization at the intake and pretreatment stages by adding sodium chlorite. As dechlorination has not be done, free chlorine remains in the permeate. It should be noted that disinfectant is further added at the posttreatment facility.

(5) Sumitomo Metal Industries, Kashima Steel Works 13,400m^3/day Desalination Facility[54]

This plant is located in the Kashima industrial zone, and raw water to be treated is taken from the Kashima industry water. This water is Kitaura lake water abundant with cultured organic material, and it easily grows algae and other organisms. As there is influence from seawater, the chlorine ion concentration fluctuates between 150–400 mg/l[55)]. The problem with this plant is fouling caused by the organic matters, microorganisms and algae in the raw water, and therefore the pretreatment has been difficult[56]. Gradually the plant was extended with technical improvements, and currently there are 10 units operating producing 13,400m^3 of water a day.

1) Currently Operating Plant[57]

<1> Facility Specifications:

Information in Table 3.2.10, Table 3.2.11 and Table 3.2.12 is provided with emphasis on especially the sterilization specifications of the plant.

Table 3.2.10 Specification of pretreatmen[57]

Capacity & Quality	18,00m^3/D≤FI 3
Coagulator	600m^3/H*2 unit
Pressurized floating separator	Kurifloator 600m^3/H*2 unit
Filter	Dual media filter 300m^3/H*4 unit
Chlorine dosing unit	NaOCl
Coagulant dosing unit	PAC
Acid dosing unit	H_2SO_4
Scale inhibitor dosing unit	Mizukurihn

Table 3.2.11 Specification of RO desalination plant[57]

Product capacity & Rejection	13,400m^3/D, 93-95%
Recovery	80%-85%
Operating pressure	20-32kgf/cm^2
Membrane module unit	Type: SW Type RO Plant 1000, 1300, 1500m^3/D*10 unit Membrane: Spiral-wound RO membrane F.System(USA) ROGA Type Toray Industries, SC Type

Table 3.2.12 Specification of post-treatment plant[57]

Capacity*Quality	14,000m^3/D*residual chlorine 0.1ppm pH 7.0 ± 0.2
Decarbonator	packed tower type 4000m^3/H*4 unit
Active carbon filter	packed tower type 200m^3/H*4 unit

<2> Flow Diagram

Fig.3.2.9 shows the flow diagram of the plant. This plant is the predesalination device of an exchange resin device for water used in the generator boiler[58] on the premises of the steel works. It was constructed at the end of 1971, has undergone renovations and expansion, and is now the plant as shown in Table 3.2.10-12. To deal with the problem of membrane fouling, it is reported that the spiral-wound type RO membrane was decided upon for use[59].

The 10 membrane elements consisted of the ROGA type made by UOP Company and the SC type made by Toray Industries. Both are the cellulose acetate type. The control of processes are summarized below[60]

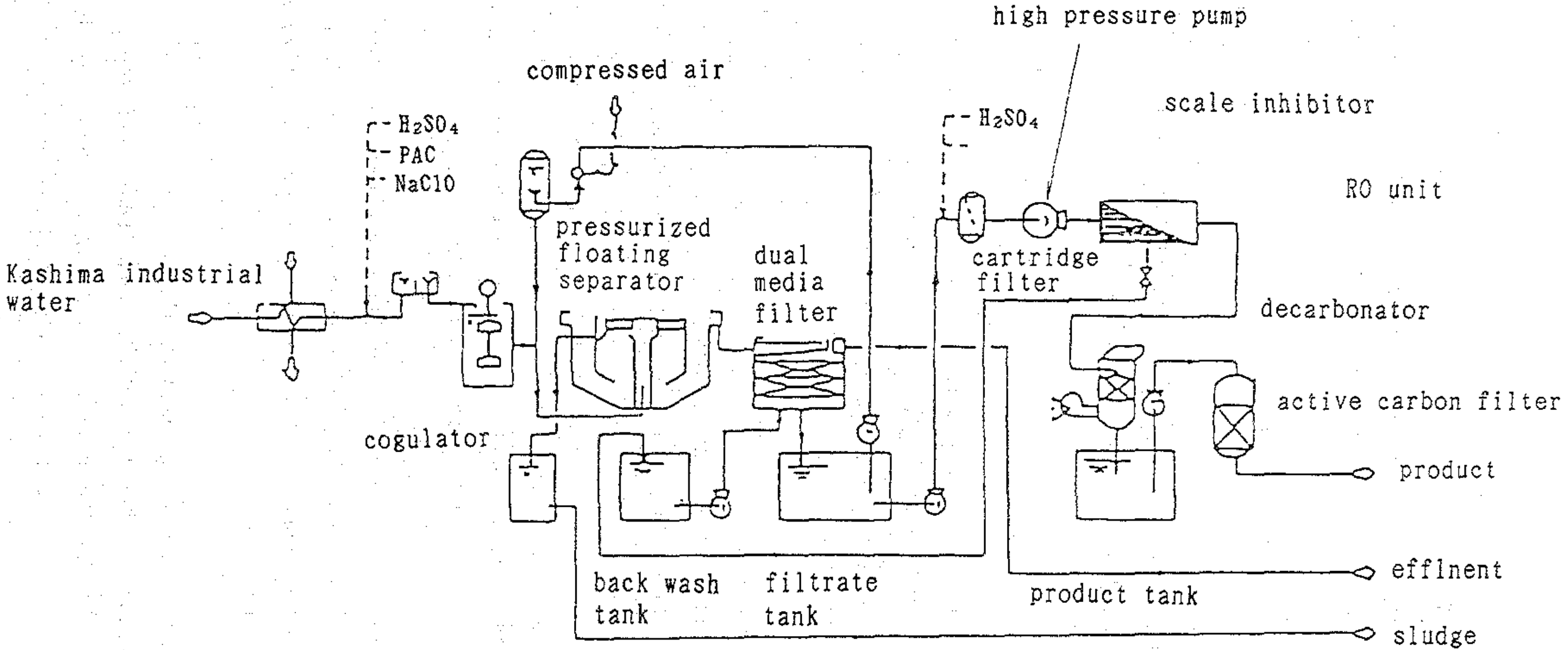


Fig. 3.2.9 Flow diagram of Sumitomo–Kinzoku–Kougyo/Kashima, 13,400m^3/D desalination plant[58]

<3> Pretreatment Chemicals

To accurately accomplish pretreatment filtration of the raw water, it is important to have filtration conditions which are suited for removal of turbid material in the raw water. Chlorine for sterilization, and polyaluminum chloride as the flocculant, and H_2SO_4 as the pH adjustment agent, are, therefore, added.

<4> Flocculation and Filtration

Due to the state of the raw water, it is necessary to reinforce the turbid matter separating performance. Accordingly, a pressurized floating separation tank is incorporated, and a double stage filter is used for filtration.

<5> RO Membrane Facilities

In the case of treating industrial water, the facility is operated with the pressure of 10–30kgf/cm^2. In the case of seawater, with a pressure of 50–60 kgf/cm^2 as the permeation pressure of seawater is high. The water is pressurized into the cellulose acetate membrane without passing through dechlorination.

<6> Sterilization of membrane

As the sterilization treatment during operations, and to prevent the growth of microorganisms inside the membrane module, chlorine sterilization is accomplished. Chlorine is added so that residual chlorine inside the membrane reaches 0.5–1.0 ppm. To prevent growth of microorganisms inside the module when operations are stopped, a disinfectant is added, and it is necessary to occassionally check the concentration[61]. The control technology of chlorine sterilization operations is basically the same as that employed by Water Re–Use Promotion Center at the Chigasaki Seaside Research Facility discussed under section 3.4.2(2).

2) Fouling of the cellulose acetate membrane by chlorine and microorganisms[62].

The same report which introduced this desalination plant, also discusses the chemical and microorganism fouling of membranes and uses the cellulose acetate RO membrane used at the plant as a typical example.

<1> Fouling of cellulose acetate membrane by chlorine:

Generally, addition of sodium hypochlorite as the disinfectant is kept to a minimum level to adjust the residual chlorine concentration below 1mg/l. A report[63] by Oya introduces the decline of the solute separating ratio for sodium chloride aqueous solution.

<2> Biological fouling by organisms:

When the membrane is eaten by microorganisms, the membrane surface becomes damaged, and due to egesta of microorganisms, the membrane is chemically fouled. Electron microscope photographs taken by Motomura and his associates indicate the fouling of cellulose acetate RO membranes[64]. On the fouled membrane surface, a 1–10 μm sized microorganism colony was observed, and $2 \times 10^6/\text{cm}^2$ of microorganisms were counted. The membrane is clearly eaten up. The desalination ratio of the membrane dropped by 25 per cent, and the permeate flux increased to $2.5\text{m}^3/\text{m}^2$day. The performance pattern with pin holes was thus indicated.

To prevent this kind of biological fouling, there is a need for adequate sterilization in the pretreatment system. Originally, the main objective of the RO system is to separate the solute, and not to separate the turbid material. Therefore, it is reported that there is a need to remove as much of the turbid material as possible during pretreatment. The report emphasizes in Table 3.2.13 that to secure a stable operating performance, the most important item is the operation control of pretreatment equipment[65]

Table 3.2.13 Decrement of membrane rejection by chlorination[65]

Object membrane of investigation	Cellulose Acetate	
Object of aqueous solution	NaCl 35,000mg/l	
Operating conditions	Pressure 60 atm Temperature 25°C	
Cl_2 concentration on membrance	1mg/l	4mg/l
Initial rejection	94.1%	94.1%
Rejection after 30,000 hrs operation	94.0%	90.1%

3) Progress in Technological Study of Plant

In regards to the operation of the plant, prior to the above report discussed, a very interesting report, by a technician at the plant, concerning the progress of technological study, was made in 1980[66]

<1> Summary of Process Configuration:

At the beginning, as shown by Fig.3.2.10, after the filtrate water tank, the water passed through the activated charcoal tower (indicated by the dotted line, arrow A). After the residual chlorine has been sufficiently removed in the tower, the water passed through a media filter, a heat exchanger, and was pressurized into the RO aromatic polyamide membrane (DuPont B-9, permeator), and block to obtain dechlorinated water. As it is an aromatic polyamide membrane it was presumed that there was no membrane fouling from bacterial growth. The residual of the chlorine used as the disinfectant at pretreatment was, therfore, completely removed in the activated charcoal tower. This treatment was originally considered (May 1975) suitable for preventing fouling by free chlorine. However, 6 months after start of operations, a large amount of bacterial slime was found in the piping system after the activated charcoal tower. Due to this, the RO membrane became blocked and resulted in decreased performance.

<2> Summary of Renovations

After various studies, the following conclusions were reached.

As a measure for suppressing growth of bacteria slime disinfected with chlorine passing the raw water through the activated charcoal tower was stopped. Instead, chlorine reducing agent (sodium bisulfite) was added immediately prior to the RO block pressure pump, and the line for the residual free chlorine was lengthened as much as possible. However, as the final measure, a disinfectant was selected and was added at the C point in Fig. 3.2.10 (which does not alter the quality of the aromatic polyamide membrane). The operating results of the plant are shown in Fig. 3.2.11.

<3> Current Operating Conditions of the Plant

Despite the above technical studies, the membranes currently used in the RO desalination equipment are not aromatic polyamide but are cellulose acetate as indicated in Table 3.2.11 above and as shown in Fig. 3.2.9, the disinfectant used is chlorine.

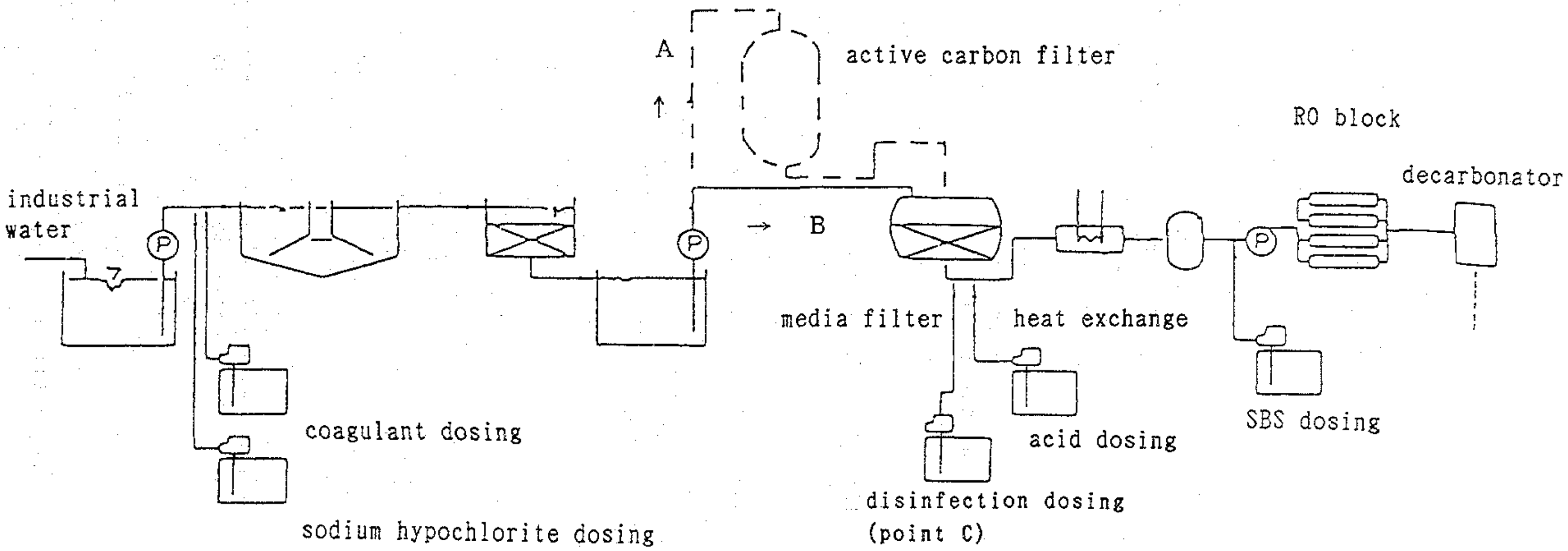


Fig. 3.2.10 Flow diagram of pretreatment section[66]

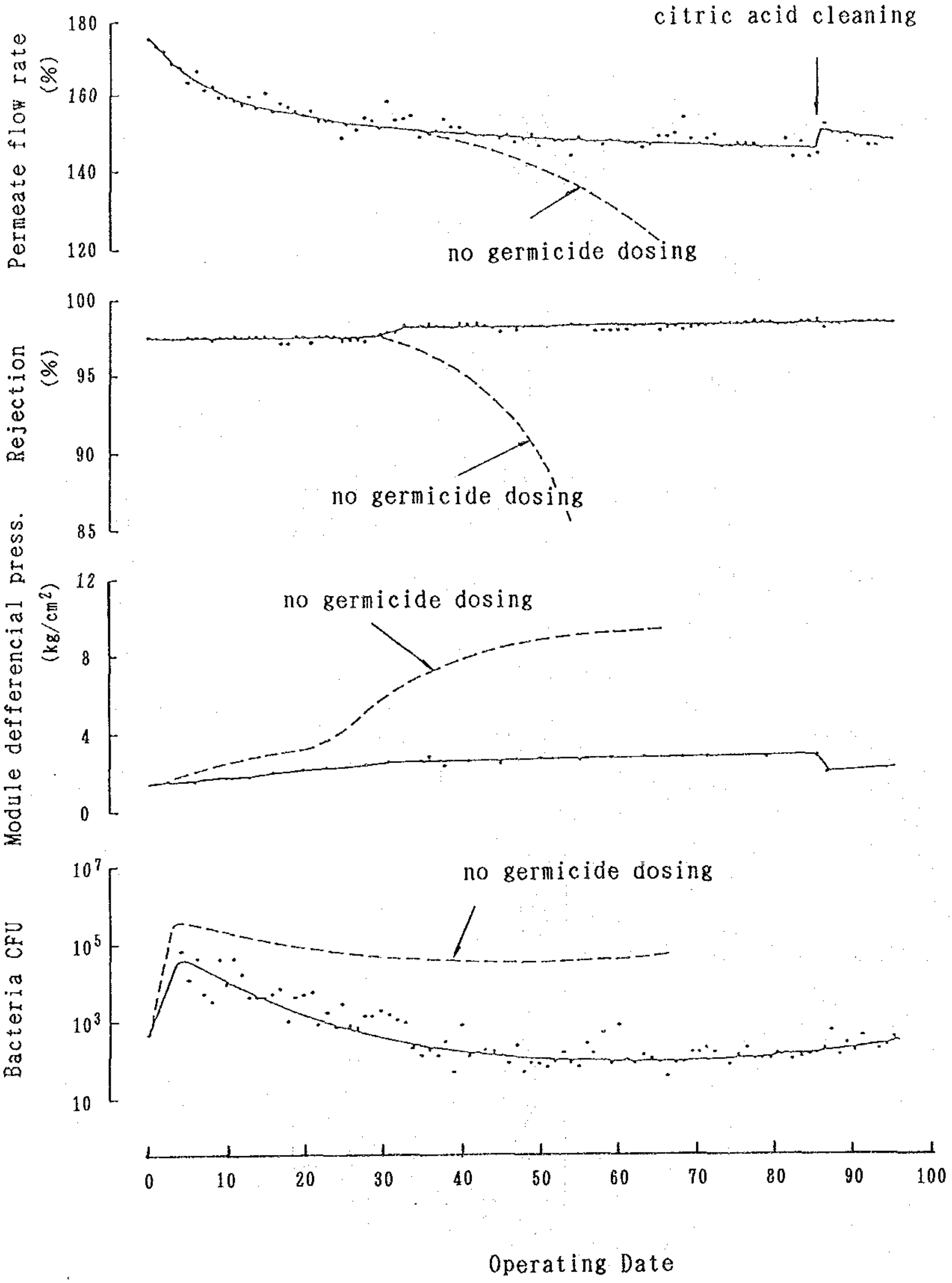


Fig. 3.2.11 Plant performance[66]

3.1.5 Chlorination Facilities

(1) Overview

The chlorinator (chlorine sterilizer) is already on the market on a commercial basis. The equipment is safe and reliable, and there are no problems with it[66]. However, as the chlorine is a toxic gas with a stimulating odor, the utmost care is required.

Especially when the amount of raw water to be treated is large (for example for city water supply), several tons of chlorine is added every day for the chlorine concentration of several ppms. For this kind of large scale chlorination facility, not only a large number of chlorinators need to be installed, but there is a need for a series of equipment such as tank lorries for transporting the liquid chlorine, receiving facilities, liquid chlorine storage tanks, evaporators, and gas chamber. Also, a pneumatic compressor for conveying pressurized chlorine, and neutralizing equipment for the facility's shutting down and in emergencies. Electrical instrumentation equipment for plant operations accounts for 30 per cent of total equipment costs.

(2) Chlorinator and Its Operation

<1> Types

Chlorinators can be broadly divided into the dry type and the wet type. The dry type directly adds chlorine gas through a diffuser or a chlorinator. With the wet type, chlorine is dissolved in a small amount of water to create concentrated chlorine water, and this is added into the water to be treated.

Fig. 3.2.12 and 3.2.13 show the dry type and the wet type respectively.

<2> Precautions for Use

The report summarizes the operating procedures for the chlorinator.

Backup unit:

As the chlorinator easily malfunctions, a backup unit should be installed. Both units should be constantly maintained and chlorine addition should under no circumstances be stopped. In this way, the standard level of chlorine concentration of the water supplied can be continuously assured.

Control of addition volume:

Stability of addition volume near the maximum addition capacity of the unit is unstable, and malfunctions occur easily. On the contrary, if the added volume is small compared to the

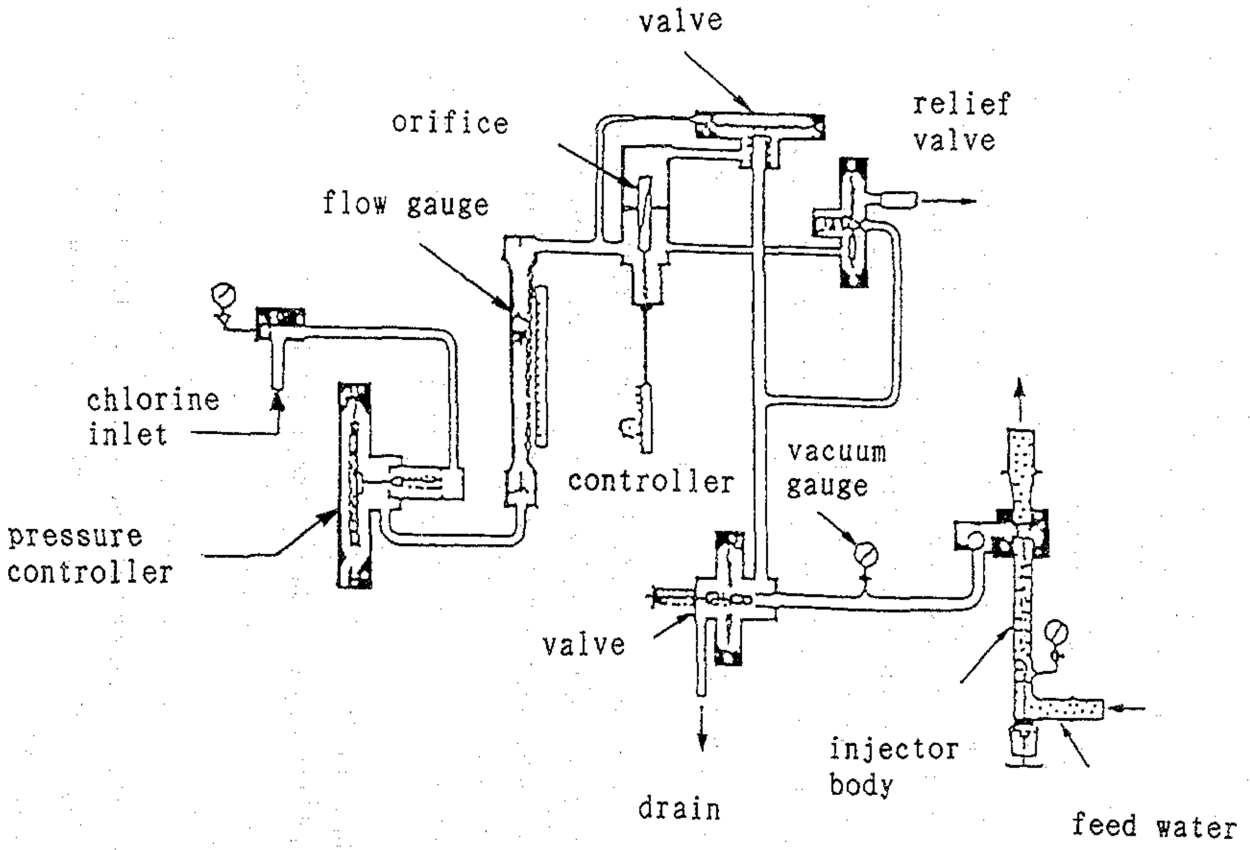


Fig. 3.2.12 Chlorine dosing device Wet A-721[67]

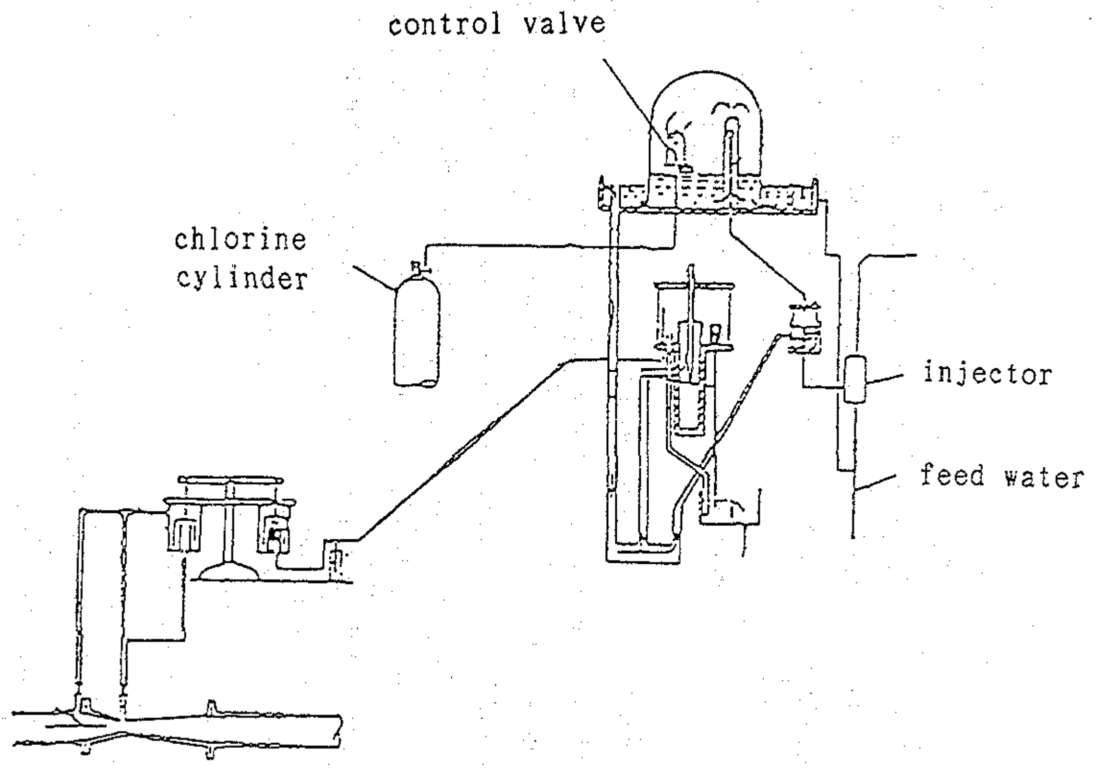


Fig. 3.2.13 Chlorine dosing device[67]

addition volume, as the measuring sacle is narrow, adjustments are not very accurate. The report states that addition should be continually maintained in the 60 to 80 per cent range of the addition capacity.

Temperature control of the chlorinator chamber:

That vapor-liquid equilibrium state of chlorine will be changed according to teh pressure and the temperature can be understood by the physical and chemical properties of chlorine as discussed under sedtion 3.2.(1). To stablized the evaporation, the temperature of the chamber where the chlorinator is located needs to be maintained between 15–20°C even in cold seasons. When the chlorine is evaporated by direct heating from an electrical heater, after the chlorine gas has been put into the chlorinator, it is rapidly cooled, and reliquified. As this can block the machinery, direct heating should be avoided, and indirect heating should be used.

<3> Estimated number of required chlorinator

Naturally the scale of the chlorination facilities depends on the amount of raw water to be treated(T/D). Furthermore, it is necessary to consider that on some days the amount of microorganisms, algae and bacteria to be sterilized will increase above average values[68] depending on change in season and the temporary increase of raw water in the season of storm and flooding. The imperativeness of the backup unit has already been mentioned under section 3.2.5.2. Some specifications of the water re-use plant needed for the estimation of the required number of chlorinators are mentioned below.

1> Specifications:

Production volume: 30,000T/D
Recovery rate: 30%
Treated raw water volume: 100,000 T/D
Chlorine concentration: 5mg/l[9]

2> Calculation of Required Number of Units:

Supposing that the chlorinators directly extract chlorine from a bomb(50kg chlorine/bomb) as shown in Fig.3.2.12 and 3.2.13.

Required chlorine volume:100,000 T/D x 5 mg/l = 500 kg/D
Required units = 500 kg/D divided by 50 kg/set = 10 sets/D

There is information that the maximum number of germs and algae over the year(number in 1ml), is several times the average number. Furthermore, considering that the backup unit must be definitely maintained, it can be estimated that the required number of units will greatly exceed the above values. It will be necessary to rationalized the process. Furthermore, it is said that compared with the process of sodium hypochlorite and other chlorine source, the advantage of rationalized chlorinator process is to quickly respond to the large fluctuation of loads such as bacteria.

<4> Chlorine gas addition equipment from liquid chlorine

Currently, sewage works consume a large amount of liquified chlorine gas and have a similar process to that of seawater desalination plants.

To avoid making mistakes in the outline of the research, manufacturers of chlorine related equipment were visited, and technical surveys were made. Here the details of information obtained concerning reception of liquid chlorine, storage, evaporation, additionm, and removal equipment will be given.

Surveyed manufacturer name, location
Company name: Nitto Machinery Co. Ltd.
Head office: 1-8-9 Yaesu, Chuo Ku, Tokyo
Preconditions of facilities design:
Raw water treatment volume: 100,000 T/D
Water product volume: 30,000 T/D
Chlorine concentration: 5mg/l
Annual average chlorine consumption per day: 0.5 T/D
Maximum chlorine consumption per day: 2.5 T/D
Flow diagram of chlorinator for 10,000 T/D

Fig. 3.2.14 gives the flow diagram for facilities suitable for the annual average consumption of 0.5T/D chlorine, and maximum 2.5T/D. The specifications presented by the authors belong to the class which is one rank over the smallest scale in the main flow and diagram series.

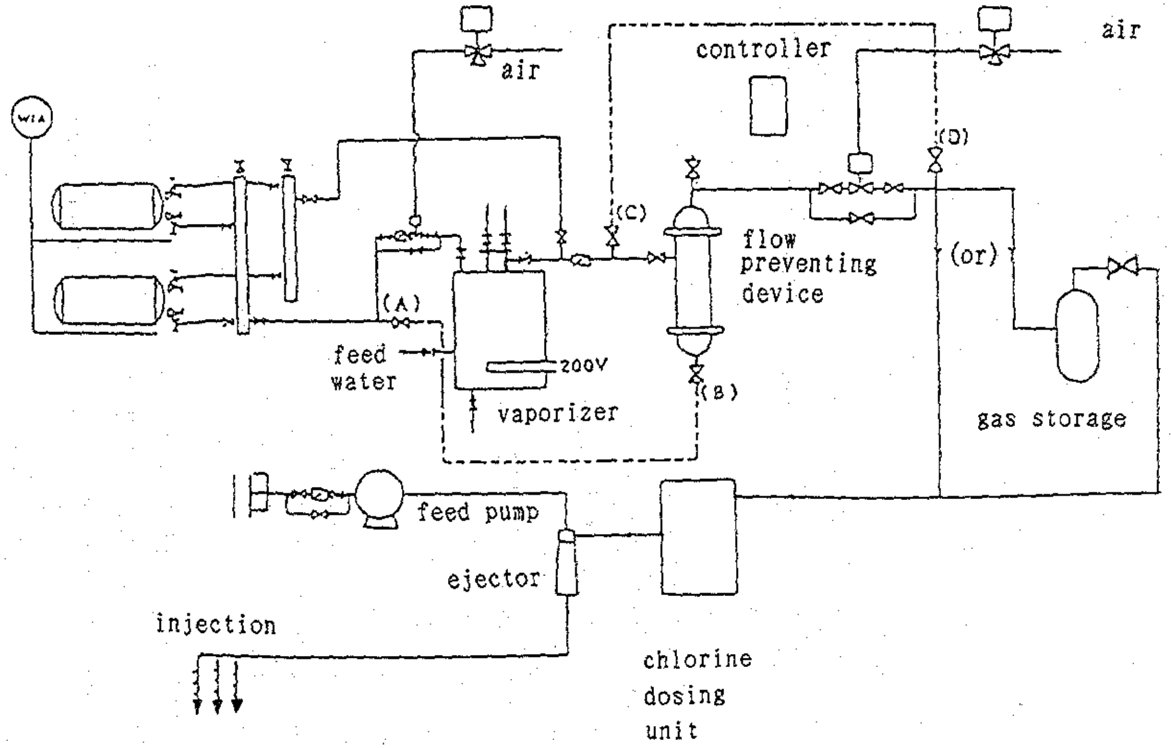


Fig. 3.2.14 Flow diagram of chlorine gas addition equipment[68]

The major equipment are 1 ton bomb containers, bomb gauges, evaporator, inflow stopper, air chamber, chlorinator, ejector, ejector water supply pump. Furthermore, it is composed of piping and gauges. In addition, the chlorine removal equipment, neutralizing equipment, and chlorine gas leakage detector are included.

After two 1-ton bombs are fixed to the gauge respectively, volume is measured. The liquid chlorine is pressured by 4-5kg/cm^2 G of chlorine gas pressure in the bomb and enters the evaporator shown in Fig.3.2.15. The evaporator is a winding pipe type heater tank(or bayone type), and the hot water tank is filled with water heated to under 40°C. The evaporated chlorine enters the inflow stopper. Fig.3.2.16 shows the liquid chlorine stopper, and the liquid chlorine flowing out of the evaporator is caught to prevent inflow into the chlorinator. If liquid chlorine is mixed with gaseous chlorine, the chlorine concentration of the raw water will fluctuate greatly, and therefore contact must be avlided.

The chlorine gas either enters the chlorinator directly, or when chlorine consumption is large, it passes through the gas chamber (approx. 1m^3) to enter the chlorinator. The chlorine gas which exits the chlorinator (mentioned previously that liquid must not be contained), is mixed at the ejector with the pressurized water coming from the water supply pump, to become chlorine aqueous solution.

Fig. 3.2.17 shows the chlorine removal equipment. At the plant's regular repairs, the equipment removes and neutralizes the residual gas inside the system equipment. Moreover, it is essential for preventing occurrence of chlorine leakage accidents. The chlorine gas applied by the blower, comes up from the bottom part of the tower, and is neutralized by caustic soda showered from the top. A chlorine gas leakage detector is also installed.

<1> Number of units of equipment

Bomb gauges: 1 gauge for two bombs and a back up to handle maximum chlorine comsumption.
Bombs: Four 1-ton bombs are continually installed.
Evaporator: 2 sets, of which 1 is a backup
Inflow Stopper: 2 sets, of which 1 is a backup
Air Chamber: 1 set, of which 1 is a backup
Chlorinator and ejector: 2 sets, of which 1 is a backup.

In normal water purifying plants, for process control, chlorine is added as pretreatment and midtreatment. Furthermore, at the post treatment, the residual chlo

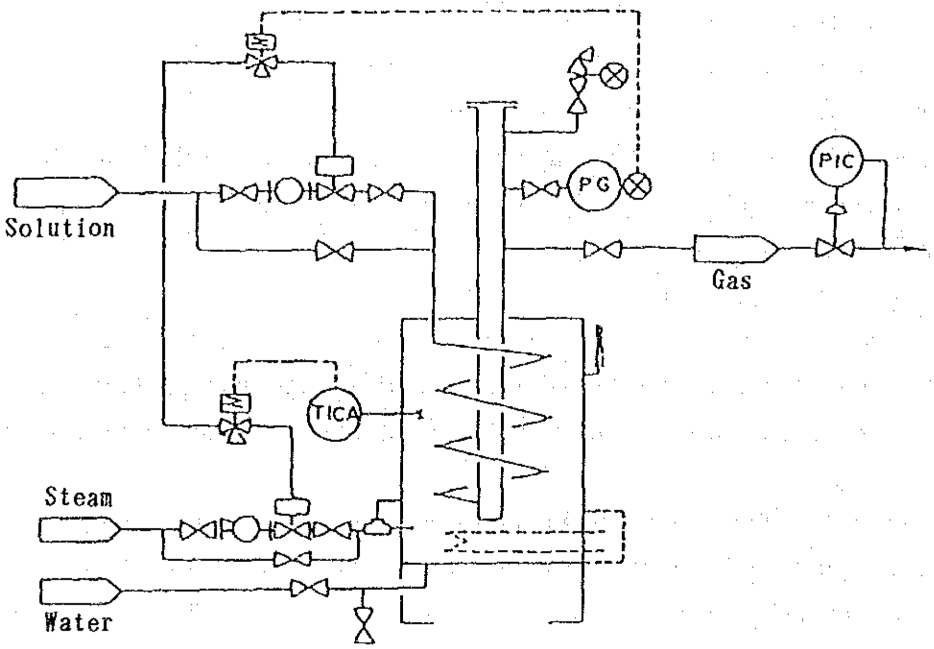


Fig. 3.2.15 Flow diagram of vaporizer[68]

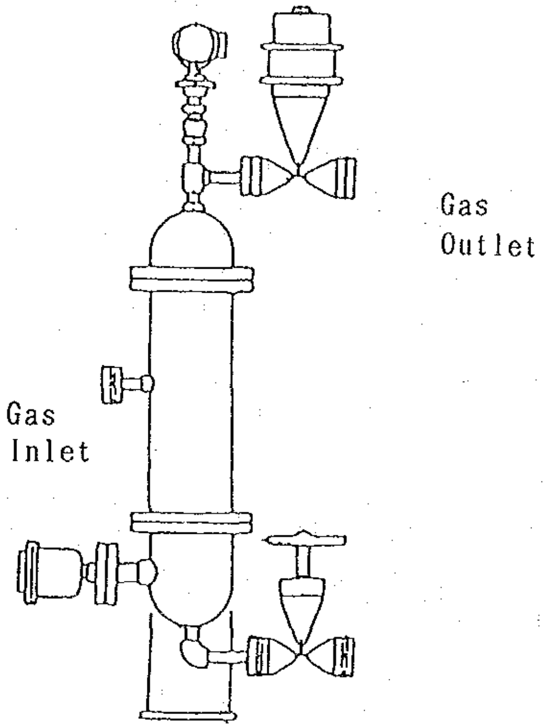


Fig. 3.2.16 Liquid chlorine stopper[68]

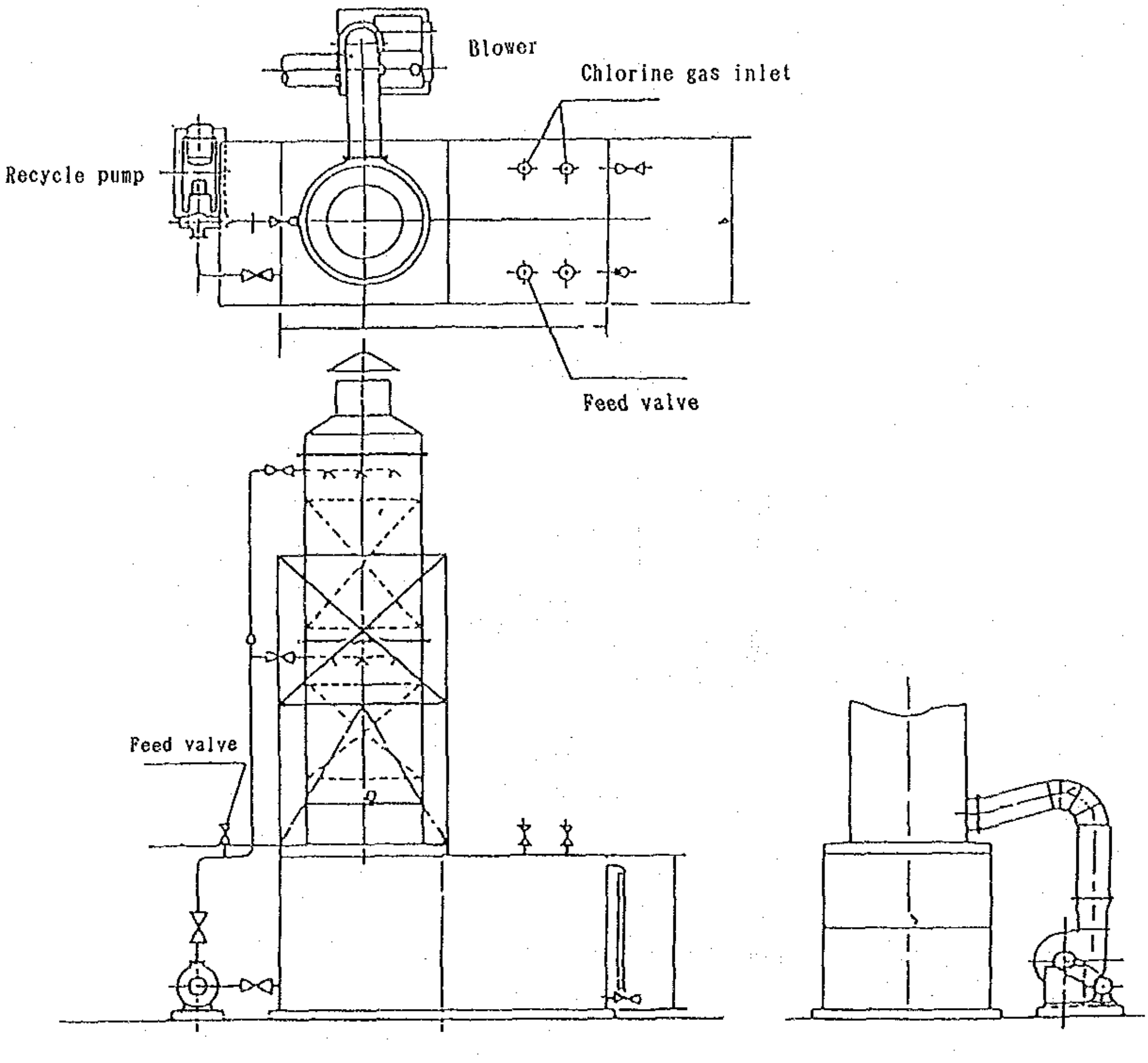


Fig. 3.2.17 Dechlorinator[68]

rine of the clean water is adjusted to 1 ppm. However, with the current technical survey, as the required specifications have not been fixed, only the pretreatment chlorinators are being mentioned.

Chlorine removal equipment: 1 set.
Depending on the length of processing capacity will change.

Piping: 2 system piping, with equipment being able to be interchangeably used.
Piping works will be limited to inside a building.

Instrumentation: 2 system instrumentation.
However, 1 system for 1 set of 1 set of equipment. Air for instrumentation is supplied, and includes chlorine gas leakage detector.

Materials: Chlorine contact part up to the chlorinator must be in accordance with STPG (JIS rating). The zero point of moisture when pressurized air is used, must be under −45°C.

Accommodation of Load Fluctuations: Must sufficiently satisfy required specifications.

Air Conditioning: In Saudi Arabia, as outdoors is hot, bombs brought in will be hot. The equilibrium vapor pressure inside the bomb will, therefore, increase. So there is a need for air conditioning in the reception and equipment rooms.

<2> Required Building Space

Reception facility room: 2 sets of gauges for 2 bombs. 10 storage bombs, 2 empty bombs.

Main Equipment Room: Main equipment, ejectors, chlorine removal equipment, piping, instrumentation are placed in this room.

The required building space is as shown in Fig. 3.2.18.

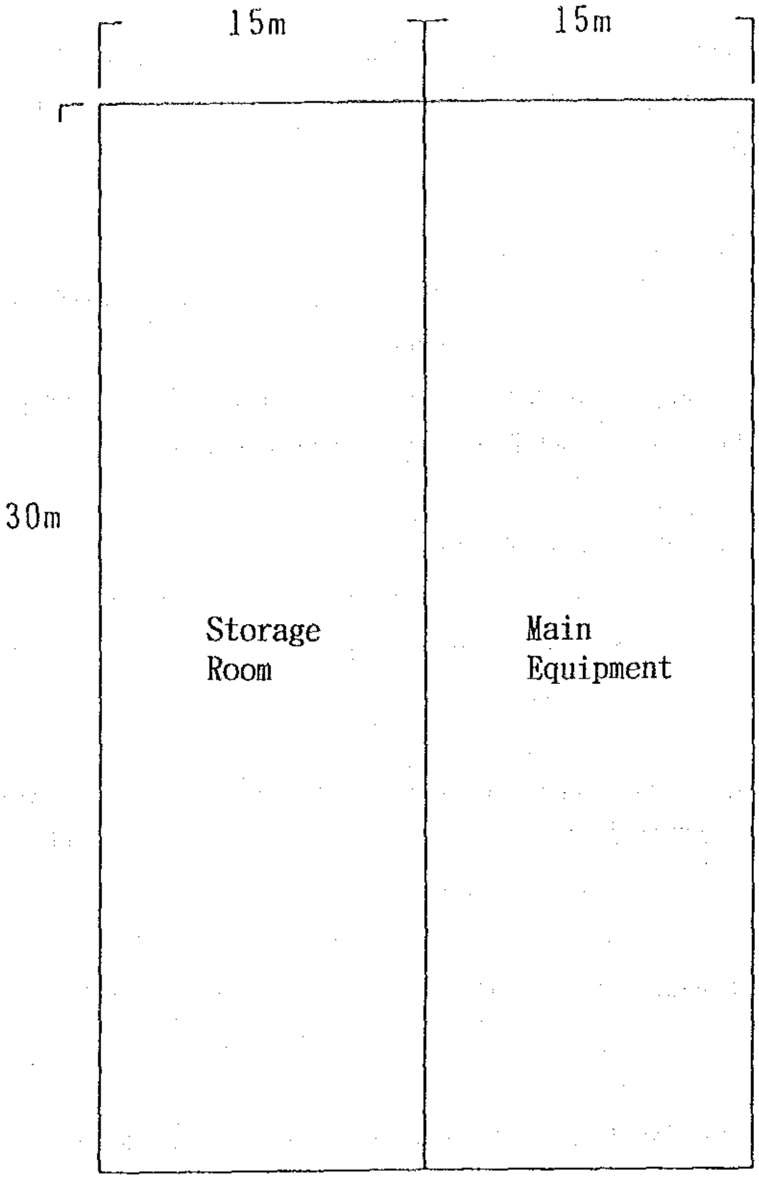


Fig. 3.2.18 Required space of building[68]

Floor space: 900m^2

Building standard: Reinforced concrete structure, which will permit air-conditioning system, and room temperature must be maintained at 22-23°C. The costs of air-conditioning system were not considered as a part of this survey.

Bomb Storage, Warehouse: Required separately.

<3> Construction Costs of Chlorination Facility:

Equipment costs: 100 million yen. Description is as under item a).

Piping costs: 20 million yen. Same as above.

Miscellaneous: 10 million yen. Packaging, inspection, shipping, and insurance costs.

Construction personnel costs: 10 million yen

Description: 60 persons/day (for the case of construction in Saudi)

Direct personnel costs 6 million yen

Travel and lodging costs 4 million yen

Total 10 million yen

Total: 140 million yen

Building construction costs: 135 million yen

Floor space: 900m^2 x unit price 150,000 yen/m^2

Reinforced concrete structure, and the unit cost indicated is that of Japan. The costs of air-conditioning system are not included.

<4> Required Operating Staff

Central Control Staff:

1 person/shift, 3 persons/day : Must be skilled and have knowledge concerning chlorine.

On site workers:

2 persons/shift, 6 persons/day: Plant is automated. However, to prevent accidents from occurring when bombs are replaced, staff will be placed from the safety aspect. Depending on the level of the worker's skill, the number of workers can be reduced.

Total: 9 persons/day

<5> Utility: Small enough to be ignored. Costs related to air-conditioning system are separate.

<6> Life: Same as general chemical industries, 7 years.

3.2.6 Cost of Chlorine Sterilization

(1) Cost of Materials

<1> Preconditions

5 ppm concentration of chlorine added into 100,000 T/D of raw water in the pretreatment process, and dechlorinization and use of dechlorization agent shall not be used. Also other chemicals are not included in the calculated costs for chlorine sterilization. Water product shall be 30,000 T/D.

<2> Cost Calculation

Chlorine consumption per day: 0.5 T/D (100,000T/D x 5ppm)
Unit cost per ton of chlorine: 60,000 yen/T (unit cost when 5 – 6T/D of Kashima
complex product is purchased).
Chlorine cost per day: 30,000 yen/D (0.5T x 60,000 yen/T)
Chlorine cost per ton of water product: 1 yen/T (30,000 yen/D/30,000T/D

(2) Labor Costs

<1> Preconditions

9 staff persons required to operate. As this number of personnel includes extra personnel in consideration of safety aspects, backup staff will not be placed. Labor costs are 4 million yen per year per person.

<2> Calculation of costs

Annual labor costs: 36 million yen/year; Staff 9 / day X 4 million yen/man year
Labor costs per day: 98,700 yen/day
Labor costs per ton of water product: 3.3 yen/T

(3) Facilities Costs and Depreciation Costs

As equipment costs, 140 million yen is appropriated as the cost of constructing the chlorination facility and 135 million yen is appropriated as the building cost. The life of of the former is 7 years for straight line depreciation, same as general chemical industries, and the latter is 35 years for straight line depreciation.

(4) Utility Costs

Small enough to be ignored. The operating costs of the airconditioning system are separate.

(5) Calculation of Sterilization Costs

<1> Annual depreciation: 23.9 million yen/year

Description:

Equipment depreciation 140 million yen/7 years
Building depreciation 135 million yen/35 years
Total depreciation costs 23.9 million yen/year

<2> Depreciation Costs per Day: 65,400 yen/day, per 30,000 T/D water product
<3> Depreciation costs per 1 ton of water product: 2.2 yen/T
<4> Utility costs: 0; However, the operation costs of the air–conditioning system are separated.
<5> Sterilization costs per 30,000 T/D

Material costs:	30,000 yen
Labor costs:	98,700 yen
Depreciation costs:	65,400 yen
Utility costs:	0 yen
Total:	194,100 yen

<6> Sterilization Costs per 1 ton water product(yen/T)

Material costs:	1.0 yen
Labor costs:	3.3 yen
Depreciation costs:	2.2 yen
Utility costs:	0 yen
Total:	6.5 yen

3.3. Disinfection by Ultraviolet Irradiation

3.3.1 Outline

Disinfection by ultraviolet irradiation has been widely used recently[76], in addition to the conventional methods to kill bacteria and algae such as heat treatment, chemical treatment, coagulation or filteration. Following the invention of the fluorescent lamp, technologies to apply ultraviolet light to disinfection treatment were studied in the 60's and 70's. As the lamp technology for the source of ultraviolet light has been established, the applicable areas of ultraviolet light have been widely expanded[77].

(1) The features of the ultraviolet light disinfection[78]

The following features can be listed in comparison with other ways of disinfection.

a. Safer and relatively easier to operate
b. Not selective to the types of bacteria and works on almost all kinds of bacterial
c. Does not produce harmful materials or cause secondary water contamination as the chemical treatment, because it disinfects bacteria without destroying biomass.
d. The required time is very short, within a minute. A large space for the system is not necessary.

These are the advantages of the ultraviolet light disinfection.

(2) Applied areas of ultraviolet light disinfection technology

With the advantages mentioned above, this type of treatment is considered to be an important process especially in the production of ultra pure water, requiring the highest level of purity. The highly clean systems are needed in the fields of most advanced technologies, including the electronics industry, the biotechnology, and medical engineering. The ultraviolet light disinfection treatment will be used more and more as an efficient way of disinfection to meet strict requirements[79]. For the water treatment in other fields such as the food industry, the fishery and fish-raising, the disinfection of cooling water, and the disinfection of water supply and sewage water, this is also an indispensable technology. It is reported that this treatment appeals as an excellent way of disinfection for the next generation[80].

(3) The possibility to apply the ultraviolet light disinfection to the seawater desalination with the reverse osmosis membrane.

The ultraviolet light disinfection has profound potentiality as mentione above. According to our technical research into Japan Photo Science Corporation which has good results in commercializing the ultraviolet light disinfection technology[81], however, not many domestic or foreign desalination plants use ultraviolet light for disinfection on a large scale as yet. They request that an appropriate disinfection technology utilizing ultraviolet light be developed that can be applied to desalination with RO membranes and they made a few suggestions[82]. In another report[83], the limitation of chlorination is described, pointing out that chlorination restrains the formation of slime but residual chlorine assists bacteria to decompose the cellulose acetate membrane. It is suggested that ultraviolet irradiation is one of the effective ways to disinfect in such a case.

The document of Japanese published patent Sho-56-129009[84] emphasizes the advantages of using the ultraviolet light for disinfection. In this case, raw water is introduced into the RO membrane without chlorination and a part of the permeate is used repeatedly to clean the membrane, and the other part of the permeate is treated by the ultraviolet light irradiation.

The use of ultraviolet rays is a technology of interest as a means of disinfection in RO membrane desalination.

(4) Disadvantages of ultraviolet light disinfection[85]

Ultraviolet light disinfection has excellent features as mentioned above, while it also has the following disadvantages in practical application.

a. The permeability of ultraviolet light varies with the water quality. The disinfection effect may be reduced depending on the water to be treated.
b. The suspended particles screen the ultraviolet light, so the ultraviolet light effect to microorganisms is reduced.
c. The UV illumination intensity of the low pressure mercury lamp used as the source of ultraviolet light depends on the ambient temperature.
d. Duration of this disinfection effect is shorter than that in the chemical treatment. Some measures to counteract these disadvantages should be taken for the ultraviolet disinfection system to be practical.

3.3.2 Principles of Ultraviolet Light Disinfection

(1) The Mechanism of the Ultraviolet Light Disinfection

The mechanism of disinfection or mutation by ultraviolet light is complicated and is being clarified by photobiology. The reports[86] referring to these principles, however, seem to have the same viewpoint.

<1> Presence of UV-C

It has been known for some time, that a sunbeam has a disinfection effect. This is the action by the ultraviolet ray of the sunbeam. Ultraviolet ray is the general term for the electro-magnetic waves of the wavelength range of about 100 – 400 nm between X ray and the visible rays of all radiant energy as shown in Fig. 3.3.1. The physical and chemical quality as well as the effect on organisms vary greatly with the wavelength.

The ultraviolet rays in the intense light of the sun are photobiologically divided into the following groups by the effect.

Table 3.3.1 shows disinfection light UV–C exists in the sunbeam.

<2> The mechanism of disinfection

a. The bactericidal effect of the 260nm wavelength ultraviolet ray

Fig. 3.3.2 shows the relative sterilization effect of each of the three wavelength groups. The energy of sterilization action is highest at the 260 nm wavelength.

The main reason why the ultraviolet ray causes disorder for bacteria is that the absorption of deoxyribonucleic acid (DNA) is the highest at the range of ultraviolet wavelength[90]. DNA is the most important component of an organism and is necessary to the life support and the duplication of the organism.

Fig.3.3.3[91] shows the absorption of ultraviolet light for DNA and protein. Fig.3.3.3 also shows that the absorption of ultraviolet for DNA is utmost at around 260 nm wavelength. This coincides with the characteristic of relative sterilization effect curves of three wave-length groups shown in Fig.3.3.2.

The first condition for photochemical reaction of an organism is that light is absorbed into a molecule. DNA absorbs the energy of the ultraviolet light of 260 nm wave length very well. With the irradiation of the ultraviolet light of 260 nm wavelength, the DNA molecule may become excited and lead to a large chemical change.

b. Lethal reaction[92]

The most important reaction of all is the dimerization of the pyrimidine base in the DNA molecule. The main cause of the death of an organism is now considered to be the formation of pyrimidine dimer by the ultraviolet light.

In addition, when ultraviolet light causes photohydration in the DNA, or when DNA and protein are in interaction, photo crosslinking occurs. These reactions may have some relation with ultraviolet light as a lethal factor.

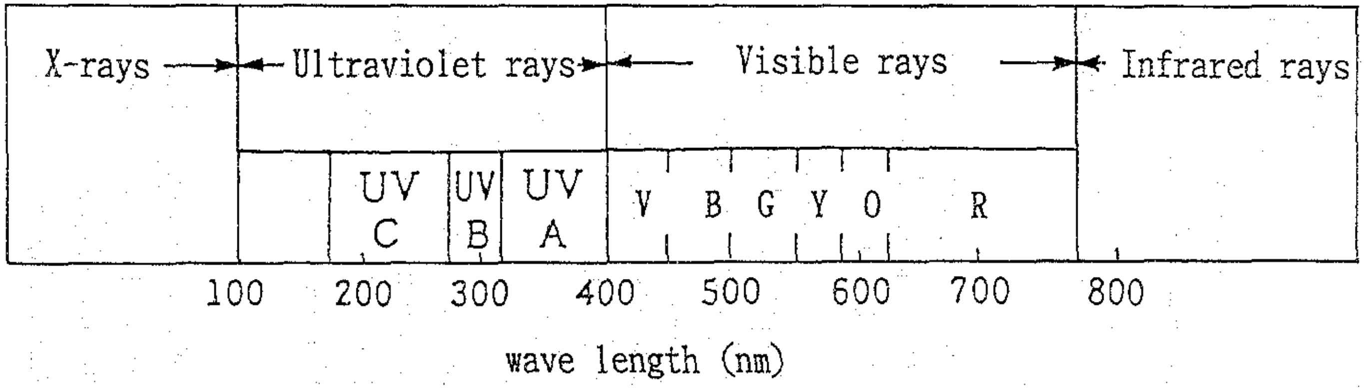


Fig. 3.3.1 Wave length and name of rays[86]

Table 3.3.1 Classification of ultraviolet rays[36]

CLASSIFICATION	WAVE LENGTH(nm)	ACTION
UV-A	320-400	Increase colouring matters in the skin by photochemical reaction
UV-B	280-320	Form red spots in the skin and subsequently turn the skin to be black Create vitamin
UV-C	180-280	Have a harmfull influence on the life Bring into existence of mutation of microorganism Cause sterilizing phenomena Create cancer cells in the skin of mouse

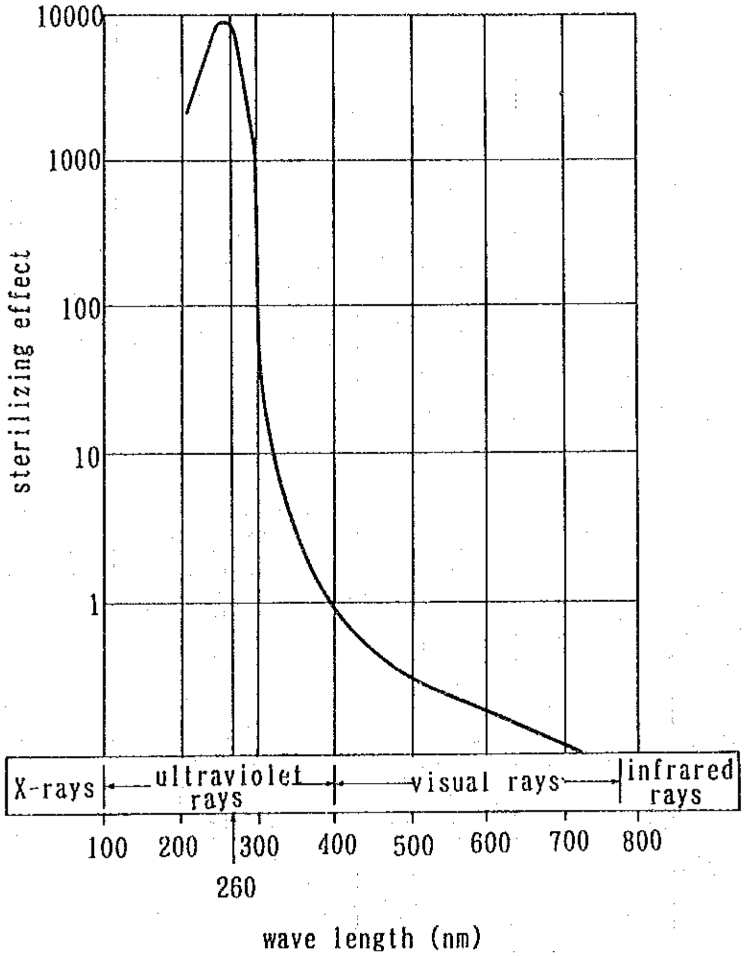


Fig. 3.3.2 Wave length and sterilizing effect[90]

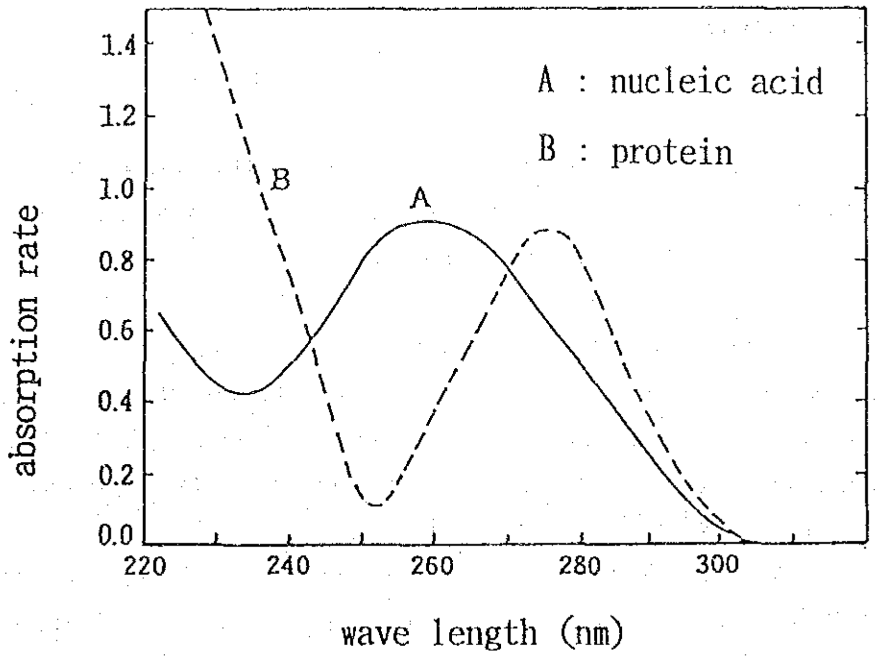


Fig. 3.3.3 UV absorption of nucleic acid and protein[91]

c. Disinfection of escherichia coli with ultraviolet light

Fig.3.3.4 compares the ultraviolet light disinfection effect for escherichia coli and the absorption characteristics for DNA. The absorption curve of wavelengths to be absorbed into DNA of escherichia coli cell shows a pretty good correlation with the ultraviolet disinfection effect.

d. Conclusion

DNA is the fundamental material for a living thing and is responsible for genetic phenomena and all of the important biological functions. Therefore it is concluded that changes or damages to DNA must be lethal[94].

(2) The Wavelength Distribution of the Low Pressure Mercury Lamp[95]

Ultraviolet disinfection is developed to make use of the prominent biological effect of the ultraviolet light. The low pressure mercury lamp artificially generates UV–C which has a several thousand times stronger disinfection effect than the sunbeam, and the ultraviolet light energy of 253.7 nm can be used efficiently for disinfection.

The wavelength distribution of the low pressure mercury lamp is shown in Fig.3.3.5. This lamp is designed to generate a 253.7 nm wavelength with almost 90% efficiency. The 253.7 nm wavelength has the most effective disinfecting property of all the energies generated by mercury resonance.

3.3.3 The Disinfection Effect by Ultraviolet Irradiation

The low pressure mercury lamp performs the efficient disinfection action, but its energy source has characteristics of light. To get desirable effects, it is necessary to consider "the disadvantages of ultraviolet light disinfection" mentioned before. Moreover there are various kinds of microorganisms with various shapes and features. The dose of the ultraviolet light required for disinfection depends on the types of microorganisms.

UV dose (mWsec/cm^2) = UV illuminance (mW/cm^2) x irradiation time (s)[96]

(1) Values of the Ultraviolet Dose Required for Killing Various Microorganisms

When irradiated at a certain illumination intensity of ultraviolet light, the survival rate of microorganisms is reduced logarithmically with the elapsed time in many cases. Therefore the ultraviolet light sensitivity of a microorganism is expressed by the dose value. Table 3.3.2 shows the ultraviolet light dose required for a 90% killing of various microorganisms. It is noted that a quadruple dose of the indicated value is required to raise the killing rate to 99%.

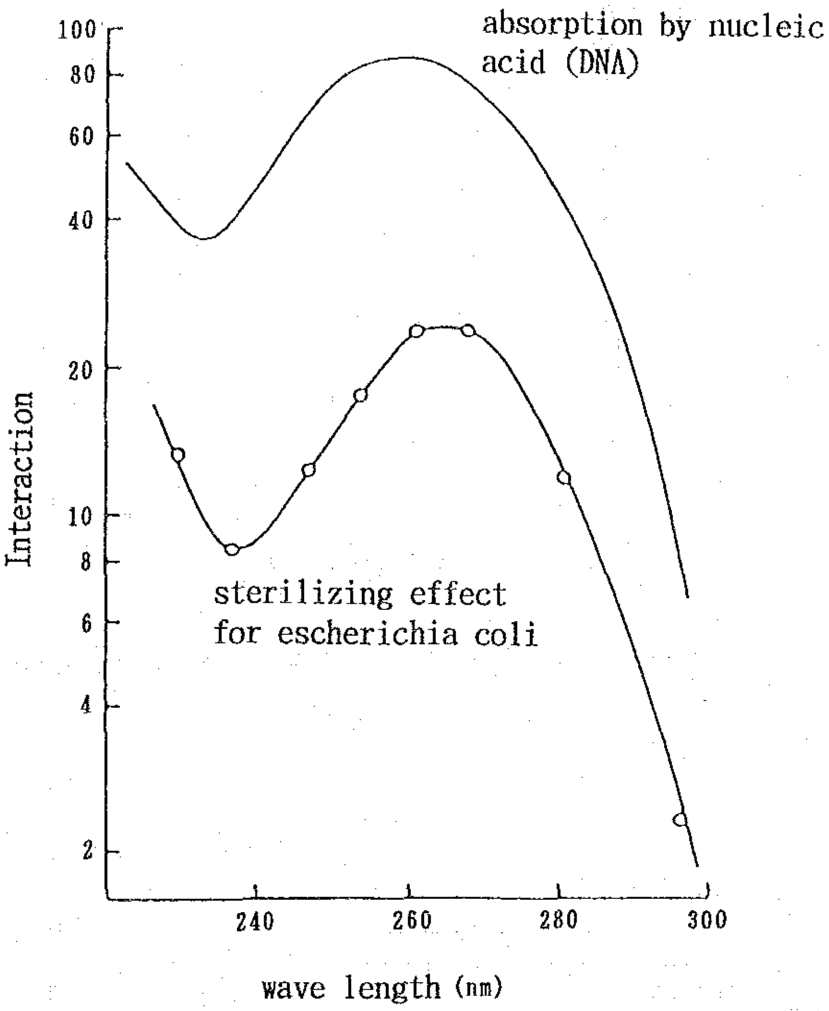


Fig. 3.3.4 UV sterilizing effect for escherichia coli and absorption by nucleic acid(DNA)[93]

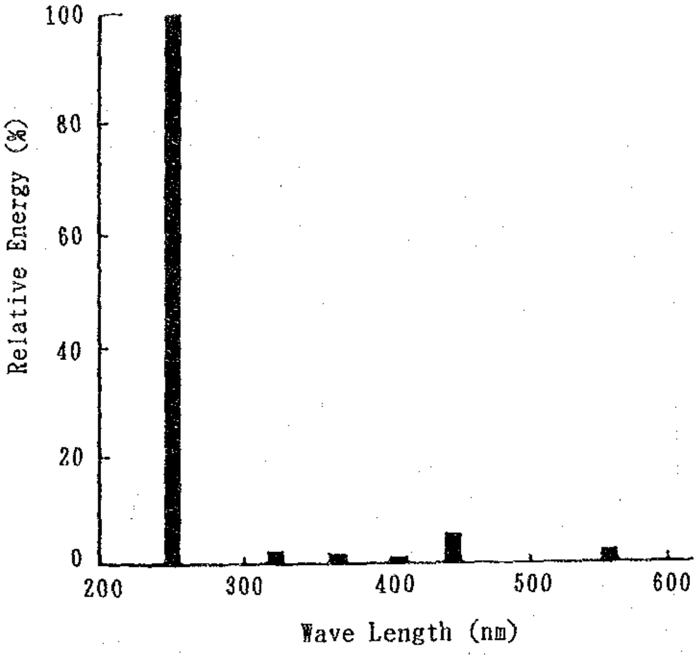


Fig. 3.3.5 Spectra diagram of low pressure mercury lamp[95]

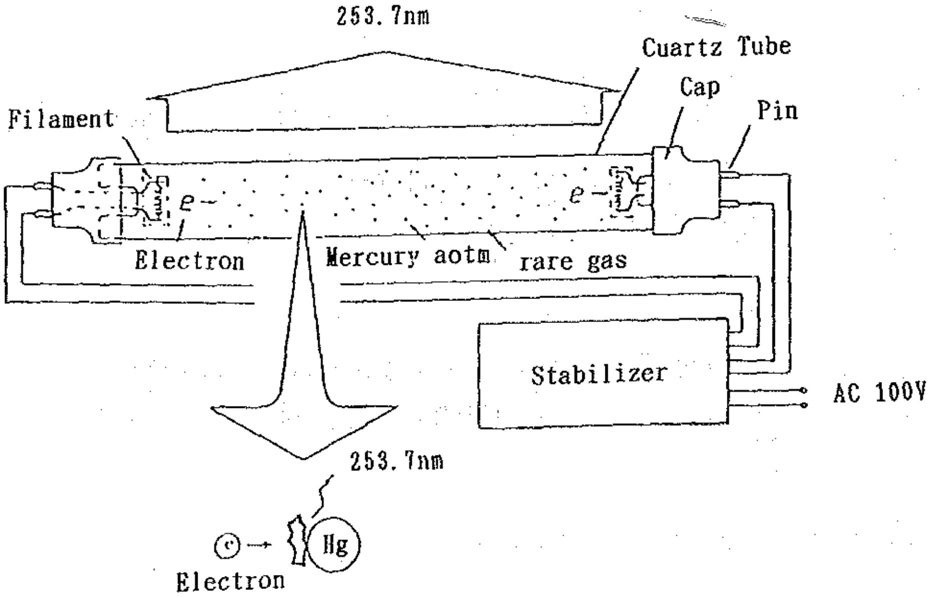


Fig. 3.3.6 Structure of low pressure mercury lamp (ex:ignition circulation)

Table 3.3.2 UV dose required for a 90% Killing of various microorganisms[97]

Organism	Dose*
Serratia marcescens	2.42
Pseudomonas aeruginosa	5.5
Mycobacterium tuberculosis	6.0
Salmonella enteritidis	4.0
Salmonella typhimurium	8.0
Shigella paradysenteriae	1.68
Escherichia coli	3.0
Proteus vulgaris	2.7
Bacillus subtilis (Cell & Spores)	7.1
Bacillus subtilis (Spores)	12.0
Clostridium tetani	4.9
Staphylococcus aureus	2.18
Streptococcus viridans	2.0
Streptococcus pyrogenes	2.16
Micrococcus candidus	6.05
Sarcina lutea	19.7
Micrococcus radiodurans	20.5
Yeasts (Average)	4.0
Algae, blue-green	300～600
Aspergillus nigar (bread)	100
Penicillium chrysogenum (fruits)	30～50

* Dose (mWsec/㎠) given above is for UV light at 254 nm required for a 90% killing. Multiply dose by 4 times for a 99% kill.

Table 3.3.3 shows the ultraviolet light dose required for a 99% killing of various microorganisms. The ultraviolet light sensitivity of gramnegative bacteria is generally stronger than that of spores. The quoted report indicates that the resistance of fungi is stronger than that of bacteria and algae is much stronger in resistance than fungi[99].

Table 3.3.3 UV irradiation volume, to be necessary for sterilizing various FUNGUS[99]

(μW·s/cm^2)

	(Sterilized Rate 99.9%)
BACTERIA (GRAM-NEGATIVE BACTERIA)	
Escherichia coli	6.600
Shigella dysenteriae	3.400
Pseudomonas aeruginosa	10.500
BACTERIA (GRAM-POSITIVE BACTERIA)	
Bacillus subtilis	11.000
Bacillus subtilis spores	22.000
Streptococcus hemolyticus	5.500
Staphylococcus aureus	6.600
Staphylococcus albus	5.700
YEAST	
Saccharomyces Sp.	17.600
Saccharomyces cerevisiae	13.200
MOLD SPORE	
Penicillium roqueforti	26.400
Penicillium expansam	22.000
Aspergillus niger	330.000
Aspergillus flavus	99.000
Rhizopus nigricans	220.000
VIRUS	
Influenza	6.600
Polio Type I	6.000
Coxsachie A2	4.800
Adeno virus Type III	4.500

(2) Selectivity for the Type of Bacteria

Apart from the report mentioned in the previous section 3.3.1, the technical report from Photoscience Japan Corporation emphasizes that ultraviolet light is the ultimate for disinfection and cites Fig.3.3.7[100] that UV disinfection is less selective and is able to kill all types of bacteria, presenting the comparison of the required dose of chlorine or ozone. Some other reports also take the same attitude to the ultraviolet light disinfection[101].

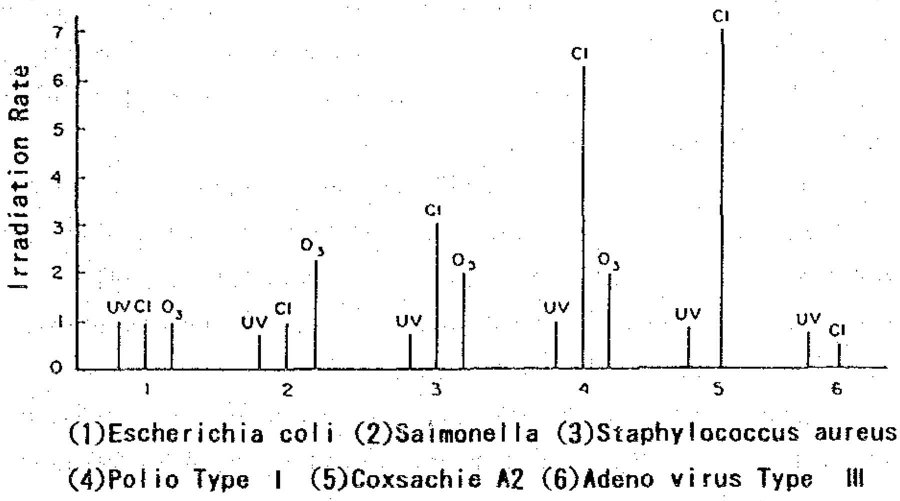


Fig. 3.3.7 Relative effect of UV against various Fungus[100]

(3) The Procedures to Decide UV Dose Required for Disinfection

The procedures to decide the ultraviolet dose required for disinfection is as follows. Initially obtain the absolute value of the ultraviolet illumination intensity as a parameter (based on the measurement of the actinometer), and then check the bacteria survival rate by irradiation time (s). In many cases the number of bacteria that survive decreases logarithmically with the irradiation time. Then from these values the ultraviolet light inactivation rate is formulated. With this rate, the ultraviolet dose required for extremely high rate disinfection can be estimated.

Fig.3.3.8 and Fig.3.3.9 show the inactivation rate of the typical bacteria, that is, pseudomonas aeruginosa, escherichia coli, bacillus subtilis, and bacillus subtils spore.

In designing the disinfection system, in addition to this basic data, the ultraviolet transmittance of the water to be treated, the ultraviolet distribution in the equipment, and the fluid mixture characteristics are, also counted to decide the ultraviolet dose required for disinfection. Therefore the types of target microorganisms, the quality and the temperature of the water to be treated, and other requirements should be carefully considered to develop the system for practical use.

(4) The Impact of Water Quality to the Ultraviolet Transmittance[103]

The ultraviolet light transmittance depends on the water quality, which in turn affects the ultraviolet light disinfection effect. In other words, in the water of low ultraviolet transmittance, the irradiated ultraviolet energy is attenuated by the raw water before reaching the target microorganisms, losing the UV dose required for disinfection.

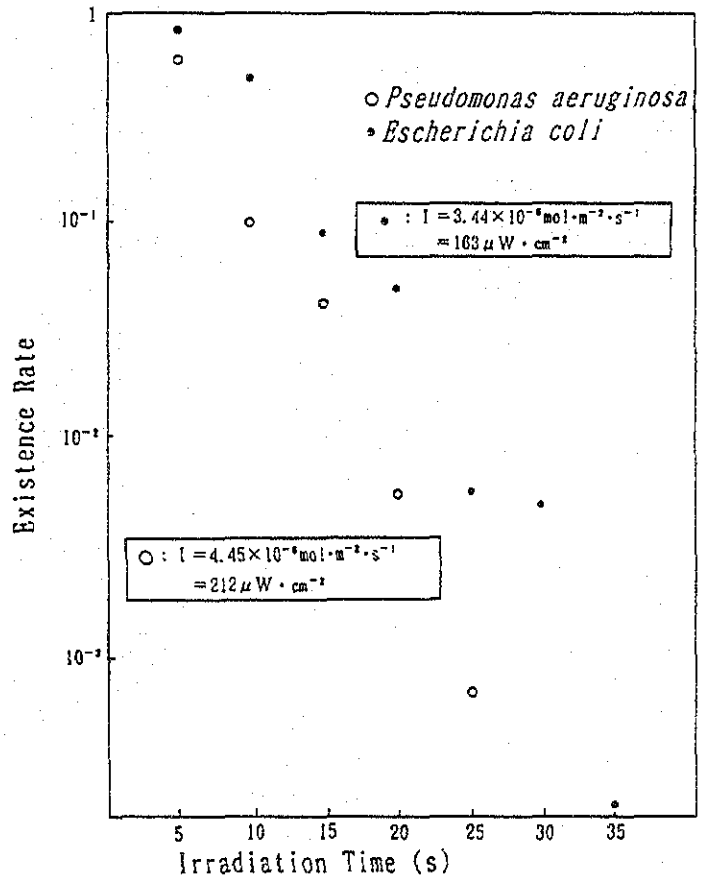
2/3 of the ultraviolet light is absorbed at every 1cm depth for the river water, and at every 5 m for the clean water, 2/3 of the ultraviolet is absorbed. When the water contains much ultraviolet absorbent material such as minute particles, iron, or organic substances, the ultraviolet light disinfection effect extremely gets attenuated. Fig.3.3.10[104] shows the ultraviolet transmittance of the various kinds of liquid and at various depths.

The relation between the ultraviolet light tranmittance and the liquid depth is expressed by Lambert–Beer's law. With measurement of one point, the ultraviolet dose attainable at a certain depth can be estimated.

$$\mathrm{Log}_{10}\, I_0/I = \alpha l$$

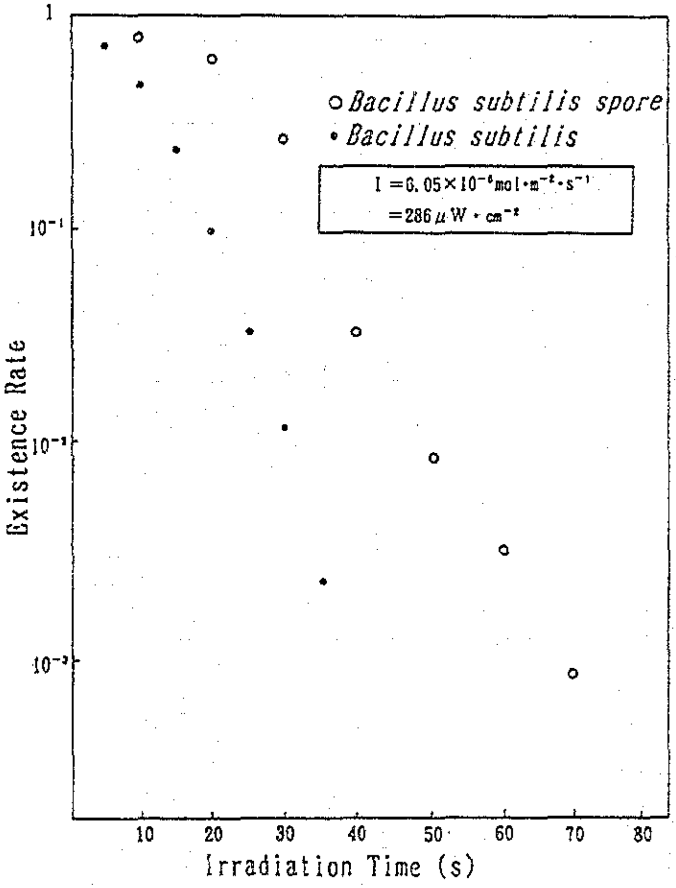
αl: absorbance
I: liquid depth
α: absorptivity
c: concentration of the target
I_0/I: transmittance   substance

Although there is no problem for pure water of 100%, ultraviolet light transmittance for other raw water, it is necessary to measure the ultraviolet light transmittance and then set the ultraviolet dose required to disinfect the volume of water to be treated.



(Irradiation Degree : $212\ \mu W \cdot cm^{-2}$: $163\ \mu W \cdot cm^{-2}$)

Fig. 3.3.8 Inactivation rate of bacteria by UV irradiation[102]



(Irradiation Degree : $286\ \mu W \cdot cm^{-2}$)

Fig. 3.3.9 Inactivation rate of bacteria by UV irradiation[102]

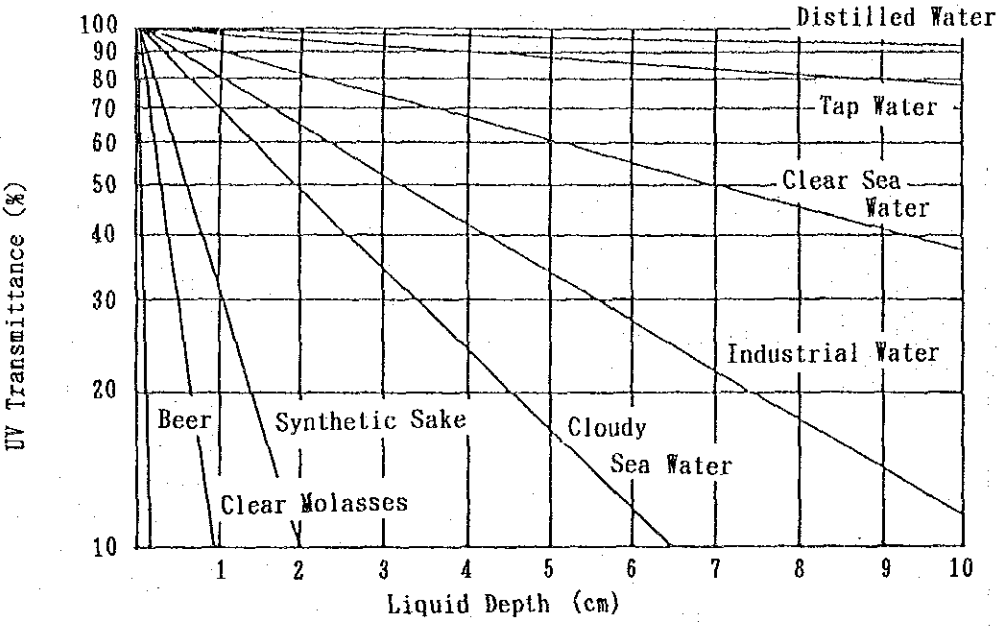


Fig. 3.3.10 UV transmittance of various liquids[104]

(5) The Duration of Disinfection Effect[105]

The ultraviolet light disinfection effect does not last long, as mentioned above. When the process line after disinfection is dirty, bacteria may enter again. To avoid secondary contamination, the lines after disinfection should always be kept clean. Above all it is necessary to use the disinfection system just before the point of use. In many cases ultraviolet light disinfection is utilized, this type of process is adopted in practice.

3.3.4 The Characteristics of the Low Pressure Mercury Lamp

(1) The Characteristics of the Temperature in Disinfection with Light Energy

As shown in Fig.3.3.6, the wavelength of the ultraviolet light radiated from the low pressufe mercury lamp, in which mercury is enclosed, varies with the vapor pressure of mercury. As the vapor pressure is higher, the quantity of the long wavelength light gets larger and the radiation quantity of the disinfection light of 253.7 nm wavelength gets smaller[106].

Fig.3.3.11[107] shows the change of the radiation quantity with the change of the tube temperature, and indicates that the mercury vapor pressure varies remarkably with the ambient temperature or the temperature of the lamp tube.

The general low pressure mercury lamp has the characteristic to radiate the disinfection light most efficiently at 40°C of the lamp tube temperature[108]. The data in Fig.3.3.11 are reported to show that the temperature characteristics can be changed if the filler gas or the lamp shape is improved. The report also gives an example of the ultra pure water production process and indicates that the disinfection lamp for medium temperature works efficiently because when the water temperature on the ultra pure water production line is 20–25°C, the tube temperature of the low pressure mercury lamp of the system rises higher than 50°C.

Photoscience Japan Corporation, which submitted the technical reports on the ultraviolet light disinfection, presents Fig.3.3.12[109] to show that the change of the ultraviolet light radiation resulting from the fluctation of the ambient temperature of the lamp can be held extremely small by enclosing special alloy (amalgam) in the lamp, and thus controlling the mercury vapor pressure which changes accompanied with the temperature fluctuation.

(2) The Compensation of the Ultraviolet Illumination Intensity for the Water Temperature

As the ultraviolet radiation from the low pressure mercury lamp depends on the ambient temperature, it is necessary to pay attention to the water temperature when utilizing an internal radiation type in which the lamp is installed in the liquid. It is required to control the ambient temperature of the lamp or to compensate the illumination intensity, when the temperature of the water to be treated is extremely high or low[111].

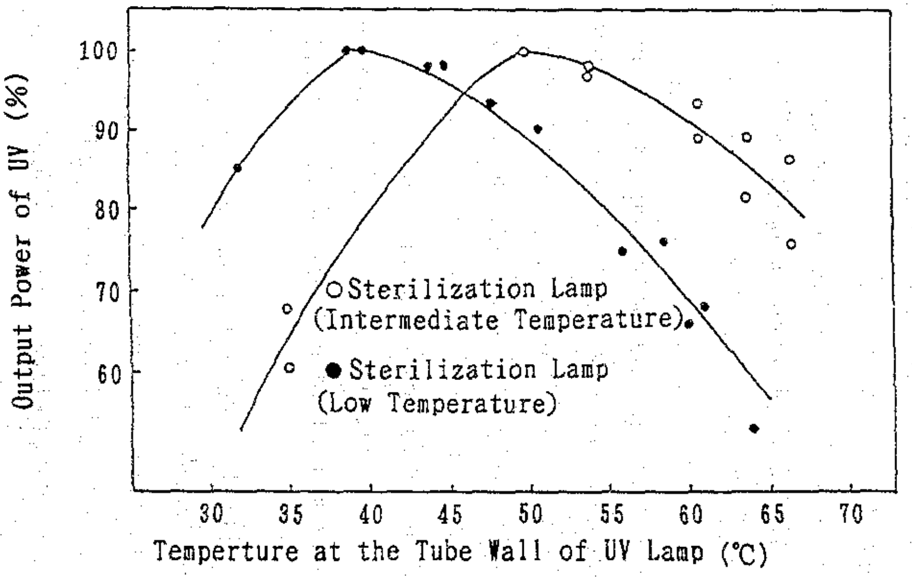


Fig. 3.3.11 Relationship between temperature at the tube wall of UV lamp and output power of UV[108]

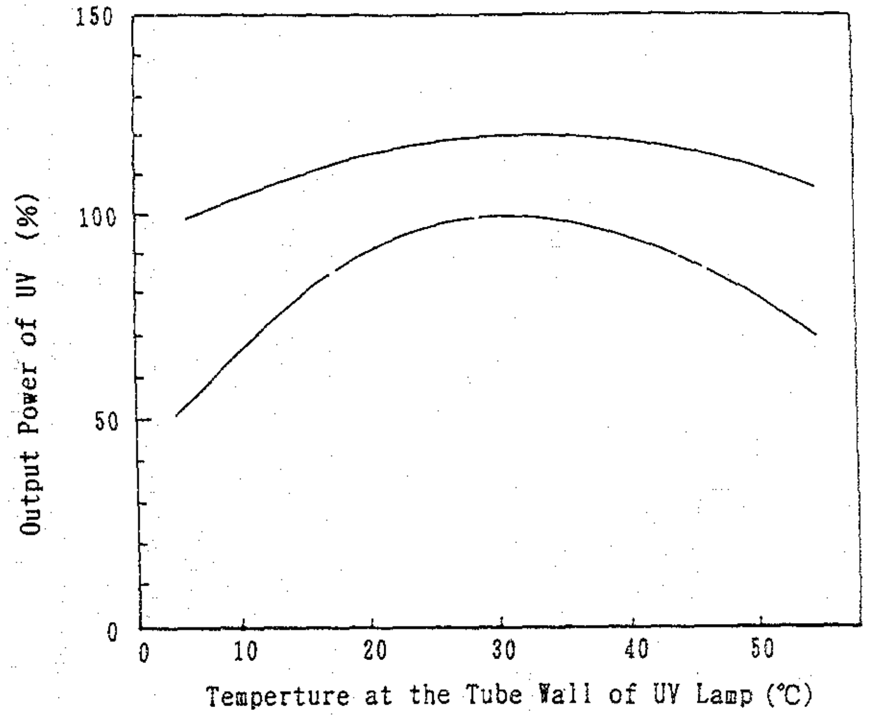


Fig. 3.3.12 Temperature characteristic curve[109]

(3) Basic Data Required for Designing the Disinfection System[112]

As mentioned before, the ultraviolet dose can be calculated by multiplying the ultraviolet illumination intensity by the irradiation time. The value of the ultraviolet dose for disinfecting microorganism thus calculated should be attained from the system in practical use. The illumination intensity distribution is calculated based on data such as the arrangement of lamps and the ultraviolet light transmittance of the treated water. The distribution of the time of irradiation to the treated water in the irradiation area, that is, the holdup time shows the hydraulic characteristic. It depends on the arrangement of the outer silica tube, the shapes of the inlet and the outlet, whether the reflux panel is used or not, and Reynold's number. The distribution of the holdup time should be obtained by measuring with the tracer.

Fig.3.3.13 shows the elapsed time after the tracer injection and the concentration distribution of the effluent tracer for a good and a bad examples of the flow.

(4) The life of the low pressure mercury lamp[113]

Like a common fluorescent lamp, the life of the low pressiure mercury lamp is limited, generally to one year. The ultraviolet radiation gradually decreases with the operating hours. This is owing to the drop of ultraviolet light transmittance resulting from degradation of the lamp tube material and the darkening of the tube inside caused by vapor of the electrode materials and others. Photoscience Japan Corporation states that they designed the system, setting the replacement time (life) of the lamp at 70% of the initial illumination intensity (after 8,000 – 8,500 hours by continuous lighting), in order to keep efficient performance. Fig.3.3.14 shows a example of decline curve on sterilizing ray of low pressure UV lamp.

3.3.5 The Types and the Structures of the UV Disinfection System

There are mainly 2 types of system, the outer irradiation type and the internal irradiation type. For the outer irradiation type, the ultraviolet light is irradiated from the outside of the treated water, while for the internal irradiation type, the lamp is soaked in the treated water in which the ultraviolet light is irradiated. Each of the types has its own advantages and disadvantages and has been used according to the purposes.

(1) The Outer Irradiation System

<1> Advantages[115]

a. The impacts of the temperature and the quality of the treated water are smaller and the disinfection effect is stable.
b. The outer silica tube is hard to get dirty because the lamp is not in direct contact with the treated water.

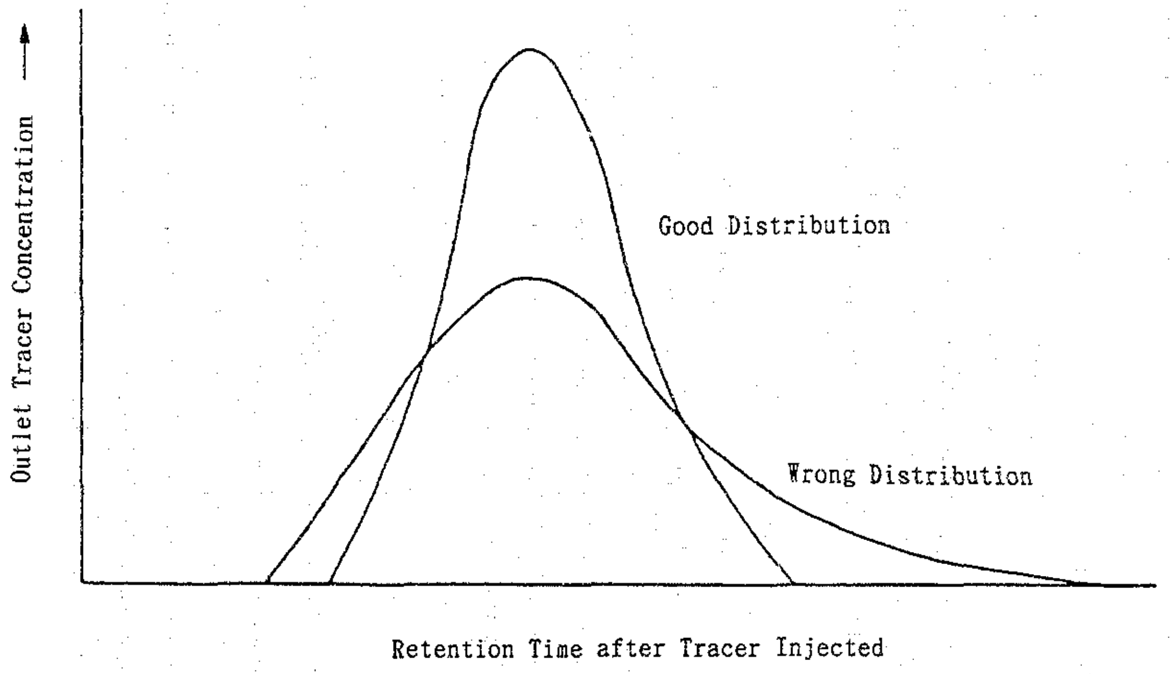


Fig. 3.3.13 Resident time distribution measured by tracer test[112]

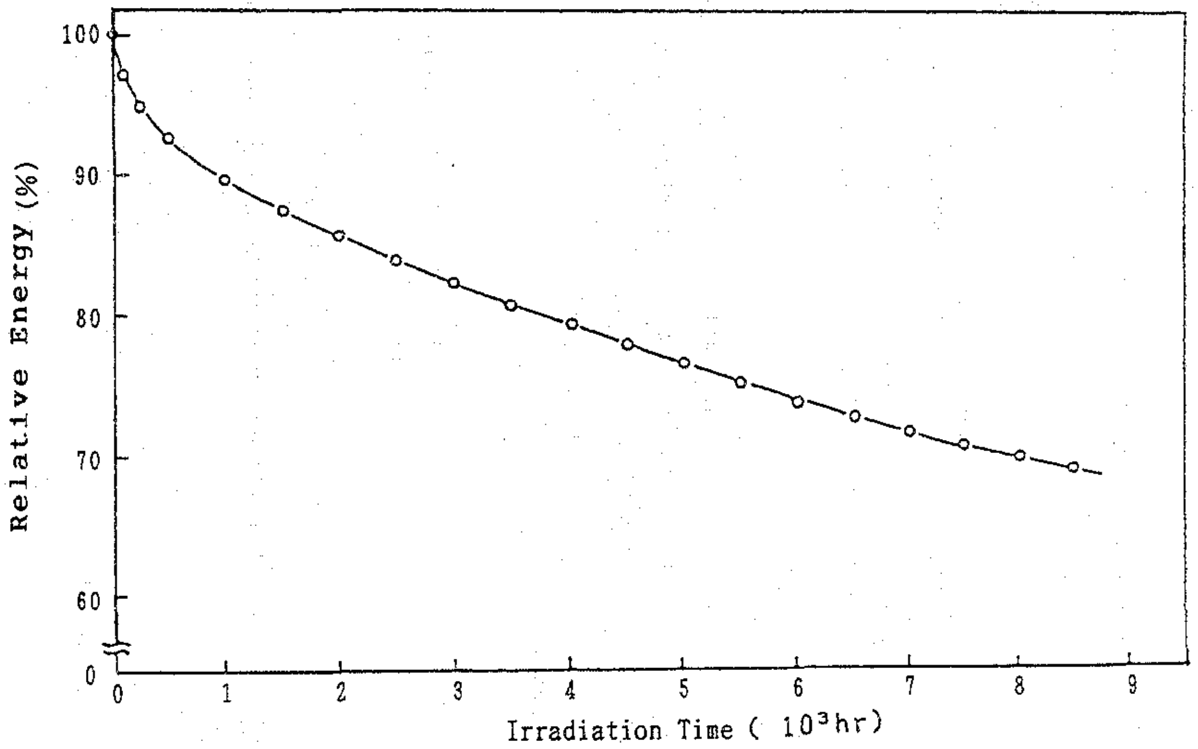


Fig. 3.3.14 Examples of decline curve on sterilizing ray of low pressure UV lamp[113]

c. The structure is relatively simple and it is easy to maintain.

<2> Disadvantages[116]

The energy efficiency is rather worse than for the internal irradiation system.

<3> Examples[117]:

a. Flonfine type

As shown in Fig.3.3.15, in this type of system, the treated water flows in the ultraviolet light transmissible tube (made of teflon resin) and the ultraviolet light is irradiated from the outside of the tube. Maintenance in the sewage treatment can be improved greatly with this type.

b. Trough type

As shown in Fig.3.3.16, the treated water flows through the trough and is irradiated by the lamp installed over the trough. This type is effective for disinfecting the water of low transmittance. This is a medium or small size disinfection system.

(2) The Internal Irradiation System

<1> Advantages

The energy efficiency is better because the lamp is installed inside of the treated water. This is a very popular system.

<2> Disadvantages

The advantages of the outer irradiation system are regarded as the disadvantages for the internal irradiation system.

<3> Examples[118]:

a. Sterifine type

As shown in Fig.3.3.17, this type of system with its structural feature can be used at high hydraulic pressure and is effective for disinfection of high level water such as pure water.

b. Open channel type

As shown in Fig.3.3.18, this type of system can be used in the gravity flow and for large scale disinfection with many units arranged horizontally. To make use of the ultraviolet disinfection treatment for desalination of the seawater, this type will be selected.

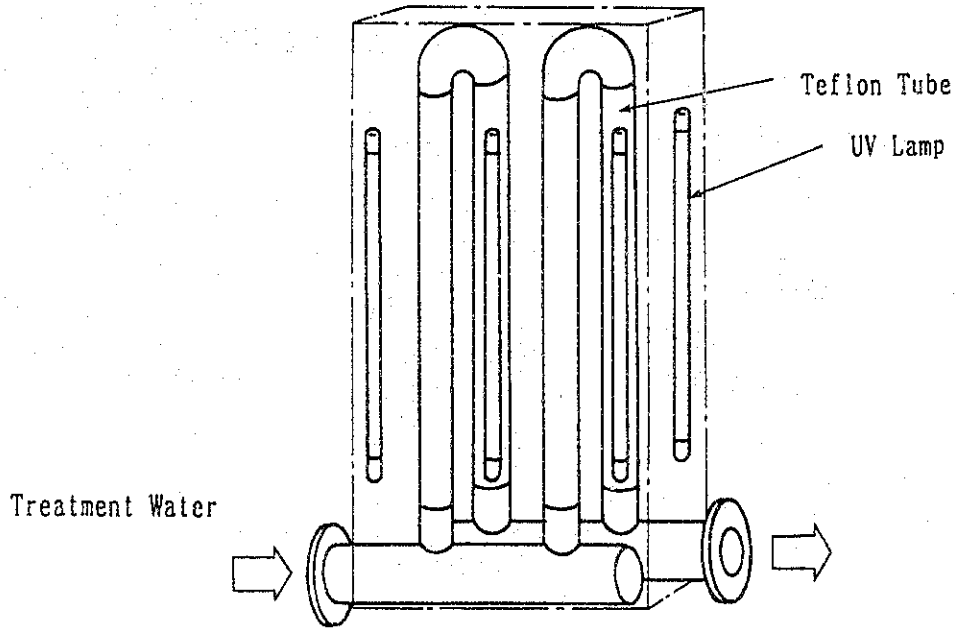


Fig. 3.3.15 Flonfine type UV disinfection system[117]

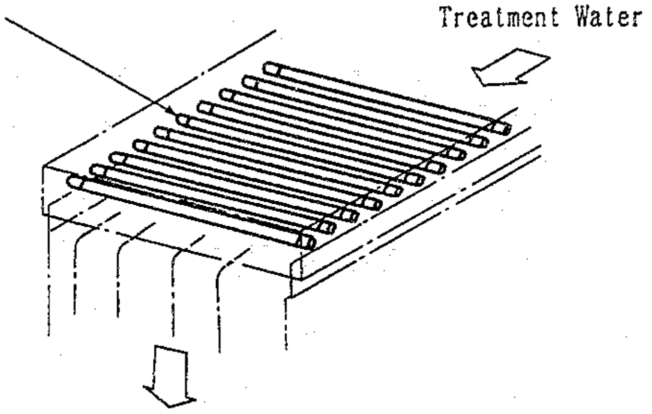


Fig. 3.3.16 Trough type UV disinfection system[117]

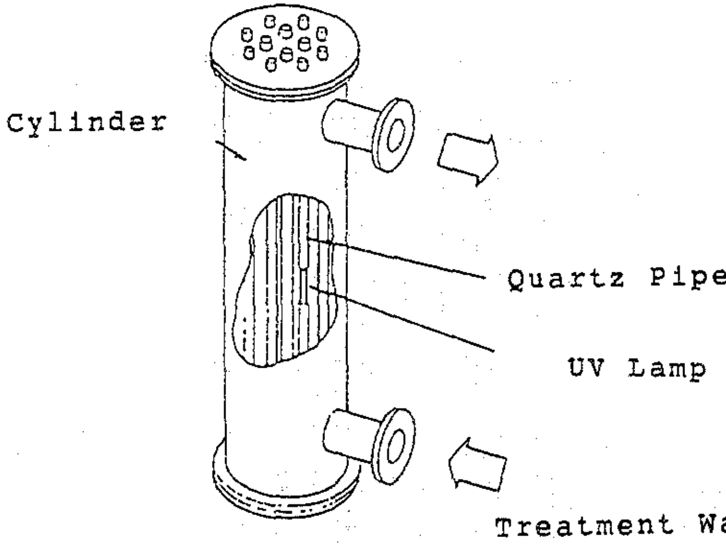


Fig. 3.3.17 Sterifine type UV lamp[118]

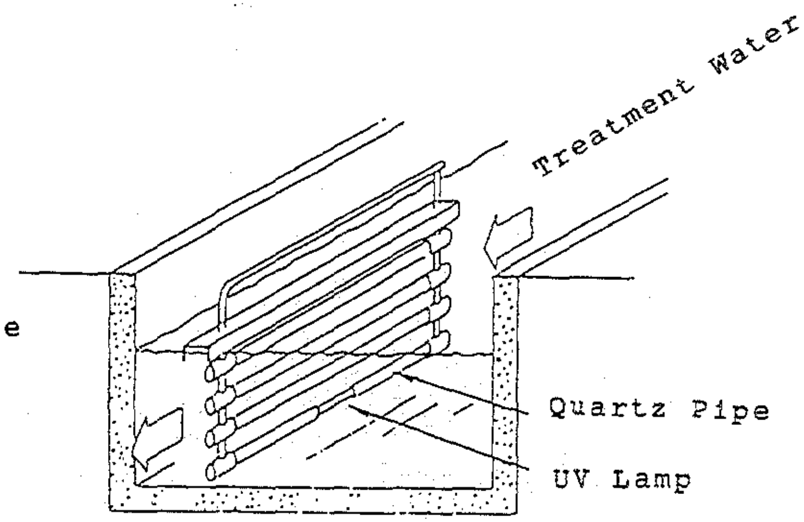


Fig. 3.3.18 Open channel type UV lamp[118]

3.3.6 The Cost for the Ultraviolet Light Disinfection

(1) Prerequisites For System Designing And Cost Of Equipment

<1> Introduction

Ultraviolet light has no selectivity to the types of bacteria, and does not cause the generation of harmful materials as the chemical treatment. Neither does it cause secondary water contamination. Ultraviolet light disinfection is therefore essential technology for various water treatment, in the most advanced industries such as electronics industry, biotechnology, medical engineering, etc. and in other fields. It has been paid much attention as the more efficient disinfection for the next generation as mentioned before, There are, however, few examples of ultraviolet light applications to disinfection for RO membrane desalination of seawater.

In considering the application of ultraviolet light to the disinfection system in the RO membrane desalination plant, we obtained information about the possibility of a practical system, the cost of equipment, the operating cost, etc. from Photoscience Japan Corporation. The report in this section is created based on the information obtained from Photoscience Japan Corporation which has unique technologies and the commercial base results in the ultraviolet disinfection field.

The name and the location of the researched manufacturer:

Photoscience Japan Corporation
Nisshin Bldg. 17-21 Nishi-Waseda 3 Chome,
Shinjuku-ku, Tokyo

<2> Prerequisites Of The System Design

Raw water quantity to be treated: 100,000 T/D
Water production: 30,000 T/D
Raw water intake: sand filter (for the installation in Saudi Arabia)
Target bacteria: Escherichia coli and vibrioid class, excluding Phytoplankton whose energy consumption is large.
Point of UV irradiation: In front of the RO membrane, not only to disinfect the raw water but also to improve the effect and the maintenance of the RO membrane.
Operating hour: For 24 hours a day

<3> The Relevant Equipment And The Cost Of The Equipment

a. Equipment

When the UV light is used for the disinfection system of the seawater desalination plant,