Chapter 6
Strategies and Limitations of Water Treatment Methods for Point-of-Use Application

N. R. Srinivasan, M. Kamaraj, and S. Venkatesa Prabhu

Abstract Water-related diseases threaten human survival across the globe. This is because of the severe contamination of water all over the world. Water is being contaminated due to the rapid growth of population and industrialization. As a result, the demand for potable water is increased in many developing countries. The surface sources for potable water are rivers, lakes, and aquifers. Particularly, to ensure its quality, water obtained from these sources should be free from chemical and biological contaminants. In developing countries, most of the people (60%) depend on the groundwater source. However, groundwater needs to be treated as the soil does not remove all the contaminants. Moreover, in various parts of the world, water supply is done through the network of pipelines to the individual consumers from water treatment plants. The quality of water may get degraded due to the formation of rust while flowing through pipes. Chlorination is a widely adopted method in many countries for water disinfection. It exhibits low disinfection efficiency and results in the generation of toxic by-products. This chapter describes the strategies and limitations of water treatment methods to remove both chemical and biological contaminants for point-of-use applications.

Keywords Chemical disinfectants · Physical disinfectants · Filtration methods · Comparison of treatment technologies

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1 Introduction

On the earth's surface, approximately two-thirds of the portion is full of water in the form of oceans and freshwater. The contribution from the oceans is around 97.2% of its availability, whereas the freshwater source contributes about 2.7%, of which 0.35% is contaminated due to various anthropogenic activities (Akshay et al. 2020). In general, water contamination occurs due to the presence of undesirable chemicals, microbes, and suspended solids. The presence of these contaminants in water could lead to waterborne diseases such as dysentery and cholera. Water scarcity and contamination are major problems in most of the places across the world. According to the World Health Organization report (WHO), 844 million people do not have safe water all over the world (WHO/UNICEF 2017).

The main reasons for water contamination are climate change, population, and industrialization (Okello et al. 2015). It is necessary to provide cost-effective water treatment technologies for low-income countries. To have safe and drinkable water, the WHO set guidelines for centralized and decentralized water treatment systems. Figure 6.1 shows the water treatment process involved in a centralized system to provide safe water. In most of the countries, water treatment plant undergoes various physicochemical processes such as sedimentation, coagulation/flocculation, etc. (Matsumoto et al. 1995). This chapter discusses the strategies and limitations involved in both chemical and physical disinfectants.

2 Chemical Disinfectants

2.1 Chlorination

The pathogenic microorganisms (viruses, bacteria, and protozoa) are of utmost concern in water treatment. Over the years, chemical control of microorganisms is the most important and widely accepted method for a water treatment plant. Chlorine compounds such as chlorine gas, chlorine dioxide, chloramines, and hypochlorite are widely used to treat water (Zhang et al. 2012). Particularly, the addition of chlorine in water reacts with organic matter, reducing agents and ammonia. Chlorination is preferred not only for killing microbes and also for odor removal in water. One of the main drawbacks of chlorination is toxic to humans if the concentration exceeds above the permissible limit. During chlorination, chlorine reacts with natural organic matter (NOM), iodide (I^-), and bromide (Br^-) to generate various harmful disinfection by-products (DBPs), including trihalomethanes (THM), haloacetic acids, etc. (Richardson et al. 2007). The predominant organic matters in water are humic and fulvic acids. The formation of DBPs is influenced by chlorine dosage, contact time, organic matter, pH, temperature, etc. Based on the animal and epidemiological studies, these by-products may produce adverse effects on humans (carcinogenicity or cytotoxic). The required concentration of chlorine for bacteria,

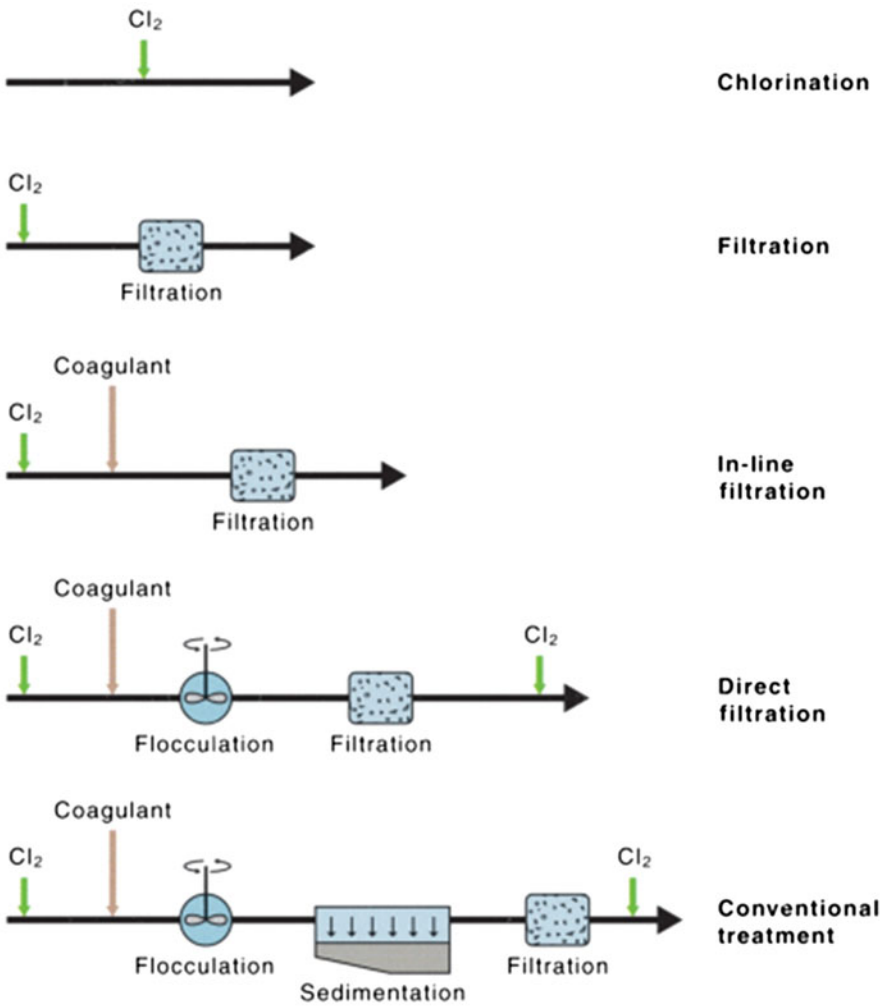

Fig. 6.1 Water treatment process (Adapted from Environmental Microbiology, Academic Press, San Diego, CA, 2000)

virus (poliovirus), and parasites are 0.1, 1, and 5 ppm, respectively (Bitton 2014). Moreover, the microbicide occurs when chlorine interacts with the surface of the pathogen. The mechanisms by which the disinfection occurs are the damage of cell surface, oxygen uptake, and oxidative phosphorylation, inhibiting the enzyme activity and physical damage to DNA. According to the WHO, the maximum value for chlorite in drinking water should not exceed 0.2 mg/L (Twort et al. 2000).

Generally, chlorine is added as a gas (Cl_2) and salt (sodium hypochlorite). Chlorine gas is rapidly hydrolyzed in water to form hypochlorous acid (HOCl), which further dissociates and generates hypochlorite ion (OCl^-) according to

Eqs. (6.1) and (6.2). These products form at different pH (particularly, HOCl is at pH – 5.0 and OCl^- is at pH –10.0). These free chlorine-based compounds react with both inorganic and organic matter present in water to form chlorinated compounds.

$$Cl_2 + H_2O \leftrightarrow HOCl + H^+ + Cl^- \tag{6.1}$$

$$HOCl \leftrightarrow H^+ + OCl^- \tag{6.2}$$

Hypochlorous acid reacts with ammonium ion to produce monochloramine, dichloramine, and trichloramine according to Eqs. (6.3), (6.4) and (6.5). The formation of different types of chloramine depends on the pH value and the concentration of NH_3 in water.

$$NH_3 + HOCl \rightarrow NH_2Cl + H_2O \tag{6.3}$$

$$NH_2Cl + HOCl \rightarrow NHCl_2 + H_2O \tag{6.4}$$

$$NHCl_2 + HOCl \rightarrow NCl_3 + H_2O \tag{6.5}$$

The term "free chlorine" is the combination of hypochlorous acid and the hypochlorite, whereas chloramines are called combined chlorine. Moreover, these chlorinated compounds have disinfection ability to oxidize sulfides (S^-), ferrous (Fe^{2+}), and manganese (Mn^{2+}) ions. As a result of impurities in water, chlorine demand is assessed based on the concentration of residual chlorine in the water. The breakeven point occurs when the amount of chlorine increases beyond the point where the residual chlorine is detected. It generally occurs in the pH range of 7.0–7.5.

The effective dosage of chlorine concentration is at the breakpoint or slightly higher. The concentration should be enough to break chemical bonds of contaminants as well as destroy pathogens. It is important to keep free available chlorine residual by supplying an excess quantity of chlorine for disinfection. Figure 6.2. shows the breakpoint chlorination curve in different stages. In the initial stage, there is no free residual chlorine as complete oxidation occurs (Point A). Next, the curve is linear until point B as the formation of chloramines and chloro-organic occurs at this stage. Furthermore, oxidative destruction occurs with an increase in the concentration of chlorine. After that, the number of residual chlorine decreases and reaches a point called breakpoint (Point C). Then, the residual chlorine keeps on increasing as the concentration increases (Point D). The breakpoint concentration varies with the quality of raw water (Al-Abri et al. 2019).

Table 6.1 shows the concentration of chlorine-based disinfectants and contact time required for the inactivation of *E. coli*. In the water treatment plant, chlorine is added in the form of elemental chlorine (chlorine gas), sodium hypochlorite, and calcium hypochlorite. Each form has distinct advantages and disadvantages of water disinfection (Table 6.2).

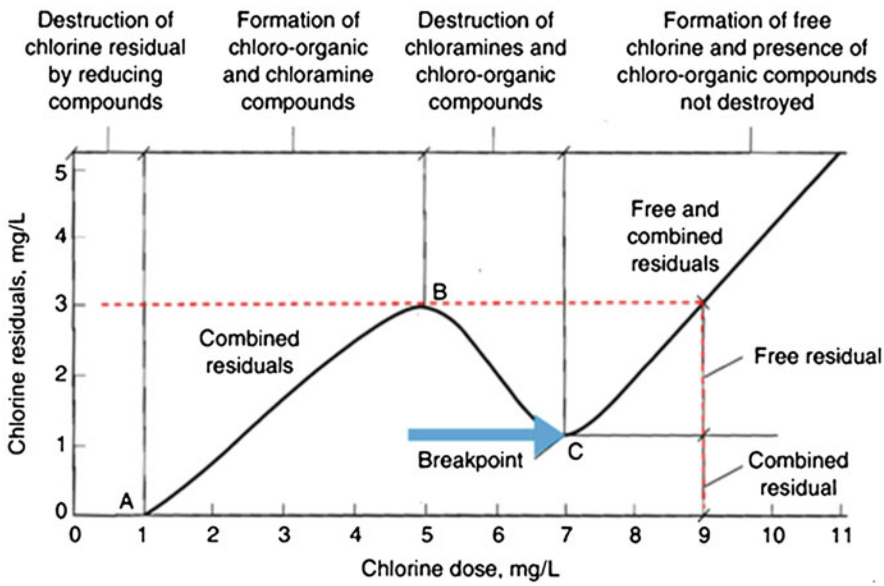

Fig. 6.2 Breakpoint chlorination curve (https://naturalpoolproducts.com/category/main/)

Table 6.1 Contact time required to inactivate *E. coli* (Hall and Hyde 1992)

	Concentration (ppm)	Contact time, min	pH	Temperature, °C
Free chlorine	0.1	0.4	6	5
	0.1	6	8.5	20–29
	0.4	1	8.5	20–29
Monochloramine	0.1	60	4.5	15
	1	6	4.5	15
Dichloramine	1	64	9	15

Although chlorination remains the most widely accepted method, the water treatment system may choose either one of the chlorine-based disinfection or their combination.

2.2 *Iodine*

Iodine is a strong oxidant, which is more effective at a lower dosage and less sensitive to pH and organic matter present in water. The active disinfectants are elemental iodine and hypoiodous acid. The main disadvantage is that it could create allergic reactions to any individual. The recommended concentration is 150ug/day for adults. The concentration below 1 mg/l is effective for bacteria. However, the same concentration will take a few hours to kill *Giardia cysts*. Iodine has more

Table 6.2 The advantages and disadvantages of chlorine-based chemical disinfectant

S. No	Chlorine-based disinfectants	Advantages	Disadvantages
1.	Elemental chlorine	Cost-effective	Hazardous and highly corrosive
2.	Sodium hypochlorite	Less hazardous	High cost, formation of chlorite and chlorate, storage difficulties, limited shelf life
3.	Calcium hypochlorite	More stable than sodium hypochlorite	Formation of by-products like chlorite and chlorate, higher cost than elemental chlorine
4.	Chloramines	Formation of trihalomethanes and brominated products, more stable than free chlorine	Weak oxidant, hazardous to humans and aquatic life
5.	Chlorine dioxide	Effective method as compared to chlorine, no formation of by-products like trihalomethanes	Formation of by-products like chlorite and chlorate, may exhibit taste and odor problems, high operating cost

advantages, such as chemical stability and less reactivity with organic nitrogenous contaminants. The required concentration shoots up when the turbidity increases. It is considered to be more effective for water containing sludges. The usage of iodine as a disinfectant is restricted because of the cost, and it cannot be applied to a large system (White 1992; Howard et al. 2000).

2.3 Silver

Recently, silver has been proven to be an effective antibacterial agent against microbes since the times of ancient Greeks. It can be added in the form of salt (silver nitrate), colloidal suspension, and a small bed of silver (Srinivasan et al. 2013). For effective usage, silver can be deposited on the surface of porous carbon to inactivate microbes. Mostly, the support could be either ceramic or polymeric materials. These kinds of systems have been established against both gram-positive and gram-negative bacteria, which shows an almost 100% reduction in bacterial colonies. In this case, several mechanisms have been proposed in the literature to inactivate microbes such as cell wall rupture, generation of reactive oxygen species, and deactivation of proteins (Klueh et al. 2000). The use of silver as a disinfectant has become popular in most of European countries and the USA, and it is accepted for the bacteriological quality of stored water.

2.4 Potassium Permanganate, Ferrates, and Ozone

Potassium permanganate is also a strong oxidizing agent that oxidizes iron and manganese ions and removes odor and taste. It is reported that it has antibacterial properties against *Legionella pneumophila* (Yahya et al. 1989). Moreover, it is also considered as a poor disinfectant because the concentration required to inactivate microbes is too high (1 g/L). Bromine has the potential to control microbes in places where a small area is to be disinfected (e.g., swimming pool). It works well by forming hypobromous acid (HBrO) and other compounds like bromamines, which can be potent antimicrobial products. The addition of bromine in water disinfection is costlier than chlorine-based compounds. The main disadvantage is its reactivity with ammonia or other amines that may hinder its effectiveness. The mechanism behind the inactivation of microbes is oxidative stress (Taylor and Butler 1982). Ferrates react with reducing agents and various compounds that can effectively inactivate microbes (Audette et al. 1971). It is reported that ferrates can kill *Pseudomonas* over a range of concentrations (0–50 ppm) (Murmann 1975). Interestingly, zero-valent iron nanoparticles are considered as an alternative to permeable reactive barriers. It is reported that nanostructured iron adsorbs on the viral capsid, leading to inactivation (Ryan et al. 2002).

Ozone is one of the strongest disinfectants and oxidants that can remove color and turbidity and inactivates microorganisms. It is generated by passing dry oxygen or air through high voltage electrodes. In literature, the inactivation of *Salmonella* is reported with a 5 and 6 log removal at a concentration (2 mg/L) after the interaction time of 45 and 60s, respectively (Jamil et al. 2017).

This process does not produce chlorinated THMs or HAAs, and it reacts with bromine-containing compounds and forms toxic contaminants like bromate and brominated organics. Ozone breaks down complex organic molecules, and smaller size molecules can increase the growth of microbes and disinfection by-products during the processes. The mechanism behind the inactivation of microbes by ozone is the attack on the double bonds of the lipid layers in the cell membrane (Smith 1967).

2.5 Coagulation

Coagulation involves the addition of chemicals to remove dissolved and suspended solids by filtration and sedimentation. The addition of coagulating chemicals (e.g., alum) will increase the settling rate of particles by combining smaller particles into flocs. The required concentration ranges from 10 to 30 mg/L of water. After the addition of chemicals, at least 30 min is required to settle at the bottom, and then the clear water above the flocs can be decanted off.

3 Physical Disinfectants

Chemical disinfectants protect humans against waterborne microbial diseases. However, it has several disadvantages, including the formation of toxic by-products, taste, and odor problems. Moreover, in the longer term, there is pressure on the industries to reduce the production of chlorine-based disinfectants for environmental issues. In a commercial scale, membrane-based processes are available to remove microbes as well as chemical contaminants. It operates without chemical disinfection, or at least to reduce the number of chemicals used for disinfection. Further, pressure-driven membrane processes (microfiltration, ultrafiltration, nanofiltration, and reverse osmosis) can remove chemical contaminants and microbes based on their pore size. From the point of toxicological issues, this membrane process would prevent the formation of disinfection by-products and undesirable chemicals. The main issues in the membrane-based processes are the removal efficiency of microbes and biofouling. As an alternative to chemical disinfection, UV radiation is also capable of removing bacteria and viruses. This part describes the principles involved in the physical processes.

3.1 Ultraviolet Radiation

UV radiation energy waves ranges from 100 to 400 nm. The optimum UV range is between 245 and 285 nm for germicidal effects. This process does not produce any toxic by-products or taste and odor problems. It has several demerits such as high cost as compared to the chemical disinfectant, UV lamps maintenance, and photoreactivation of enteric bacteria. The effectiveness is decreased in the effluents by substances like phenolic compounds, humic substances, lignin sulfonates, and ferric iron. The presence of suspended matter may protect microbes from UV light, leading to the implementation of pretreatment techniques (filtration) in water treatment.

UV light damages microbial RNA or DNA at a wavelength of 260 nm. This causes thymine dimerization, which blocks the replication of nucleic acid and effectively inactivates microbes. In the case of viruses, UV light attacks the genome, followed by a virus protein coat. Figure 6.3 shows the UV treatment of drinking water. A minimum dose (16,000 μW s/cm^2) is recommended for drinking water. This leads to a 99.9% reduction of bacterial coliforms (DeMers and Renner 1992). The factors that affect the performance are biological films, reactor geometry, short circuiting, microorganism clumping, and turbidity (Sawyer 1992). The continuous exposure to UV radiation does not change the water chemistry. As a result, there is no formation of THM or other disinfection by-products.

Fig. 6.3 UV treatment for drinking water (https://www.gigahertz-optik.de/en-us/basics-light-measurement/apps/uv-disinfection-lamp-control/)

3.2 Solar Disinfection

The use of sunlight is an ancient practice for many centuries. In developing countries, it is considered as a low-cost, simple technique for safe drinking water. The water is to be filled in a transparent glass or plastic bottle and is exposed to sunlight for a few hours. It is reported that many microbes are eliminated by solar disinfection. There are two pathways in solar disinfection to inactivate microbes. Figure 6.4 shows the direct and indirect effects of sunlight exposure.

The efficacy of this process depends on several factors. The factors that affect the performance are organic compounds (humic acids), inorganic salt, dissolved oxygen, light intensity, temperature, and type of container (Davies and Evison 1991). In literature, *Entamoeba histolytica* and cysts of *Giardia* spp. are inactivated by sunlight exposure (e.g., within 10 min at 56 °C) (Ciochetti and Metcalf 1984). The temperature is achieved with a solar hot box cooker. Thus, solar disinfection is not applicable all over the world, but it may be appropriate for sunny places in which

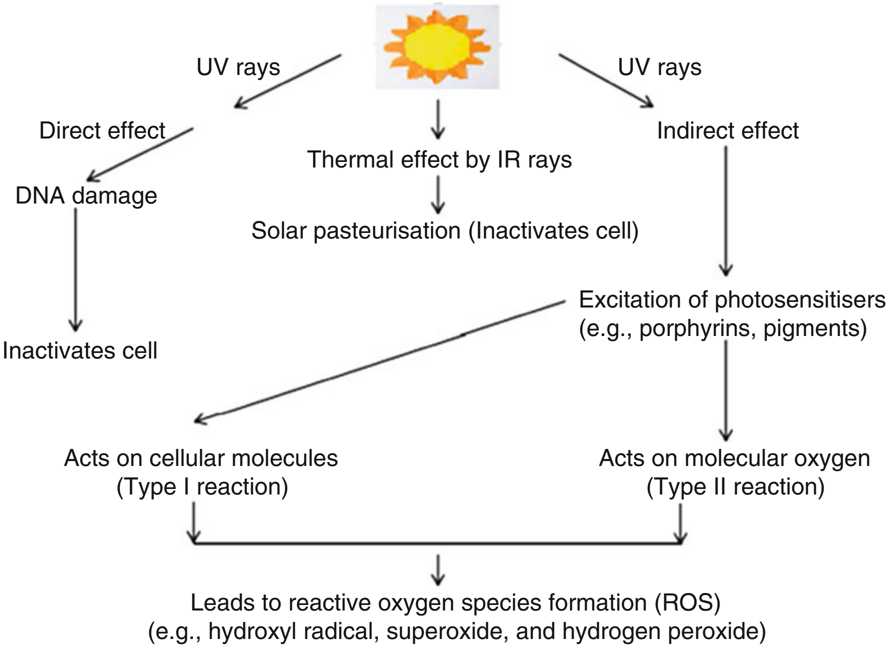

Fig. 6.4 Water disinfection by solar radiation

there is no realistic alternative treatment process. Boiling is another way of inactivating microbes. To achieve this, water is to be maintained in a boiling state for 5–10 min.

3.3 Filtration Methods

3.3.1 Membrane-Based Techniques

Membrane-based techniques are used to remove suspended solids, microbes, and ions from water. The principle behind these filters is physical separation. The separation is dependent on the size of the pores in the membrane. Substances that are larger than the size of the pores are completely removed, whereas the smaller size substances are partially removed, depending on the structure of a refuse layer. Figure 6.5 shows membrane-based techniques for water purification. Microfiltration (01.–2 bar) and ultrafiltration (2–10 bar) filters are low pressure-dependent processes that can remove microbes and suspended solids. The other substances (monovalent species and small colloids) in water can be effectively removed by nanofiltration (8–10 bar) and reverse osmosis (10–80 bar).

Osmosis is the flow of small size organic molecules through a semipermeable membrane from one side to another due to the difference in concentration across the

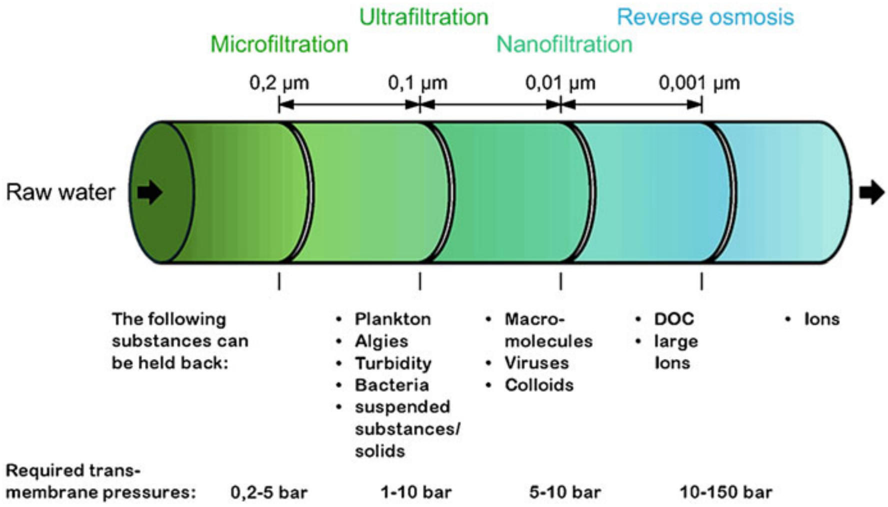

Fig. 6.5 Membrane-based water treatment (https://hbsciu.com/2015/10/12/modifying-water-purification-membranes-with-nanomaterials/)

membrane. At equilibrium, the concentration is balanced on both sides by the osmotic pressure. If the applied pressure is greater than the osmotic pressure, the solvent will flow to the side of the solution through the membrane (Fig. 6.6). This phenomenon is called reverse osmosis (RO) (Kim et al. 2005). Nanofiltration is effective for the removal of Mg^{2+} and Ca^{2+} ions and small size organic compounds. This method could be used to remove disinfection by-products (trihalomethanes) formed during the process of chlorination. RO is known for desalting brackish water and seawater. This process can be used to remove low-molecular-weight organic compounds from water.

The main issues in membrane-based technique are fouling and scaling issues. Fouling issues are due to organic or inorganic colloids and biological contaminants, while scaling is due to the presence of high levels of salt (calcium sulfate fluoride or carbonate or magnesium hydroxide) (Cheryan 1998; Baker 2004).

3.3.2 Sand Filtration

Slow sand filtration (SSF) is a simple process that allows water to pass through the bed of sand, and it can remove solids, microbes, and heavy metals in water. SSF is filled with a combination of coarse and fine sand as a filter medium, as shown in Fig. 6.7. To support this medium, pebbles and gravels are used. It consists of a layer of sand with a height of 60–120 cm and a gravel layer of 30–50 cm. The combination of coarse and fine sand helps in the removal of organic matter in water. Moreover, the flow rate is low in the case of SSF and maturation period (up to 40 days). The water is fed into the column from the top, and treated water is to be collected at the

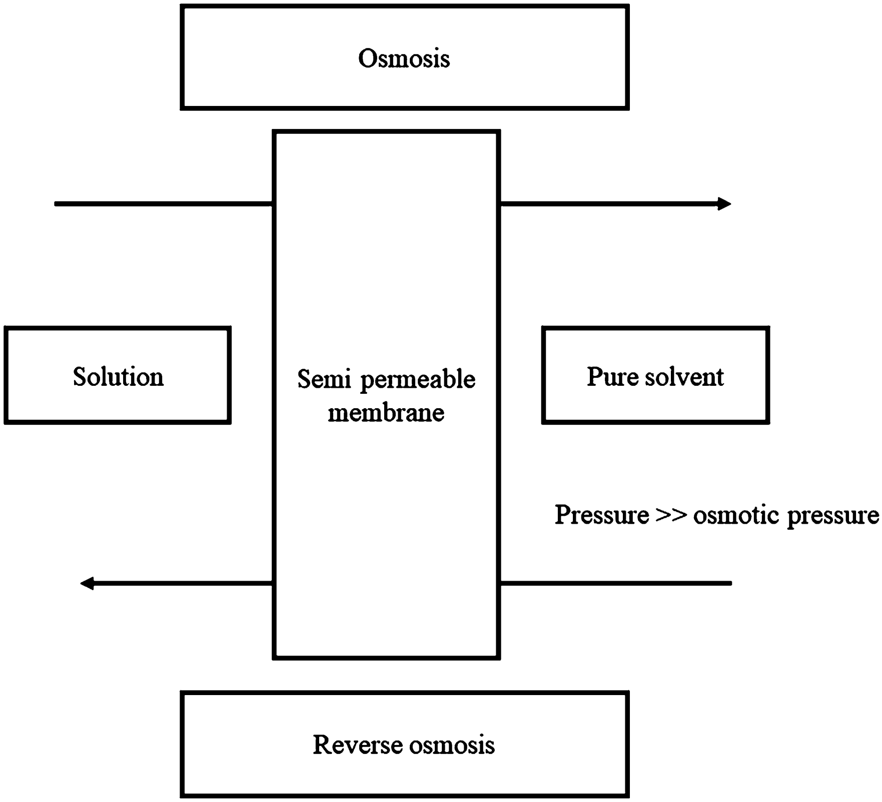

Fig. 6.6 Water disinfection by reverse osmosis

bottom of SSF. In the SSF process, biological filtration takes place in parallel with the physical filtration. Further, bacterial colonies form a slimy layer (schmutzdecke) during the operation of SSF. This layer hosts bacteria, protozoans, diatoms, and metazoan. The biological activity is enhanced by the accumulation of inorganic and organic debris on the surface of the layer. It helps in the removal of NOM, transforms synthetic organic compounds, retains microbes, and purifies water without any biological contaminants. The formation of this biolayer (schmutzdecke) increases head loss, thereby increasing SSF performance during the start-up phase. The quality of water remains poor in the initial period of operation, and it can be improved after the ripening process. The main operational problem in SSF is clogging. It is caused by the suspended solids and biofilm formation on the surface of media. During the process of clogging, head loss increases in the filter, and beyond a point the filter is aborted. To overcome this issue, the removal of the top layer of the sand particle is the most preferred method. In remote areas, SSF performance can be improved by reducing the loading rate, pretreatment of water, and increasing the frequency of dosage. However, SSF demands large areas per unit volume of water to be treated,

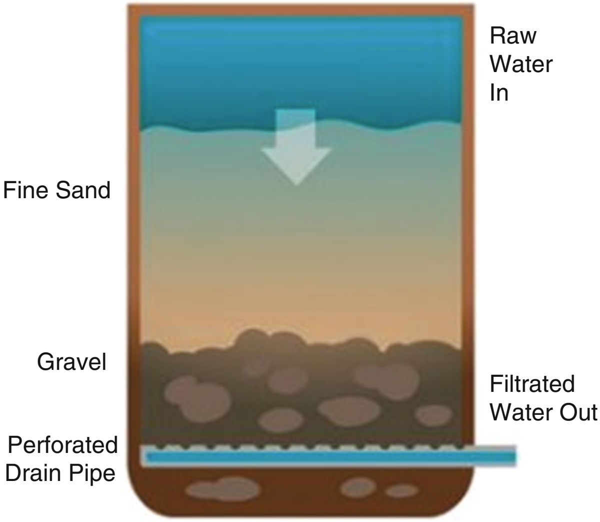

Fig. 6.7 Slow sand filter (https://www.fertinnowa.com/technologies/slow-sand-filtration-removal-algae-diseases-horticulture/)

which in turn suits for the rural community. SSF is known for the removal of turbidity, waterborne pathogens, and suspended solids. The factors that affect the performance of the filter are temperature, sand size, filter depth, and flow rate. Particularly, some of the microbes are small enough to pass through filters as individual particles. Further, the turbidity in water can affect the removal efficiencies of microbes due to the presence of particulate matter, which can block the filters and results in bypasses. Therefore, the combination of pretreatment, sand filtration, and chemical disinfection methods are possible ways to reduce microbial contaminants in drinking water (Alvarez et al. 2008; Hoslett et al. 2018).

3.3.3 Rapid Sand Filtration

RSF is an important physical process in drinking water treatment plants that involves the removal of large suspended solids. It requires prior and post-treatment stages. Typically, RSF is made of a sand bed with a height ranging from 1.5 to 2.5 m, and it is operated in the downward direction at a filtration velocity of 3–8 m/h (Fig. 6.8). The filter medium (usually sand) size ranges from 0.4 to 1.5 mm. These filters perform well when the turbidity of the incoming water should be below 20 NTU. RSF involves solid-liquid separation with an objective to reduce turbidity (0.3–1.0 NTU). Recently, the removal of *Giardia* cysts and *Cryptosporidium* oocysts with a turbidity of 0.1–0.3 NTU. It requires preparatory treatment (coagulation and flocculation) to have a higher throughput of water. The hydraulic loading rate depends on the incoming water quality, temperature, and the media that we used in the filter. Typically, the filter with sand exhibits around 6 $m^3/m^2/h$. It is reported that a clean filter will possess a head loss of around 0.3 m. Once the filter reaches the head loss of

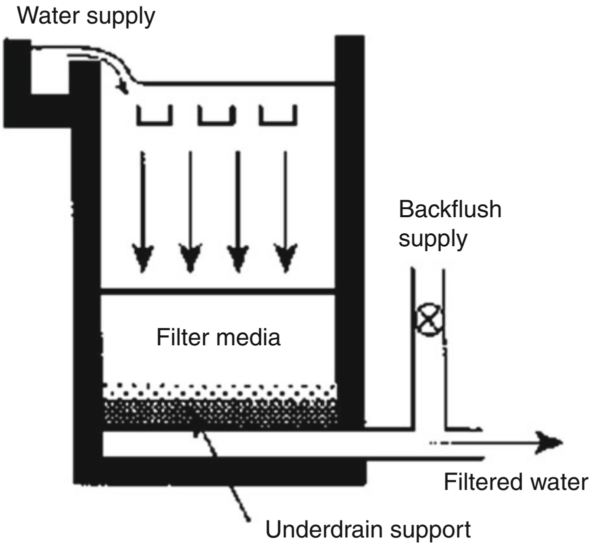

Fig. 6.8 Rapid sand filtration (http://www.ce.memphis.edu/1101/notes/filtration/filtration-1.html)

around 1.5–2 m, the filter needs to be cleaned by backwashing. A critical stage is reached when the frictional resistance increase in the top layer of the sand bed exceeds the static head of water above the bed of sand. The bottom sand layer behaves like a vacuum, and water just passed through the media rather than getting filtered through them. As a result, the developed negative pressure tends to release the gases present in water. The gas bubbles prefers to stick with the surface of the sand. This phenomenon is called as air binding. Hence, the filter needs to be cleaned as soon as it reaches the optimum value. Interestingly, mud accumulates on the surface of sand particles. Over a period of time, mud becomes like a ball when there is an inadequate washing. Once mud balls are formed in a filter, it is difficult to clear. The fine sand particle in the top layer of the bed shrinks and causes cracks in the bed. The crack becomes large with the increase in loss of head.

In RSF, backwashing is to be done periodically to avoid clogging. During the backwash, water is fed into the filter in the upward direction at a velocity to expand the filter media. The mechanism of cleaning is based on the hydraulic shear forces on the media. After backwashing, larger particles tend to settle at the bottom of the filter, and fine particles will settle at the top of the filter. As a result, large particle creates void spaces at the bottom, and clogging will start to occur at the top of the filter, which leads to incomplete use of bed that is restricted. The ripening period of RSF is 1–4 days which is very low as compared to SSF. This process requires preparatory treatment (coagulation and flocculation) to have a higher throughput of water (Han et al. 2009; Management SS and W 2012).

3.3.4 Granular Activated Carbon

Activated carbon used in water purification is produced from different materials such as coconut shells, wood, and coal. Particularly, it has high potential in water treatment due to its characteristics such as porous texture, high surface area functional groups, etc. Granular activated carbon (GAC) is used in both filtration and post-filtration methods to capture organic and odor compound. Generally, GAC is replacing RSF in most of the places, thereby reducing the necessity of further filtration. The filters that are based on GAC can operate at a high flow rate than SSF. Therefore, it is predominantly used in water treatment plants where the space is limited. Furthermore, the life cycle of GAC depends on how long the bed gets saturated with the targeted pollutants. As far as the biological activity is concerned, it can generate issues like clogging, dead zones, and detachment of microbes from the surface of GAC.

Nowadays, silver impregnation is being done to the surface of carbon to remove chemicals and inactivate microbes. Further, activated carbon in combination with a chemical disinfectant is also employed to remove both chemical and biological contaminants. To obtain maximum efficiency, it is desirable to have a maximum specific surface area in the smallest volume. Generally, GAC exhibits a specific surface area ranging from 300 to 1500 m^2/g and an abundant quantity of polar functional groups. As a result of functional groups, microbes are trapped on the carbon surface. It is reported that GAC removes microbes (*E.coli*, MS2, and spores of *Cryptosporidium parvum* and *Giardia lamblia*) by the process of adsorption. The centralized system involves various treatment stages such as sedimentation, coagulation/flocculation, filtration, and disinfection. On the other hand, the decentralized system consists of MF/UF and GAC filters.

4 Future Perspectives on Water Disinfection

The quest for safe water has been the main priority across the globe. The necessity increases for humans due to population growth, natural resources depletion, and ill-managed treatment methods for water, especially the lack of disinfection and treatment methods, specifically in small villages. We need to create awareness about the existing systems and motivate people to have clean and safe drinking water. This chapter describes a variety of disinfection techniques for safe water, such as chemical and physical methods, including traditional systems such as chlorination and sand filtration. Each technique has merits and demerits for having safe water. Therefore, the hybrid process can be accepted as a solution to have safe water. The factors that decide the economics of the process are the physical, chemical, and biological quality of water and environmental factors like pH, temperature, etc.

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