



Application of Membrane Filtration to Cold Sterilization of Drinks and Establishment of Aseptic Workshop

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Abstract

Aseptic packaging of high quality beverage is necessary and its cold-pasteurization or sterilization is vital. Studies on application of ultrafiltration or microfiltration membrane to cold- pasteurization or sterilization for the aseptic packaging of beverages have been reviewed. Designing and manufacturing ultrafiltration or microfiltration membrane systems for cold-pasteurization or sterilization of beverage are based on the understanding of size of microorganisms and theoretical achievement of filtration. It is concluded that adaptability of membrane filtration, especially its combination with other safe cold method, to cold- pasteurization and sterilization for the aseptic packaging of beverages should be assured without a shadow of doubt in future.

Keywords Beverages · Membrane · Microorganism removal · Theory and practice

Introduction

Inadequate water intake can cause dehydration which subsequently results in poor health or diseases (Perrier et al., 2013). The European Food Safety Authority Dietary Reference Values for the Adequate Intake of water was recommended to be 2.5 and 2.0 L/day, respectively for men and women from 14 years of age onward, and 2.1 and 1.9 L/day, respectively for boys and girls from 9 to 13 years of age (EFSA, 2010) while the Institute of Medicine of USA recommended higher water intake (3.7 and 2.7 L/day for males and females respectively) (Institute of Medicine, 2005).

Solid foods intake can only contribute about 20% water so that 80% water should be provided by drinks (Popkin et al., 2010) including all kinds of liquids which are drinkable (Nissensohn et al., 2016). This promotes drinks production. About 1.6 trillion L (231 L/person/year) drinks were produced in 2009 (top 8 including tea (20.9%), bottled water

(15.3%), milk (12.8%), carbonated soft drinks (12.5%), beer (11.2%), coffee (8.2%), still drinks (2.7%) and juices/nectars (2.6%) according to market share) (Neves et al., 2011). Although most drinks just mentioned above excepting bottled pure water also provide some other nutrients, the overwhelming majority of their component is water.

Large quantity of water may cause the growth of microorganisms which spoil drinks quickly. Therefore, sterilization and proper packaging are essential for storing drinks with extended periods (or a targeted shelf life), which is a prerequisite to their marketability. To achieve this goal, sterilization (complete elimination of microorganism including spores) and aseptic packaging must be assured, which must be economical, relatively simple and capable of keeping nutritional or beneficial components and sensory properties as constant as possible.

Sterilization by high temperature is traditionally used for producing drinks, e.g. milk, juices/nectars, teas, coffee and alcoholic drinks (Fellows, 2017; Pearce et al., 2012). Ultra-high temperature (UHT; heated at 130–150 °C for 1–2 s and then flash cooled to 70 °C) (Fellows, 2017) and high temperature at 120 °C with or without high pressure have usually be used. However, this technique has disadvantages, e.g. losses of B vitamins, vitamin C and other heat labile functional compounds (e.g. proteins) as well as adverse changes of sensory attributes, e.g. color, volatile

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aroma compounds and taste (Myer et al., 2016) in addition to high energy consumption.

Therefore, cold sterilization of drinks has attracted extensive studies (Koutchma, 2009; Reineke et al., 2015). Physical [including ultraviolet irradiation, pascalization (high pressure), high intensity pulsed electric field, ultrasound, etc.] and chemical methods have been developed as cold sterilization methods. Although the physical methods can efficiently kill the vegetative cells of microbes, none of them alone is able to completely inactivate spores (Cho & Chung, 2020; Lopes et al., 2018; Lv et al., 2021; Modugno et al., 2020). The spores of bacillus species are not sterilized by hydrostatic high pressure (at 400 MPa) at room temperature (Takeo et al., 1994) so that its combination with other methods is proposed. Complete inactivation of spores by high intensity pulsed electric field is difficult to be achieved (Reineke et al., 2015). Chemical methods may be efficient to completely inactivate spores, but they are not safe for consumption and environment protection. Ultrasound alone is difficult to meet a 5-log reduction of microbes required by FDA (Rupasinghe & Yu, 2012). Ultraviolet irradiation may be efficient for sterilizing transparent drinks, but it has weakness, e.g. poor efficiency for cloudy drinks, cause of losses of nutrients, e.g. vitamins and quite harmful when it directly irradiates human body (Koutchma, 2009). Gamma or X-ray irradiation is usually not recommended for using in food industry because of safety concern.

Ultrafiltration membrane has a pore size ranging from 0.001 to < 0.1 μm (McCarron et al., 2016) while microfiltration membrane has a pore size ranging from 0.1 to 10.0 μm (Ismail and Goh, 2014). An ultrafiltration or microfiltration membrane can reject particulates larger than its pore size with avoidance of causing adverse chemical or physical changes. Any pathogenic microbe has a certain size so that it can be removed by a membrane with a proper pore size. This means that ultrafiltration or microfiltration membranes with proper pore sizes have the potential of cold-sterilizing drinks.

The aim of this paper is to review the application of membrane filtration to cold sterilization of drinks. This should provide the systematic knowledge about scientific basis and practical technology.

Scientific Basis of Membrane Filtration to Remove Microorganisms

For production of bottled water and other drinks, use of water is inevitable. It must be assured that pathogenic microorganisms are eliminated before proper packaging or consumption. Waterborne and airborne microorganisms are usually involved.

In theory, the ability of removing microorganisms from drinks by a membrane depends on their sizes. For sphere-like shaped microorganisms, their removability by a membrane usually depends on their diameter. For rod-like microorganisms, their removability by a membrane is normally determined on their wide (the size being normally < that of length). Therefore, this section reviews the size of common water- or air-borne microorganisms.

Size of Waterborne Microorganisms and their Removability

Three types of pathogenic microorganisms have been identified to commonly exist in water. They are bacteria, viruses and parasites with their sizes being measured.

Size of Waterborne Bacteria and their Removability by Membrane

The size of bacteria is between that of parasites and viruses. Both bacillus and non- bacillus pathogenic bacteria can be found in water.

Table 1 indicates that the size of bacillus bacteria ranges from 0.2 to 6 μm long by 0.2 to 1 μm wide depending upon different varieties. In addition to pathogenic microorganisms, those which are usually used for preserving or producing some kinds of drinks as probiotics or beneficial bacteria (lactic acid producing bacteria, e.g. *Lactobacillus* having a cell size of $\geq 0.3 \mu\text{m}$ in diameter or wide—the bacteria which produce lactic acids from sugars or acetic acid producing bacteria, e.g. *Acetobacteraceae* having a cell size of 0.6–0.8 μm by 1.0–4.0 μm —the bacteria which produce acetic acids from ethanol). This means that the ability of this kind of bacteria to penetrate a membrane with a certain pore size should be determined on their wide or diameter rather than their length. It appears to be that waterborne bacillus bacteria with size (width) less than 0.2 μm have not been reported.

Non-bacillus bacteria usually have a sphere-like shape, which are measured by diameter. Table 1 also shows that the size of the smallest water-borne non-bacillus bacteria is 0.5 μm in diameter.

In practice, it should be noted that bacteria may be deformed by membrane so that they can transfer through the membrane with a pore size < their size (Gaveau et al., 2017). Furthermore, soft pathogens were reported to penetrate the membrane with a pore size < their size (Helling et al., 2017). This conclusion is supported by the report that a membrane with 0.1 μm pore size was suggested to assure the efficient removal of *Burkholderia pseudomallei* (*Ralstonia pickettii*) though the penetration of this kind of

Table 1 Size of some waterborne bacteria, viruses and parasites

Species	Size
Bacteria	
<i>Burkholderia pseudomallei</i>	2–5 µm long and 0.4–0.8 µm in wide (Dillon, 2014)
<i>Campylobacter jejuni</i> , <i>Campylobacter coli</i>	0.2–0.5 µm long and 0.2–0.9 µm wide (Pielsticker et al., 2012)
<i>Escherichia coli</i>	1–2 µm long or longer by 0.25–1 µm wide (Gu et al., 2016)
<i>Legionella</i> spp.	0.3–0.9 µm wide by 2–20 µm long (Żbikowska et al., 2014)
Non-tuberculous mycobacteria	0.5–5 µm in diameter (Bédard et al., 2016)
<i>Pseudomonas aeruginosa</i>	0.5–1.0 µm wide by 1.5–5.0 µm long (Bédard et al., 2016)
<i>Salmonella enterica</i> (serovar Typhimurium)	2–5 µm long by 0.5–1.5 µm wide (Andino & Hanning, 2015)
<i>Shigella</i> spp.	0.3–1 µm wide by 1–6 µm long (PHE, 2015)
<i>Vibrio cholerae</i>	1–3 µm long by 0.5–0.8 µm wide (Nazar-ul-Islam & Malik, 2015)
<i>Yersinia enterocolitica</i>	0.5–0.8 µm wide by 1–3 µm long (Van Damme, 2013)
Viruses	
<i>Adenoviruses</i>	ca. 0.063 µm in diameter (Bondoc & Fitzpatrick, 1998)
<i>Enteroviruses</i>	0.025–0.30 µm in diameter (Yates, 2014)
<i>Hepatitis A</i>	0.027–0.032 µm in diameter (Konduru et al., 2009)
<i>Hepatitis E</i>	0.027–0.034 µm in diameter (Kamar et al., 2014)
<i>Noroviruses</i>	0.027 µm in diameter (Griffith, 2018)
<i>Sapoviruses</i>	ca. 0.030–0.038 µm in diameter (Robilotti et al., 2015)
<i>Rotavirus</i>	ca. 0.07 µm in diameter (Oka et al., 2015)
Parasites	
Protozoa	
<i>Acanthamoeba</i> spp.	25–40 µm (Marciano-Cabral & Cabral, 2003)
<i>Cryptosporidium parvum</i>	0.4–0.6 µm (sporozoite) to 6 µm in diameter (mezoite) (Leitcha & He, 2011)
<i>Cyclospora cayetanensis</i>	8–10 µm (Naito et al., 2009)
<i>Entamoeba histolytica</i>	10–20 µm (Aguilar-Díaz et al., 2010)
<i>Giardia intestinalis</i>	8–20 µm long by 5–16 µm wide (Zeibig, 2013)
<i>Naegleria fowleri</i>	7–35 µm (Gutierrez, 2000; Claydon, 2017)
<i>Toxoplasma gondii</i>	6–12 µm (Dumètre et al., 2013)
Helminths	
<i>Dracunculus medinensis</i>	1.4–2.7 cm long by 295–350 µm in wide (Eberhard et al., 2016)
<i>Schistosoma</i> spp.	ca. 254.9–500 µm long by 64 µm wide (cercariae) (Braun et al., 2018) or 70–175 µm long by 40–70 µm wide (egg) (Jing et al., 2018)

bacteria to a membrane with 0.2 (or 0.22) µm pore size was not observed by others (Sundaram et al., 1999). In conclusion, it can be predicted that a microfiltration membrane with its size at 0.1 µm or an ultrafiltration membrane may be useful for removing all kinds of water-borne bacteria.

Size of Waterborne Viruses and their Removability by Membrane

Viruses have the smallest size among three types of waterborne pathogenic microorganisms. Table 1 shows that the size of waterborne viruses including noroviruses ranges from 0.025 to 0.07 µm in diameter. However, virus size as small as 0.02 µm in diameter (Yin, 2015) and even 0.01 µm in diameter (Hai et al., 2014) were also reported.

Ultrafiltration membrane with its pore size > 0.02 µm may not be able to remove the smallest viruses. Notwithstanding, ultrafiltration membrane with its pore size < 0.01 µm should be theoretically efficient for removing all kinds of viruses; the smaller the pore size of membrane, the higher the reliability of sterilization. Furthermore, a study on purification (or cold sterilization) of water indicated that the formation of a fouling (or named cake) layer may facilitate the removal of viruses by the membrane with a larger pore size (Chaudhry et al., 2015). On the other hand, too small size of ultrafiltration membrane can result in problem of serious fouling and low permeability of water or nutrient with a small molecular weight so that production efficiency is reduced. A study on cold sterilization of beer showed that precipitation of large particles, chemical interaction between solutes and membrane, absorption of small particles or solutes by inside wall

of membrane pores, etc. may be associated with the mechanism of membrane fouling which may cause the following consequences: complete pore blocking, partial pore blocking, cake formation and internal pore blocking (Esmaeili et al., 2015). Therefore, selection of proper pore size of ultrafiltration membrane should always be necessary for obtaining a good balance of complete removal of viruses and reasonable production efficiency. It is reasonably believed that microfiltration membrane (pore size $> 0.1 \mu\text{m}$) may not be able to efficiently remove this kind of microorganisms. However, microfiltration may be used for the pretreatment of water to remove large microorganisms (including some viruses) before ultrafiltration so that fouling problem of membrane can be efficiently reduced. This kind (microfiltration/ultrafiltration) of filtration system found to be effective for the production of drinking water or other beverages (Ahmed et al., 2017). This kind of system may still need a pretreatment to tackle fouling problem depending upon varieties of beverages treated or the extend of contamination. For example, the use of coagulation reagents was reported to be effective for controlling the formation of fouling layer (Erdei et al., 2008; Jang et al., 2010).

Size of Waterborne Parasites and their Removability by Membrane

Parasites have the largest size among three types of waterborne pathogenic microorganisms. Table 1 indicates that the size of parasites ranges from 0.4 to 350 μm . *Cryptosporidium parvum* appears to be the smallest waterborne parasite, which has a size of 0.4 μm in the stage of sporozoite.

Therefore, in theory, a microfiltration membrane with its pore size $< 0.4 \mu\text{m}$ should be able to remove all kinds of water-borne parasites; the smaller the pore size of membrane, the higher the reliability of pasteurization or sterilization. Ultrafiltration membrane should theoretically be able to efficiently remove all kinds of water-borne parasites.

Size of Waterborne Fungi and their Removability by Membrane

The size of fungi found in water which may also contaminate beverages is quite large. Yeasts (e.g. *Saccharomyces cerevisiae*, a kind of fungi—producing ethanol from sugars; see Table 2 for its size) may also need to be removed for producing some packaged drinks, e.g. clear fruit juices and for preventing their spoilage (e.g. acidification of alcoholic drinks or fermentation of fruit juices to result in ethanol). The size of spores or conidia of most fungi or that of cells of some fungi is $\geq 1 \mu\text{m}$ in diameter or wide (Table 2). The spores

Table 2 Size of some fungi as potential microbe-contaminants in water or air

Genus	Size
<i>Absidia</i>	2.0–3.0 $\mu\text{m} \times 4.0$ –5.0 μm (cylindrical sporangiospore) (Nguyen et al., 2016)
<i>Alternaria</i>	2.38–13.09 $\mu\text{m} \times 12.30$ –43.63 μm (spore size) (Abbo et al., 2018)
<i>Aspergillus</i>	$\geq 5 \mu\text{m}$ in diameter (spore), or $\geq 2.0 \mu\text{m}$ in diameter (conidia) (Kwon-Chung & Sugui, 2013)
<i>Blastomyces</i>	$\geq 1.5 \mu\text{m}$ in diameter (ascospore) (Lippman, 2001)
<i>Candida</i>	$\geq 1.5 \mu\text{m}$ in diameter (blastospore) (Whitley-Williams, 2006)
<i>Cladosporium</i>	$\geq 1.5 \mu\text{m}$ in diameter or wide (conidia) (Bensch et al., 2012)
<i>Cryptococcus</i>	1–2 μm in diameter (spore) (Wang & Lin, 2012)
<i>Geotrichum</i>	$\geq 3 \mu\text{m}$ in diameter or wide (arthrospore) (Ochoa et al., 2015) or $\geq 2 \mu\text{m}$ in diameter or wide (chlamidospore) (San-Martin et al., 2008)
<i>Hansenula (Pichia)</i>	$> 1 \mu\text{m}$ in diameter or wide (ascospore) (Kurtzman, 1987)
<i>Histoplasma</i>	$> 1 \mu\text{m}$ in diameter (hypha) or $\geq 2 \mu\text{m}$ in diameter (microconidia) (Nosanchuk, 2016)
<i>Microsporium</i>	$> 1.5 \mu\text{m}$ in diameter or wide (ascospore) (Dexter, 2003)
<i>Mucor</i>	$\geq 2 \mu\text{m}$ in diameter or wide (sporangiospore) (Álvarez et al., 2011)
<i>Penicillium</i>	$\geq 1.8 \mu\text{m}$ in diameter or wide (conidia) (Houbraken et al., 2011)
<i>Rhizopus</i>	$\geq 2.7 \mu\text{m}$ in diameter or wide (sporangiospore) (Hartanti et al., 2013)
<i>Rhodotorula</i>	$\geq 2 \mu\text{m}$ in diameter or wide (cell) (Chang & Wang, 2002)
<i>Saccharomyces</i>	$\geq 1 \mu\text{m}$ at the small diameter (cell) (Waites et al., 2013)
<i>Scopulariopsis</i>	$\geq 2 \mu\text{m}$ in diameter or wide (ascospore I) (Sandoval-Denis et al., 2013)
<i>Sporotrichum</i>	$\geq 1.4 \mu\text{m}$ in diameter or wide (blastoconidia) (Stalpers, 1984)
<i>Trichophyton</i>	$\geq 2 \mu\text{m}$ in diameter or wide (microconidia) (Armon & Cheruti, 2012)
<i>Trichosporon</i>	$\geq 2 \mu\text{m}$ in diameter or wide (arthroconidia) (Taj-Aldeen et al., 2009)

or conidia, etc. may have cylindrical or elliptical shape, etc. so that their ability of penetrating a membrane with a fixed pore size normally determined on their diameter or wide rather than their length. Microfiltration membrane with its size $< 1 \mu\text{m}$ should be able to remove spores or conidia, etc. of most fungi or cells of some fungi as well as whole part of them. Ultrafiltration membrane should theoretically be able to efficiently remove all kinds of water-borne fungi.

In conclusion, the size of water-borne microorganisms ranges from 0.01 to 350 μm . Ultrafiltration membrane at a proper pore size (e.g. $< 0.01 \mu\text{m}$) should theoretically be able to remove all kinds of water-borne microorganisms whereas a microfiltration membrane with its size at 0.1 μm is suitable for removing bacteria with their size $> 0.1 \mu\text{m}$.

It is suggested that pathogenic microorganism having the smallest size in water be determined before selection of a proper membrane for cold pasteurization or sterilization of water. Countermeasures for contaminations by "unexpected" pathogens may also be important for avoiding health risks in consumers (Amalfitano et al., 2018). This aspect has also been highlighted by other studies (Orhan et al., 2021; Beck et al., 2021). These can assure that the pasteurized or sterilized water can be directly used for producing bottled water (or for directly drinking) and preparation of other beverages.

Size of Airborne Microorganisms and their Removability

Table 2 indicates the size of airborne fungi. In addition, the size of many other airborne bacteria not indicated in the table is as follows: for example, size in term of diameter or wide, *Staphylococcus* ($\geq 0.5 \mu\text{m}$), *Streptococcus* ($\geq 0.4 \mu\text{m}$), *Haemophilus* ($\geq 0.35 \mu\text{m}$), *Pasteurella* ($\geq 0.2 \mu\text{m}$), *Bordetella* ($\geq 0.2 \mu\text{m}$), *Francisella* ($\geq 0.2 \mu\text{m}$), *Corynebacterium* ($\geq 0.25 \mu\text{m}$), *Borellia* ($\geq 0.2 \mu\text{m}$), *Treponema* ($\geq 0.09 \mu\text{m}$), *Neisseria* ($\geq 0.4 \mu\text{m}$), *Chlamydia* ($\geq 0.2 \mu\text{m}$), *Rickettsia* ($\geq 0.1 \mu\text{m}$), *Bacillus* ($\geq 0.1 \mu\text{m}$), *Clostridium* ($\geq 0.3 \mu\text{m}$), *Morganella* ($\geq 0.6 \mu\text{m}$), *Proteus* ($\geq 0.3 \mu\text{m}$), *Klebsiella* ($\geq 0.3 \mu\text{m}$), *Citrobacter*, *Coxiella* ($\geq 0.2 \mu\text{m}$).

The size of many airborne viruses has also been known, for example, *Parvovirus B19*, *Rhinovirus*, *Coxsackievirus* (*Coxsackievirus* 16 having a diameter of ca. 0.030 μm), *Echovirus* (0.024–0.030 μm), *Hantavirus* (0.070 to 0.350 μm), *Togavirus* (0.040 to 0.080 μm), *Reovirus* (0.050 to 0.080 μm), *Adenovirus* (0.070 to 0.100 μm), *Orthomyxovirus* (0.080 to 0.120 μm), *Coronavirus* (*Coronavirus* -19 having a size of 0.070 to 0.090 μm), *Arenavirus* (0.050 to 0.300 μm), *Morbillivirus* (e.g. Measles virus belonging to *Paramyxovirus* of the genus *Morbillivirus* having a size of 0.120 to $> 0.300 \mu\text{m}$), *Respiratory Syncytial Virus* (0.150 to 0.250 μm) and *Poxvirus—Vaccinia* (0.240 to 0.300 μm). Among these viruses, *Parvovirus B19* and *Rhinovirus* have the smallest size, i.e. 0.018 μm while *Paramyxovirus* has the

biggest size, i.e. 0.31 μm (Seregin et al., 2020; Kim et al., 2020; Flowers et al., 2016; Gong and Mita, 2014; Hepojoki et al., 2012; Utley et al., 2008; Rager et al., 2002; Doerfler, 1996; Couch, 1996; Baxby, 1996).

Airborne microorganisms normally adhere to the particle matters in air (or named airborne dust). The adherence of microorganisms including bacteria, fungi and viruses to airborne dust can form bioaerosols having 0.5 to 50 μm size (Lee, 2011), which is much larger than that of the original bacteria, fungi and viruses.

The airborne microorganisms may also contaminate drinks during their processing, filling and packaging. Therefore, removal of airborne microorganisms is very important for producing high quality drinks.

For the avoidance of contamination by airborne microorganisms during the production of drinks, two kinds of approaches may be effective. One way is to remove them from contaminated drinks. Another way is to remove them from air to prevent drinks from contamination, i.e. establishing an aseptic workshop.

According to the particle size of airborne microorganisms, they can even be removed by microfiltration membrane except for *Treponema*, *Rickettsia* and *Bacillus*. Ultrafiltration membrane at a proper pore size (e.g. $< 0.01 \mu\text{m}$) should theoretically be able to remove all kinds of airborne microorganisms, which should be suitable for cold sterilizing drinks which are contaminated by airborne microorganisms.

It is suggested that pathogenic airborne microorganism having the smallest size be determined before selection of a proper membrane for cold pasteurizing or sterilizing a beverage as the end filtration of aseptic packaging. As the basis of selecting a proper membrane, the size of the largest nutritional or functional particle in beverage should also be determined.

Summary of Relationship Between Nominal Pore Sizes of Membrane and Rejection of Microorganisms

It should be noted that the nominal pore size of membrane provided by membrane manufacturers may be generally the size of the maximum pore of the membrane. Normally, a membrane with pore size distribution is manufactured and the rejection size of microorganism is determined on the maximum size of the membrane.

At the initial stage of filtration, microorganisms with larger sizes may transmit the membrane with a smaller pore size due to deformation effects. This conclusion is supported by the report which described that an ultrafiltration membrane with a nominal pore size of 1 kDa was unable to completely remove norovirus from water (Matsushita et al., 2013).

On the other hand, cake formation may result in the efficient removal of some microorganisms with their particle size smaller than the pore size of membrane though this process can reduce permeate flux. This conclusion is supported

by the report which indicated that cake layer changed the cutt-off size of membrane (Zheng & Liu, 2007).

Application of Membrane Filtration to Cold Sterilization of Drinks

Ultrafiltration Membrane for Sterilizing Transparent Drinks

Sterilization of Natural Water by Ultrafiltration Membrane Water from a natural source may contain certain amount of waterborne pathogenic microorganisms and airborne microorganisms derived from contamination. To assure that water is directly drinkable or applicable to drink or other sectors of food industry, its sterilization is inevitable.

Nutrients in drinking water are minerals with molecular weights which are much smaller than the pore size of ultrafiltration membranes. Therefore, cold sterilization of natural water by ultrafiltration with a proper pore size is able to remove all kinds of microorganisms (see “2.” described above) with avoidance of losing valuable nutrients.

Many studies on the cold sterilization of water by ultrafiltration can be found in literature. For example, a membrane made of polyvinylidene fluoride (PVDF)/ polyethersulfone (PES) blend was reported to be sufficient for cold sterilization of water (> 99.99% rejection of microbes) (Madaeni & Pourghorbani, 2013). The ultrafiltration membrane made of silica/polyvinyl alcohol was found to completely remove all microorganisms in water (Gan & Wu, 2020). Another study indicated that permeate of ultrafiltration membrane with 200 kDa separation contained virus particles (Bray et al., 2021), which may require the use of a membrane with smaller pore sizes for completely removing pathogens from water. A membrane with a pore size of 10,000 or 50,000 Da was reported to remove all microorganisms from apple juice and the quality attributes of the filtered juice resembled the untreated fresh juice except for slight browning of color (Ortega-Rivas et al., 1998).

A pilot study using hollow fiber (Multibore PES membrane) for producing drinkable water for a long time was carried out (Molelekwa et al., 2014). The pore size of ultrafiltration membrane was 0.01–0.1 μm , which reduced total coliform from > 2419.2 CFU/100 mL (unacceptably high) to ca. 7 CFU/100 mL (< acceptable level of 10 CFU/100 mL recommended by WHO) and completely removed *E. coli* and enterococci. This means that cold sterilization of water by ultrafiltration membrane on an industrial scale is very likely feasible.

The degree of fouling is dependent upon type of the material for making membrane and the extent of contamination; for example, if the membrane is made of PP, its permeability can decrease 68–97% after filtration of 150 L/ m^2 of water (Howe & Clark, 2002). The combination of coagulation/flocculation and ultrafiltration (using 0.1 μm ceramic membranes) for the treatment of drinking water (containing 39.55 CFU/mL total coliforms and 8.00 CFU/mL *Escherichia coli*) removed 99.0% of microorganisms with 368–626 L/ hm^2 average permeability as well as 14.07–23.55% R_m (intrinsic resistance of membrane), 5.18–75.65% R_f (fouling resistance of membrane) and 0.90–71.27% R_{cp} (concentration polarization resistance of membrane) depending upon variation of ΔP and coagulation reagents used (Bergamasco et al., 2011). On the other hand, proper fouling layer formed can facilitate the rejection of microorganisms, especially those with small sizes, such as virus. A membrane filtration system for cleaning water was reported to have a pore size capable of retaining most bacteria and incapable of rejecting virus, but achieve 3.3-log₁₀ reduction of viral pathogens because of formation of a fouling (or named cake) layer (Verbyla and Rousset, 2018). It was also reported that a 4- to 5- day fouling layer formed was able to result in a 1.6 log₁₀ increase in removal (5.5–7.1-log₁₀ reduction) of adenovirus (Chaudhry et al., 2015) whereas removing fouling layer immediately decreased the efficiency of removing virus (Lv et al., 2006). Therefore, proper balance between permeability and fouling

Table 3 Comparison of indicative log₁₀ removals of microorganisms from water between ultrafiltration and other cold sterilization methods (Collivignarelli et al., 2018)

Microorganisms	Ultrafiltration	Ultraviolet radiation	Ozonation	Chlorination
Viruses (Including Adenoviruses, Rotaviruses and Enteroviruses)	> 3- > 6	> 1.0 adenovirus; > 3.0 enterovirus, hepatitis A virus	3.0–6.0	1.0–3.0
Bacterial Pathogens (Including Campylobacter)	> 6.0	2.0–> 4.0	2.0–6.0	2.0–6.0
<i>E. coli</i>	5.5–> 6	2.0–> 4.0	2.0–6.0	2.0–6.0
<i>Giardia</i>	> 6.0	> 3.0	-	0.5–1.5
<i>Cryptosporidium</i>	> 6.0	> 3.0	-	0.0–0.5
<i>Clostridium</i>	> 6.0	-	0–0.5	1.0–2.0
<i>Perfringens</i>				

Log 2 and log 3 correspond to 99% and 99.9% removals of microorganisms, respectively

may be important for the efficient sterilization of water by membrane filtration.

Table 3 indicates that the efficiency of cold sterilization of ultrafiltration is better as compared with ultraviolet radiation, ozonation and chlorination. Ultrafiltration membranes with proper pore sizes can be efficient for cold-sterilizing water in drinking water industry and at home or public places. Also, it should be noted that the quality of raw water sources should significantly affect the efficiency of cold-sterilization and life-span of membranes used.

Sterilization of Clear Fruit Juice by Ultrafiltration Membrane All kinds of clear fruit juices should not contain any colloids with large particle sizes, such as proteins and polysaccharides. Otherwise, they are very likely to be turbid. Furthermore, the presence of proteins and polysaccharides such as pectin may lead to the formation of precipitates during storage. Therefore, these macromolecules should be removed so that clear fruit juices only contain the nutrients with small molecular weights. Ultrafiltration has been found to effectively remove such macromolecules as proteins and polysaccharides from fruit juices (Hounhouigan et al., 2014; Perreault et al., 2021; Toker et al., 2014). Since the particle size of some proteins and pectin is much smaller than 0.01 μm , the membrane able to completely remove them should be capable of removing all kinds of microorganisms. This means that clarification and cold-sterilization of fruit juices by ultrafiltration can be simultaneously achieved.

Table 4 indicates the examples of studies on the application of ultrafiltration to cold sterilization of clear fruit juices. It is obvious that ultrafiltration is very effective on the removal of microorganisms. If the pore size of ultrafiltration membrane is small enough, microorganisms in

clear fruit juices can be completely removed. Furthermore, ultrafiltration for removing microorganisms from apple juice on a pilot scale was studied (Li et al., 2006). This study compared ultrafiltration membranes made of ceramic and organic polymer. The logarithm value of bacteria reduction rate of ceramic membrane was found to be 9 whereas that of organic polymer membrane was only about 5.

In addition, some studies proved that the sterilization of clear fruit juices by ultrafiltration is more effective than that by ultraviolet radiation while combination of both methods may improve sterilization effect. For example, study on white birch sap indicated that the total account of microbes was decreased from 1.6×10^4 CFU/mL to 2.0×10^2 CFU/mL by ultrafiltration using a membrane with 0.03 μm pore size, to 2.1×10^3 CFU/mL by ultraviolet irradiation, and to the level which was not detected during 40 days of storage at 4 or 25 °C by the combination of ultrafiltration and UV (Jeong et al., 2013).

Storage studies also proved that ultrafiltration was effective on the cold sterilization of clear fruit juices. For example, banana juice filtered by ultrafiltration membrane can be stored up to 1 month without any deterioration (Sagu et al., 2014). Cold sterilization of bottle gourd (*Lagenaria siceraria*) juice by hollow fiber ultrafiltration was feasible since the quality of sterilized juice did not adversely alter after 8 weeks of storage (Mondal et al., 2016).

Microfiltration Membrane for Sterilizing Drinks

Sterilization of Fruit Juice

Microfiltration may also play an important role in sterilization of drinks. The use of this method alone for sterilizing

Table 4 Application of ultrafiltration (UF) to cold sterilization of clear fruit juices

Ultrafiltration information	Examples of drinks				
	Jamun juice ^a	Coconut water ^b	Pear juice ^c	Apple juice ^d	Pineapple juice ^e
Scale (L)	Laboratory	Laboratory	Pilot	Pilot	Pilot
Membrane pore size	50 kDa	< 0.01 μm	0.05 μm	0.1 μm	30 and 100 kDa
Membrane material	Polysulfone	Polyethersulfone	Ceramic	Ceramic	Polyethersulfone
Membrane module form	Hollow fibre	Flat	Tubular	Tubular	Hollow fibre
Microbial removing efficiency	Total plate count and yeast/mold count (log CFU/mL): 0, respectively	Decrease from 4.16 to 0.0 log CFU/mL	Total plate count decreased from $1.26 \pm 0.36 \times 10^5$ to $1.12 \pm 0.54 \times 10^2$ CFU/mL	Logarithm value of bacteria reduction rate > 9	Completely removed

^aGhosh et al. (2019)

^bLamdande et al. (2020)

^cZhao et al. (2016)

^dLi et al. (2006)

^eLaorko et al. (2010)

drinks may be feasible depending upon their initial status of microbial contamination and membrane pore size. This method may be used before the ultrafiltration of drinks for attenuating the fouling problem of membrane. In another way, this method may be used for removing microbial spores or microbes with large cell sizes before or after other cold sterilization methods such as high pressure homogenization of cloudy (or called opaque) drinks.

A microfiltration system made by using the module of PSF (polysulfone) hollow fiber membrane with a pore size of 0.2 μm reduced total plat account of microorganisms from 1.53×10^4 – 3.34×10^6 CFU/mL to <25 CFU/mL in pineapple juices, which had 37 L/m²h average permeability and decreased protein content (g/100 mL) from 0.409 to 0.375 (Laorko et al., 2010). When coconut water with 140 CFU/mL total plat account of microorganisms was filtered by first using a 0.8 μm followed by 0.45 μm cellulose nitrate membranes, the growth of microbes was not observed in the permeate stored in a glass bottle after 180 days of storage while the filtration processing increased total soluble solids (Mahnot et al., 2014). Positive results of membrane filtration for cold sterilization of juices from other fruits, e.g. mosambi (Nandi et al., 2009; Rai et al., 2006) and passion fruit (de Oliveira et al., 2012) have also been stated. Microfiltration for cold sterilizing pineapple juice on a pilot scale was found to be effective (Carneiro et al., 2002). The membrane with a pore size of 0.3 μm was able to reduce total microorganism of pineapple juice to the level that is safe for human consumption while content of its solid nutrients dissolved was not decreased.

The solid particle size of opaque or cloudy fruit juices is much larger than the pore size of ultrafiltration membrane and the size of the smallest waterborne or airborne microorganisms (see “2.”). For example, cloudy apple juice is a suspension of particles with sizes of 0.25–5 μm (Tetik et al., 2013). Ultrafiltration can remove the solid particles from opaque fruit juices, which therefore is not a proper method for cold-sterilizing them. However, the solid particles of cloudy fruit juices can penetrate through microfiltration membranes with proper pore sizes. Therefore, microfiltration may be applicable to cold sterilization of cloudy fruit juices.

Although the smallest pore size of microfiltration membrane is much larger than the size of the smallest waterborne or air borne microorganisms so that the complete removal of microorganisms by the microfiltration membrane may not be assured, the membrane with a proper pore size (e.g. $0.1 \leq 0.4 \mu\text{m}$) should completely remove the smallest spore of the waterborne or air borne microbes. Especially, as described in “Introduction”, such a cold method as high intensity pulsed electric field can effectively kill all microbes except for the inactivation of microbial spores. This means that the combination of microfiltration (for removing spores) with other methods of cold sterilization

(inactivating vegetative cells) may be very effective for the sterilization of cloudy fruit juices. Some solid particles in a cloudy fruit juice may have larger sizes as compared with the smallest spore (0.4 μm , see Table 1) of some waterborne or airborne microbes so that they may be lost when the complete removal of the microbial spores is assured. This problem should be easily tackled by homogenizing all solid particles of the cloudy fruit juice to the size less than 0.4 μm , which also makes the cloudy fruit juice to be more stable. It should be noted that the size of solid particles in different varieties of cloudy fruit juices may vary a lot. Therefore, the measurement of size of solid particles in the cloudy fruit juice going to be sterilized is necessary before the designation of a proper microfiltration membrane.

Milk

Mainly, application of microfiltration to sterilization of milk has been studied because it contains quite large particles (water-protein-oil emulsion) which may be rejected by ultrafiltration. Ceramic membranes with a pore size of 0.8 μm in diameter effectively removed the spores of pathogenic bacteria which cannot be deactivated by high-temperature and short-time pasteurization from milk without loss of nutritional macromolecules, e.g. proteins (Tomasula et al., 2011). Microfiltration using a membrane with a pore size of 1.4 μm in diameter can reduce 5.63 log microorganisms and greatly extend the shelf life of skim milk while protein transmission is as high as ca. 99% (Dhineshkumar & Ramasamy, 2017; Elwell & Barbano, 2006).

Removal of spores from skim milk by microfiltration on a pilot scale was also studied (Griep et al., 2018). This study indicated that two ceramic membranes with pore sizes of 1.2 and 1.4 μm reduced *B. licheniformis* spores (1.37 μm long and 0.64 μm wide) in skim milk from 6.98 to 2.41 log CFU/mL and from 6.11 to 3.94 log CFU/mL, respectively. Both membranes near-completely removed *Geobacillus* spores (1.59 μm in length and 0.81 μm in width). Protein losses from skim milk were ca.10% and ca.4% for membranes with pore sizes of 1.2 and 1.4 μm , respectively. This level of protein loss may be acceptable considering the benefit (low cost of producing safer products) of microfiltration at a proper pore size to milk industry.

Membrane Filtration for Cold Sterilization of Alcoholic Drinks

For cold sterilization of beer, membrane filtration has also attracted scientist's attention. The advantages of membrane filtration include guarantee of impeccably sterilizing permeate, improvement of productivity and complete removal of microbes with avoidance of negative effect on quality caused by traditional heat treatment method, etc. though

disadvantages, e.g. fouling problem and detectable change in beer quality may occur. Recent study indicated that the quality of sterilized beer obtained by microfiltration using ceramic membrane could be a reliable substitution for the traditionally used sterilization method though the permeation flux was not quite ideal (Kazemi et al., 2013). Membrane filtration was reported to be an effective method for cold pasteurization or sterilization of *Jujube* wine while microfiltration had better production efficiency than ultrafiltration (Kang et al., 1998). Tuber ceramic membranes with pore sizes of 0.2–1.4 μm effectively removed microorganisms from apple cider with the avoidance of changes in pH and soluble solids (Zhao et al., 2015).

Microfiltration has been applied to production of microorganism free beer on a commercial scale (Cimini, 2014). For solving the problem of fouling caused by large amounts of yeast cells, pretreatment by employing absorbent or centrifugation is helpful. Large quantities of macromolecules e.g. proteins unlikely exist in beer. Since trace amount of them may cause precipitates during storage, use of ultrafiltration membrane to remove them should be helpful for improving beer quality.

Sterilization of Drinks by Membrane Filtration on an Industrial Scale

Application of microfiltration or ultrafiltration to cold sterilization of drinks on an industrial scale has been extensively studied and applied in practice (de Oliveira et al., 2012; Dhineshkumar & Ramasamy, 2017; Karmakar & De, 2017). The schematic diagram of cold sterilization or pasteurization based on microfiltration or ultrafiltration is summarized in Fig. 1.

Adaptability of Membranes

Hydrophobic membranes cannot be wetted spontaneously by water (Mulder, 1996). They must be pre-wetted usually by alcohols (e.g. ethanol) when they are used in filtering aqueous solutions. This appears to indicate that these membranes may have a quite limited potential in terms of sterilizing drinks. However, such treatments are required only at the initiation of use of the membranes. In other words, once wetted membranes can be used without any additional treatment until the end of their life. Therefore, the requirement of pre-wetting may not be a big issue when the membrane used has reasonably long lifetime. Membranes made of polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC) are samples of this kind of membrane, which can be used to make micro- or ultra-filtration membranes.

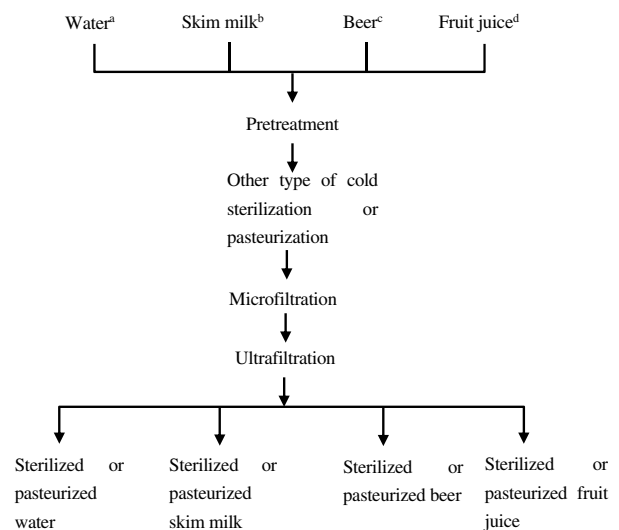


Fig. 1 Schematic diagram of cold sterilization or pasteurization of some soft drinks or beverages by microfiltration or ultrafiltration. **a** Pretreatment might be coagulation, or sand filtration, or carbon filtration, or centrifugation, or their combination while other type of cold sterilization or pasteurization might be UV irradiation, or pascalization (high pressure), or high intensity pulsed electric field, or ultrasound and sometimes these pretreatments might not be necessary depending upon the purity of water source; **b** Pretreatment might not be necessary and ultrafiltration or UV irradiation may not be applicable; **c** Pretreatment might be centrifugation or might not be necessary while other type of cold sterilization or pasteurization might not be used and in most cases microfiltration might be enough; **d** Pretreatment might not be necessary and ultrafiltration or UV irradiation may not be applicable

Hydrophilic membranes usually have good water permeability. They have a potential in terms of cold sterilization of drinks. Membranes made of polyethersulfone (PES), polysulfone (PSF), polyacrylonitrile (PAN), silica/polyvinyl alcohol and cellulose acetate (CA) are good samples of this kind of membranes (Carneiro et al., 2002; Ghosh et al., 2019; Karmakar & De, 2017; Mahnot et al., 2014). Especially, if membrane used for sterilization should be replaced frequently, then the hydrophilic polymeric membranes with limited long-term stability may be of choice as well because the effect of possible degradation of membrane is not likely to be prominent in relatively short operation time.

Good chemical resistance of a membrane is necessary for treating alcohols and acids. For this reason, the use of ceramic membrane may be the best selection for sterilizing alcoholic or acidic drinks. Another advantage of ceramic membrane is that it is able to withstand the harsh conditions of high frequency cleaning (including backflushing) (Charcosset, 2021). On the other hand, ceramic membrane has the following major disadvantages: brittleness, low area to volume ratio, low selectivity in large scale and high production cost (Bolto et al., 2020).

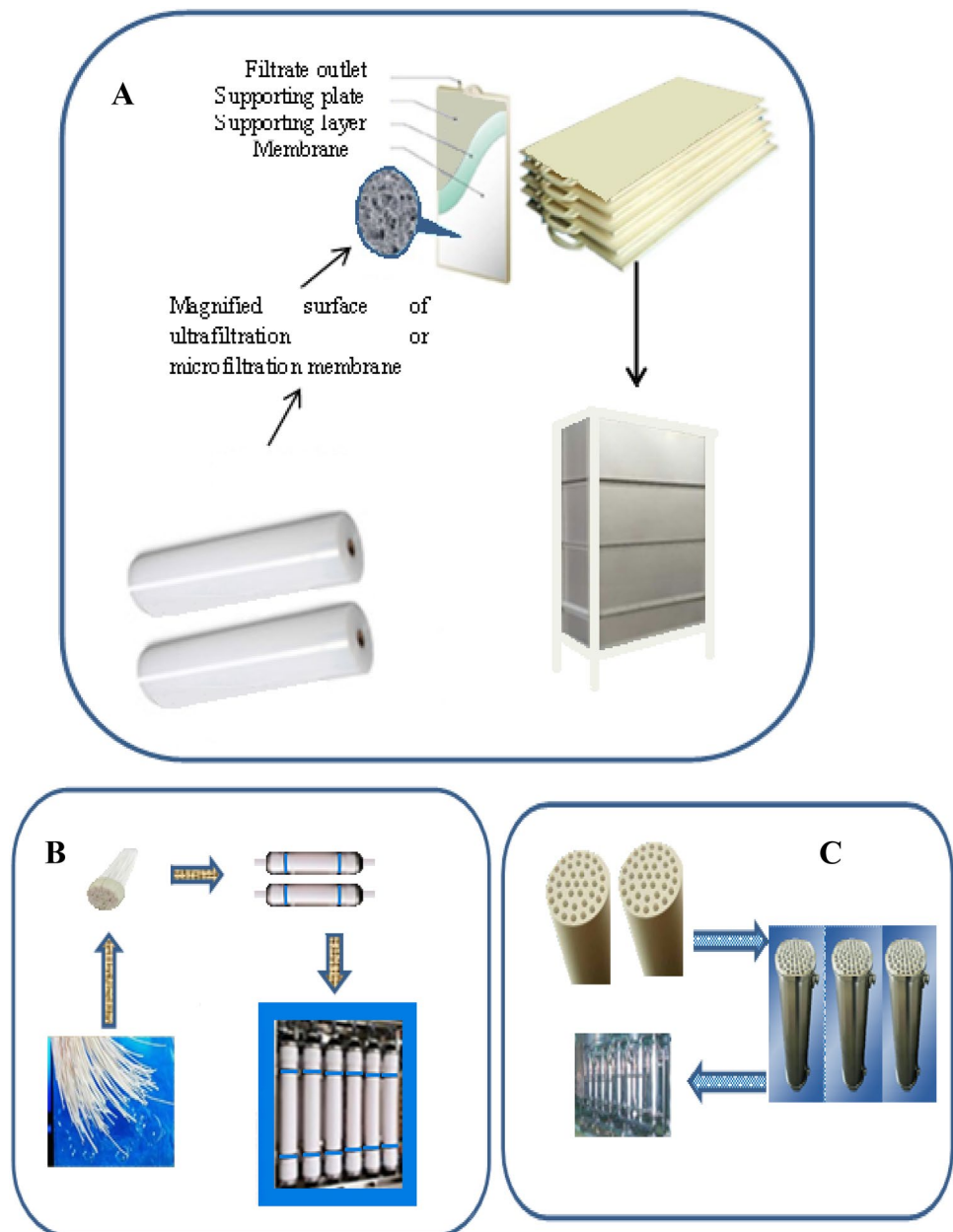
Some organic membranes currently and commercially available, such as PSF and PAN can be damaged by alcohols and acids so that their performance was degraded for a long term filtration operation (Shukla & Cheryan, 2003). The use of this kind of membranes for sterilizing alcoholic or acidic drinks for a long term potentially takes the risk of penetration of microorganisms through degraded membrane. However, there still have been extensive studies on application of this kind of membranes to the sterilization of alcoholic or acidic drinks. The application of membranes made of polypropylene and polyethersulfone having the same pore size ($0.2\ \mu\text{m}$) to filtration of white wine was compared (Ulbricht et al., 2009). The results indicated that

polyethersulfone strongly absorbed polyphenols and polysaccharides to develop a fouling layer, but polypropylene did not. A filtration system using poly(vinylidene fluoride) hollow fiber membranes was reported to withstand aggressive solutions for cleaning (El Rayess et al., 2011).

Module and Material of Membrane Filtration Membrane

For engineering purpose, proper assembly of membrane is important. Several kinds of membrane filter systems have been developed, which are shown in Fig. 2.

Fig. 2 Different filter systems based on flat ultrafiltration or microfiltration membrane for sterilization or pasteurization of beverages. **a** flat membrane, **b** hollow fiber membrane, **c** tubular membrane (Cui et al., 2010)



Organic membrane can be made into flat-sheet or hollow fiber form of structure because it is elastic and soft, which can also be made into tubular type of membrane. Inorganic membrane (e.g. ceramic membrane) is made into tubular form of structure to overcome its inflexibility and fragility. Flat-sheet ceramic membrane is also commercially available.

Membranes are assembled into a module and then several modules are mounted on a rack, having the same feed and discharge pipeline. Nowadays, Plate and Frame Module (A in Fig. 2), Hollow Fiber Module (B in Fig. 2), Tubular Module (C in Fig. 2), Capillary Module and Spiral Wound Module have been developed. Most organic membranes are assembled into Plate and Frame Module, Spiral Wound Module or Hollow Fiber Module whereas inorganic membrane is assembled into Tubular Module or Capillary Module. One application sample is the cold sterilization of beer using a cross-flow systems based on hollow fiber module (Esmaili et al., 2015). A comparison study found that hollow fiber module using 0.1 µm membrane was better for cold sterilization of coconut water than that using 0.2 µm microfiltration and ultrafiltration membranes (Laorko et al., 2017). The ability of a plate and frame module using microfiltration membrane to remove bacteria and spores from beer was reported in a literature (Shekin, 2021). A tubular module using 1.4 µm membrane reduced the total mesophilic microflora about 4 Log and 2 Log from ovine milk and bovine milk, respectively (Panopoulos et al., 2020). Spiral Wound and Hollow Fiber Modules pack membranes with the highest area per unit volume whereas Plate and Frame or Tubular Module packs membrane with the lowest among all kinds of membrane modules. The characteristics and prices of different membrane modules are shown in Table 5, which usually vary with the variation of original materials used for making the membrane.

Materials most commonly used for making ultrafiltration or microfiltration membranes and their characteristics are shown in Table 6. The membranes shown are commercially available and they do not have health or environmental safety

problems. The removal of bacteria from water by PVDF with 0.2, 0.1 or 0.05 µm was found to be 100% (Ghayeni et al., 1999). PES with 0.45–0.65 µm pore sizes was reported to be good for removing yeasts from beer (van der Sman et al., 2012) and cold-sterilizing peapple juice (Carneiro et al., 2002). The feasibility of PAN for cold-sterilizing tender coconut water was indicated (Karmakar & De, 2017). Membranes made of other materials with the pore size of microfiltration for removing bacteria from fruit juices and other drinks were summarized in literature (Liu et al., 2022; Conidi et al., 2020; Bhattacharjee et al., 2017; Cassano, 2015). Preparation of an ultrafiltration or microfiltration membrane using silica/polyvinyl alcohol is also feasible (Ran & Wu, 2017).

Modeling of Membrane Filtration Process

The design, production, selection and operation of ultrafiltration or microfiltration systems need process modeling. Extensive studies on the modeling of membrane filtration can be found in literature (Khac-Uan & Félix, 2020; Krippa et al., 2020; Yusuf et al., 2016). Here, some models widely accepted are described.

The production capacity of a membrane (i.e. amount of a beverage sterilized or pasteurized per hour per m²) is one of the critical index for its application in practice. The production yield of a selected membrane may be predicted by the following equation:

$$V = A_m \times B_f \times t, \quad (1)$$

In the equation, V is the total volume (m³) of beverage filtered during working time; t is working time (h) required for finishing filtration; A_m is membrane area (m²); B_f is beverage flux (m³/m².h) of membrane at a certain pressure, which may be established through experimentation. By using Eq. (1), the required area of membrane can be calculated according to a required amount (i.e. V) of sterilized or pasteurized beverages. B_f is dependent upon pore volume and polarity of

Table 5 Characteristics and prices of different membrane modules commonly used (Zirehpour & Rahimpour, 2016)

Items	Name of module				
	HFM	PFM	SWM	CM	TM
Packing density	High	Low	Intermediate	Intermediate	Low
Cleaning	Difficult	Easy	Intermediate	Easy	Easy
Pressure drop	High	Intermediate	Intermediate	Intermediate	Low
Operation under high pressure	Upto 6.9 MPa	Upto > 0.7 MPa	Upto 6.9 MPa	Upto 0.7 MPa	Upto > 0.7 MPa
Restriction of membrane form	Yes	No	No	Yes	No
Manufacturing cost	Low	High	Moderate	Moderate	High

HFM hollow fiber module, *PFM* plate and frame module, *SWM* spiral wound module, *CM* capillary module, *TM* tubular module

Table 6 Materials most commonly used for making ultrafiltration membranes and their critical characteristics

Membrane material	Characteristics			
	pH range	Maximum inflow pressure	Maximum operating temperature	Adaptable membrane form
PAN	3–9	0.3 MPa ^a	50 °C	Flat, hollow fiber, spiral wound
PS	2–12	0.3 MPa ^a	90–95 °C	Flat, hollow fiber, spiral wound
PSF	2–12	0.3 MPa ^a	80 °C	Flat, hollow fiber, spiral wound
PES	1.5–13	0.3 MPa ^a	80 °C	Flat, hollow fiber, spiral wound
PVC	3–9	0.3 MPa ^a	80 °C	Flat, hollow fiber, spiral wound
PVDF	2–11	0.3 MPa ^b	140 °C	Flat, hollow fiber, spiral wound
Ceramic	1–14	Up to > 1.6 MPa	Up to 200 °C	Tubular
PPO	2–11	Up to > 0.3 MPa	Up to > 90 °C	Flat, hollow fiber, spiral wound
PFE	2–13	0.3 MPa	40 °C	Flat, hollow fiber, spiral wound
PTFE	1–14	Up to > 0.5 MPa	Up to > 130 °C	Flat, hollow fiber, spiral wound
FEP	2–13	Up to > 0.3 MPa	90 °C	Flat, hollow fiber, spiral wound
PE	2–13	0.4 MPa	35 °C	Flat, hollow fiber, spiral wound
PP	2–13	0.3 MPa	90 °C	Flat, hollow fiber, spiral wound
CA	6–8	0.3 MPa	35 °C	Flat, spiral wound

PAN polyacrylonitrile, PS polystyrene, PSF polysulfone, PES polyethersulfone, PVC polyvinyl chloride, PVDF polyvinylidene fluoride, PPO polyphenylene oxide, PFE polyfluoroethylene, PTFE polytetrafluoroethylene, FEP fluorinated ethylenepropylene; PE polyethylene, PP polypropylene, CA cellulose acetate

^aInside-out pressure mode hollow fiber

^boutside-in pressure mode hollow fiber

membrane as well as properties e.g. viscosity, solutes concentration and pH of beverages at a certain pressure. Therefore, B_f is changeable during filtration process. Particles (i.e. from permeate) with their size < membrane pore size may deposit on or be absorbed by pore walls, which results in decreases in pore volume and subsequent reduction of B_f or production yield of sterilized or pasteurized beverage. Furthermore, selection of proper pore size of membrane is also important for achieving acceptable production capacity. Therefore, adequately desired production yield is achievable by selecting a proper membrane with an adequate filtering area, pore size, thickness and polarity.

The fouling problem of membrane which greatly reduces production yield should not be ignored during stage of designing and establishing a membrane system for sterilizing or pasteurizing a beverage. Several studies have attempted to reduce fouling problem of membrane, which involve production of antifouling membrane (Wang et al., 2017), addition of reagents, e.g. enzyme or chitosan and centrifugation (Domingues et al., 2014). Furthermore, pretreatment of beverage by microfiltration or other method before ultrafiltration is usually implemented to tackle fouling problem.

Furthermore, macromolecules (i.e. from concentrated solution) rejected by membrane may develop a fouling layer

on the surface of membrane. The decrease in pore volume (DV_p) during working time (t) is predicted by the following equation (Bowen et al., 1995):

$$DV_p = [1/(P_{m0}t) + (K_s/2)]^{-1}. \quad (2)$$

In the equation, K_s is standard blocking constant [m^{-1} ; which may be considered as combination of membrane intrinsically hydraulic resistance (R_M) which is a fixed value for certain membrane unless significant change in the status of the membrane, concentration polarization resistance (R_{CP}) and fouling resistance (R_F)] while P_{m0} is initial permeability of membrane. It would be worthwhile further investigating into the relationship between R_M and K_s as well as that between R_M and R_F in future.

Total R_m changes as filtration time is extended or number of runs is increased. Total R_m of a particular membrane for filtering a particular beverage can be established through experimentation. Establishment of R_M and R_F for filtering coconut water by hollow fiber membrane based on copolymer of PN has been well described (Karmakar & De, 2017). The authors supposed that total R_M after N th run may be modeled by the following equation:

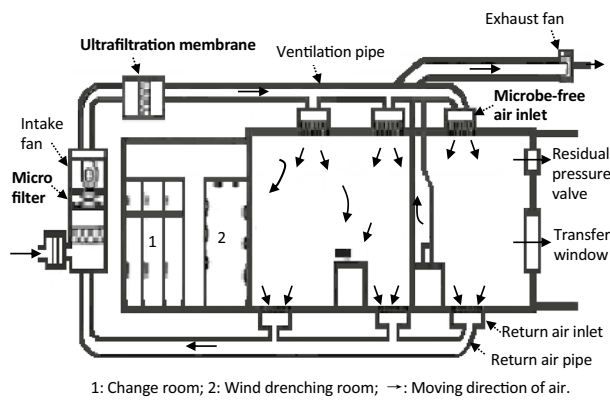


Fig. 3 Critical constitute of sample of an aseptic workshop based on ultrafiltration membrane

$$R_{M(N)} = R_{m(N-1)} + R_{irr(N)} = \Delta P / (\mu_w P_{mw(0)}). \quad (3)$$

In the equation, $R_{M(N)}$ or $R_{m(N-1)}$ is membrane resistance corresponding to N th or $N-1$ th run, respectively; $R_{irr(N)}$ is irreversible membrane resistance after filtering at the end of N th run which is associated with the irreversible change in membrane permeability because of irreversible membrane fouling or aging of membrane; ΔP is trans-membrane pressure; μ_w is water viscosity (0.9×10^{-3} Pa s at 30°C , determined by a U -Tube viscometer); $P_{mw(0)}$ is permeability of membrane at the end of N th run. Trans-membrane pressure (ΔP) required for beverage to permeate through a membrane can be calculated by the following equation (Sampath et al., 2014):

$$\Delta P = R_m \times P_m \times \eta. \quad (4)$$

In the equation, R_m is beverage (or hydraulic) resistance (1/m) of membrane while η is beverage viscosity (Pa.s) at 25°C .

The sterilization or pasteurization efficiency (SE or PE; %) of membrane is another one of the critical index for its application in practice. SE or PE can be described by the following equation:

$$\text{SE or PE} = [(O_{cfu} - P_{cfu}) / O_{cfu}] \times 100\% = [1 - (P_{cfu} / O_{cfu})] \times 100\%, \quad (5)$$

In the equation, O_{cfu} is total number of bacterial colonies in culture medium of 1 mL original beverage while P_{cfu} is total number of bacterial colonies in culture medium of 1 mL pasteurized beverage by membrane.

Application of Ultrafiltration Membrane to Aseptic Workshop

Establishment of aseptic workshop can be a very good or efficient method to achieve the aseptic packaging of beverages. Critical unit of an aseptic workshop is an air purification system. Ultrafiltration membrane can be used for building the core part of the critical unit of an aseptic workshop. Figure 3 illustrates the critical part of structure of an aseptic workshop based on ultrafiltration membrane. Control of proper air pressure and seal are also critical for constructing the aseptic workshop. Generally, the air pressure of clean (or aseptic) area should be 5 Pa higher than that of non-clean (or non-aseptic) area and 10 Pa higher than that of outside of the aseptic workshop. The quantity of positive air volume required (V_{air}) can be calculated by using the following equation:

$$V_{air} = k \Sigma (\nu L). \quad (6)$$

In the equation, k is the calibration constant which is determined on quality of seal of enclosure structure (normally being 1.1–1.3); ν is the leakage air volume per unit length of the enclosure structure ($\text{m}^3/\text{h.m}$); L is the total length of cracks of the enclosure structure. The air purification efficiency (APE) of the ultrafiltration membrane can be described by the following equation:

$$\begin{aligned} \text{APE} &= [(OAC_{\text{particle}} - PAC_{\text{particle}}) / OAC_{\text{particle}}] \times 100\% \\ &= [1 - (PAC_{\text{particle}} / OAC_{\text{particle}})] \times 100\% \end{aligned} \quad (7)$$

In the equation, OAC_{particle} is the concentration of dust particles in original air while PAC_{particle} is the concentration of dust particles in the air purified by the ultrafiltration membrane. The prediction of air volume passing through a selected membrane during the stage of design can be done by the equation similar to Eq. (1). By using this equation, the area of membrane can be determined for producing required amount of purified air for establishing the clean (or aseptic) area of the aseptic workshop. The traditional classification of aseptic workshop and conditions for air purification is shown in Table 7.

The ultrafiltration membranes suitable for pasteurizing clear beverages or drinking water may also suitable for removal of dust or fine particles from atmosphere since their size is normally larger than that of water borne micro-organism. The stopper made of elastic rubber might not be needed when drinks are processed and packaged in an aseptic workshop.

Table 7 Traditional classification of aseptic workshop and conditions for air purification

Class	Level 100	Level 1000	Level 10,000	Level 100,000
Particles (number/m ³)				
≥ 0.5 µm	≤ 3500	≤ 35,000	≤ 350,000	≤ 3,500,000
≥ 5 µm	0	0	≤ 2000	≤ 20,000
Living microbes (number/m ³)				
Settling	≤ 1	≤ 2	≤ 3	≤ 10
Floating	≤ 5	≤ 75	≤ 100	≤ 500
Room temperature (°C)	20–24	20–24	20–24	18–26
Room humidity (%)	45–65	45–65	45–65	50–65
Percentage (%) of fresh air accounting for total air intake	Vertical flow, 2%; Laminar flow, 4%	10%	20%	30%
Airflow velocity through indoor section (m/s)	Vertical flow, ≥ 0.25; Laminar flow, ≥ 0.25	–	–	–
Air changes	–	≥ 50	≥ 25	≥ 15

Conclusion

Ultrafiltration or microfiltration membrane for cold-pasteurization or sterilization of beverages has been widely studied. The illustration of size of the smallest microorganism and theoretical achievement of solving fouling problem provide a solid scientific ground for designing and manufacturing ultrafiltration or microfiltration membrane systems for cold-pasteurization or sterilization of beverage. It is therefore concluded that adaptability of membrane filtration, especially its combination with other safe cold methods, to cold-pasteurization and sterilization of beverages on an industrial scale may be assured without a shadow of doubt in future.

Data availability All data generated or analysed during this study are included in this published article (and its supplementary information files).

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