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Fundamentals and applications of nanobubbles: A review

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ABSTRACT

Nanobubble technology is an emerged solution to address climate change, environmental challenges, cost and energy reduction in industrial processes, optimization of therapeutic and diagnostic techniques and other applications. Although nanobubble production and exploitation is a recently developed field, there are numerous reports and studies of their properties and promising implementation in various sectors. This work aims to give a condense information regarding the most recent (since 2017) scientific findings in the potentials of nanobubbles as a versatile and sustainable technology. Environmental, agricultural, medical/bio-medical and otether applications are reviewed and the most indicative of each sector is presented in detail. A special focus is given on water and wastewater treatment implementation.

Contents

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1. Introduction

Nanobubble production and exploitation gain exponentially increasing attention the past two decades. In fact, nanobubble or fine bubble technology comprises many sectors and its ever growing market share, in 2016, was forecasted to reach €145 M in EU until 2030 (Fig. 1). A recent global report of 2022 refers to multi-billion dollar market with wide spread of applications (BUSSINES WIRE). The main application falls under environmental technologies and specifically water and wastewater treatment. Agriculture and bio-medicine follows, while nanobubbles have entered energy sector as well.

The topic of “Applications of Nanobubbles” is very popular. However, until now, the majority of scientists have investigated the principles and fundamentals of nanobubbles (NBs) (Nazari et al., 2022; Haris et al., 2020). Recently, there is an extreme increasing rate of publications regarding the use of NBs. Unfortunately, until now there is no any collection of the application of NBs. Only a limited number of review articles exist, in which there is a description of the applications of each sub-field in each publication. In particular, review articles have been published regarding environmental areas (Azevedo et al., 2019; Xiao et al., Jun. 2019), water and wastewater treatment (Sakr, 2022; Seridou and Kalogerakis, 2021; Singh et al., 2021; Temesgen et al., 2017; Atkinson et al., 2019), groundwater remediation (Haris et al., 2020; Ye et al., 2019; Alazaiza, 2021), flotation (Li and Zhang, 2022; Rosa and Rubio, 2018), medical/bio-medical applications and food processing (Su, 2021; Batchelor, 2021; Wu et al., 2021). The purpose of this work is to provide a summary of the most recent scientific research on the potential of nanobubbles as a versatile and sustainable technology. The presented findings are both examples of effective techniques successfully applied on a large scale and experimental laboratory bench scale results that can be potentially applied in industrial and field processes.

1.1. *Fundamentals*

Nanobubbles (NBs) are nanoscopic gaseous (typically air) cavities in aqueous solutions that have the ability to change the normal characteristics of water (Kyzas and Mitropoulos, Oct. 2021). Nanobubbles are classified into surface NBs and bulk NBs. Ordinary bubbles have a diameter which range from 1 μm and larger. Nanobubble size classification, according to ISO-20480–1–2017, is given in Fig. 2. Larger bubbles quickly rise to the surface of a liquid and collapse. Nanobubbles which are < 100 nm in diameter will randomly drift owing to what is termed, Brownian Motion and can remain in liquids for an extended period of time (Michailidi et al., 2019). The existence of NBs was confirmed in the last 2 decades.

There are numerous applications in various fields as medicine, biology, chemistry, agriculture, decontamination, materials science, food technology, etc. Recently, there is an extreme increasing rate of publications regarding the use of NBs. Fig. 3a) presents the contribution of research on each sector, while Fig. 3b) shows the exponential research increase in the field.

From the academic perspective, there is a major issue about NBs that have to be addressed. The topic of this debate

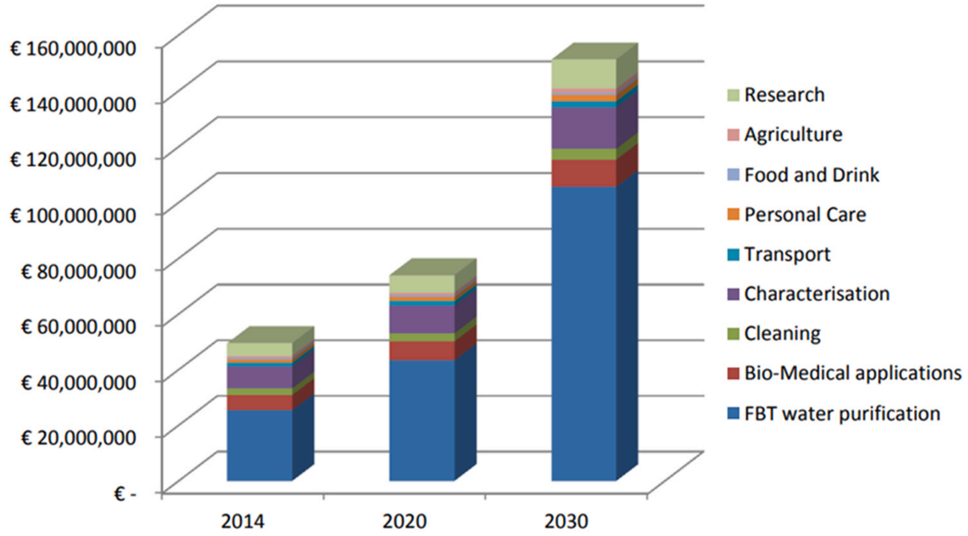

Fig. 1 – Nanobubble market share by sector of application, as foreseen in (BREC Solutions Limited, 2016).

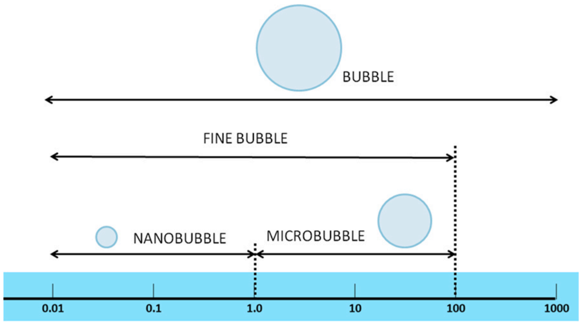

Fig. 2 – Nanobubble size classification, ISO-20480–1–2017.

is the need for a theoretical approach to explain why bulk NBs in water last so long. Long-range hydrophobic attraction, microfluidics, and adsorption onto hydrophobic surfaces are also all areas where NBs are of interest. Furthermore, NB enriched water has entirely different physicochemical properties than water without NBs (Michailidi, 2020). Nanobubbles present high mass transfer efficiency and oxidation ability because of the increased gas-liquid contact area and the generation of hydroxyl radicals when collapsing (Michailidi et al., 2019; Xiao and Xu, 2020; Takahashi et al., 2021).

1.2. Stability of Nanobubbles

The most unique characteristic of bulk NBs is their remarkable longevity. While microbubbles (MBs) have a lifespan of seconds, NBs have been observed to exist for weeks or months (Favvas et al., 2021; Babu and Amamcharla, 2022). A contradiction emerges in systems including NBs considering the fact that their longevity does not follow the Young-Laplace law:

$$\Delta P = \frac{2\gamma}{R} \tag{1}$$

where $\Delta P = P_{vap} - P_{liq}$, the pressure inside (vapor phase) and outside (liquid phase) of the bubble' respectively, and γ is the surface tension. This equation estimates a massive inner gas pressure for NBs and, as a result, the Epstein–Plesset theory predicts that they should dissolve on a timescale of milliseconds (Epstein and Plesset, 1950).

The most popular theory about NBs stability is the electrostatic repulsion theory (Meegoda et al., 2019). Electrostatic repulsion and van der Waals attraction are the two types of surface forces that determine the stability of any colloidal system. As a result, the colloidal stability may be explained using the Derjaguin, Landau, Verwey, and Overbeek (DLVO) theory (Trefalt and Borkovec, 2014). Nanobubble interfaces in pure water have been shown to be negatively charged, indicating the development of an electric double layer around the NBs. The accumulated ions surrounding the bubble surface form a thin layer that functions as a diffusion barrier, decreasing gas dissolution and therefore prolonging the NBs lifespan. This process is known as the ion shielding effect. The external electrostatic pressure generated by the charged NB interface is believed to balance the internal Laplace pressure as shown in Fig. 4, resulting in no net gas diffusion at equilibrium and the NBs remaining stable (Nirmalkar et al., 2018).

Another important theory is the hydrogen bonding model (Jadhav and Barigou, Feb. 2020). The stability of NBs, according to this theory, is due to differential hydrogen bonding at the gas-water interface. The length of hydrogen bonding in NBs is stated to be 0.273 nm rather than 0.295 nm. As a result, the interfacial layer reduces gas diffusivity. The supersaturation theory is also a widely accepted idea. According to this theory, NBs stability is attributed to the slow rate of gas dissolution into the surrounding aqueous phase, which is already gas supersaturated. It was experimentally found that when water is gas supersaturated, the gas transfer rate from the bubble to the liquid is reduced (Alheshibri et al., 2021; Tan et al., 2021).

Other theories like the dynamic equilibrium model (Yasui et al., 2018) and the skin model (Sun, 2018; Wang et al., 2019) suggest that solid materials are involved in NBs stability. The dynamic equilibrium theory assumes that a NB is stabilized against dissolution when part of its surface is coated with a hydrophobic material. Water is repelled by the hydrophobic material and as a result a depletion layer forms on its surface. Gas is confined preferentially in this depletion layer. As a result, the gas pressure at the hydrophobic material's surface rises substantially above ambient pressure. According to the skin theory, a skin of organic material, surfactants etc. fully cover the nanobubbles surface. Gas diffusion from the nanobubble’s interior to the surrounding liquid is totally blocked.

The degree of nanobubbles stability is associated with the absolute value of zeta potential. Zeta potential is defined as the electrical potential at the slipping plane boundary of NBs electrical double layer. Higher zeta potential increases stability for nanobubbles in suspension due to the repulsion between the bubbles. Less stability and coagulation occur from a reduced zeta potential. Zeta potential of bulk NB suspensions is a function of electrolytes, surfactants in the

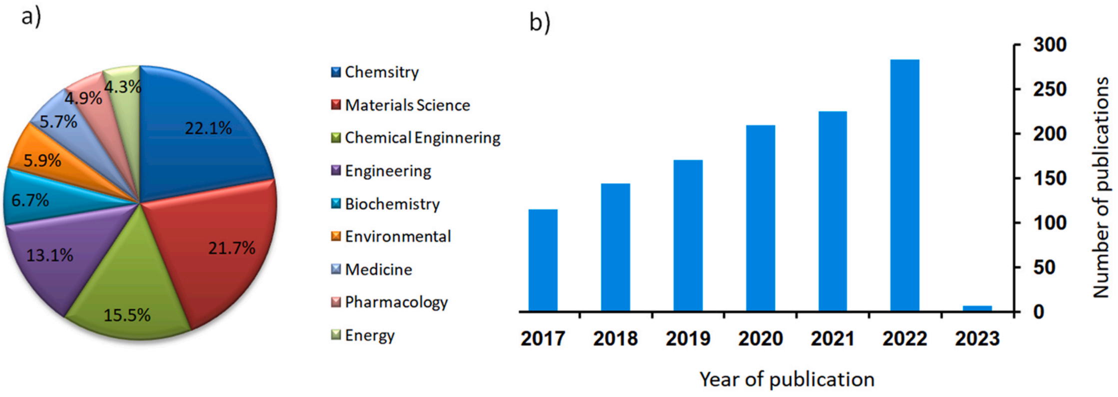

Fig. 3 – Research publication since 2017 with “nanobubble technology” as search term; (a) publications by sector and (b) number of published works per year.

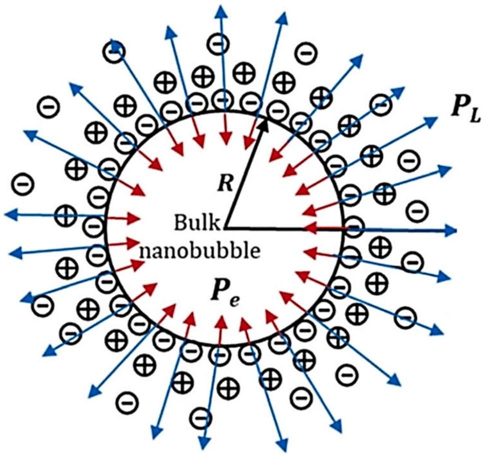

Fig. 4 – Schematic representation of the electrostatic pressure of a charged nanobubble interface.
Reproduced from (Nirmalkar et al., 2018) with permission from the Royal Society of Chemistry.

solution, pH and gas type. It has been reported that nanobubbles are negatively charged at pH values between 4 and 12 with zeta-potential values between – 4.3 and – 62 mV respectively (Han, 2022; Bui et al., 2019). At a neutral pH, zeta potential values were measured to be between – 20 and – 30 mV (Alam et al., 2022).

Meegoda et al. (2018), experimentally studied the effect of the infilled gas type on zeta potential of NB suspensions. The results showed that the zeta potential values are function of the gas solubility and gas diffusion rates. In specific, ozone had the highest magnitude negative zeta potential value followed by oxygen, air, and nitrogen which can be attributed to the different ability of each gas to generate OH^- ions at the water/gas interface.

1.3. *Generation methods*

In the case of surface nanobubbles the following generation methods have been reported: solvent exchange (Fang, 2018; Millare and Basilia, 2018), electrochemical reactions (Gadea et al., 2020; Suvira and Zhang, 2021), immersion of hydrophobic substrate into water with temperature increase or pressure decrease (Li et al., 2019). However, the scientific interest is primarily focused on bulk NBs generation techniques. According to Zhou et al. (2021), there are two basic formation mechanisms for bulk NBs: bubble nucleation where the gas phase emerges from the liquid phase and bubble collapse where microbubbles shrink to NBs. Bulk NBs can be generated by cavitation, electrolysis, nanopore membranes, solvent exchange, compression of MBs and application of electric/magnetic field.

Cavitation occurs when there are sudden variations in pressure in a liquid in areas where the pressure is relatively low. Hydrodynamic cavitation causes vaporization and bubble formation as a result of changes in system geometry due to the variation of pressure in a flowing fluid, like in a Venturi-type circulation (Li et al., 2021; Wu et al., 2022). Hydrodynamic cavitation can occur by mechanical agitation (Counil et al., 2022), depressurized flow constriction (Nazari, 2020) and axial flow shearing (Syaeful Alam et al., 2022) as was developed by Favvas et al. (2021), depicted in Fig. 5. The specific device uses air/water counter flow in a modified Venturi-type tube with a specific internal roughness.

Applying ultrasonic waves to liquids can produce a phenomenon known as acoustic cavitation, which causes changes in local pressure and the consequent creation of bubbles (Bu and Alheshibri, 2021). Short-pulsed lasers that are directed into fluids with low absorption coefficients are utilized in optical cavitation (Hashimoto and Uwada, 2022).

Using electrolysis method, nanobubbles are produced through electrochemical processes on electrodes. In particular, gas bubbles initially form on the electrode surface and then develop until they eventually separate and float (Jadhav and Barigou, 2021; Postnikov et al., 2018). Bulk nanobubbles also may be produced effectively by forcing gas through a porous medium, such as a membrane, into a flowing aqueous or liquid. Porous membranes, primarily made of ceramic, are already employed (Shi et al., 2021; Ahmed, 2018). Solvent exchange has been shown to be a practical and efficient method to generate surface and bulk NBs by replacing a gas/oil-saturated solution (i.e. ethanol) with a poor solvent (i.e. water), which provides a condition of oversaturation for nucleation. Furthermore, it has been reported that bulk NBs can be created by repeatedly compressing microbubbles (Jin, 2020; Jin et al., Mar. 2019). The creation of bulk NBs by external electric/magnetic fields has been developed recently and is highly innovative (Quach et al., 2020; Ghaani et al., Aug. 2022). According to this technique, bulk NBs may be created by passing ionized water via an oscillating electric/

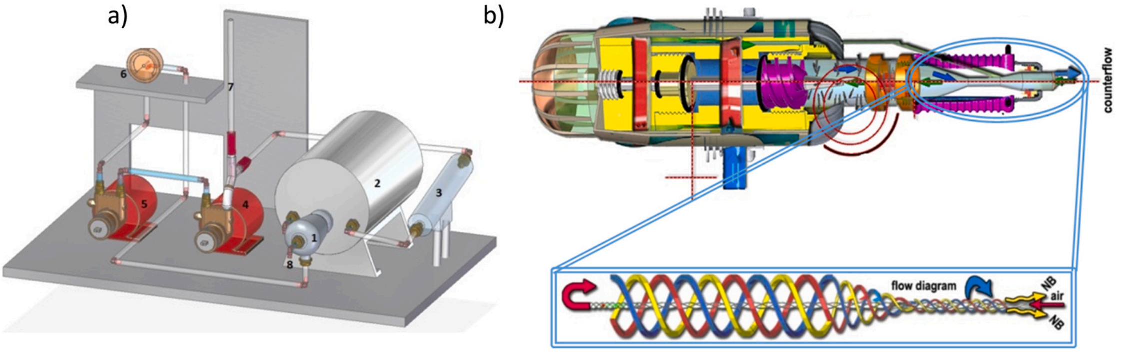

Fig. 5 – Schematic representation of nanobubble generation device by means of advanced hydrodynamic cavitation; a) Batch 4lt NB generator comprised of: 1) NBs' generator, 2) thermostable liquid vessel, 3) transparent tube for liquid observation, 4) 1st pump, 5) 2nd pump, 6) pressure transmitter, 7) gas input through the 1st pump, and 8) gas input through directly the generator; b) Continuous flow NB generator.
Reproduced from (Favvas et al., 2021).

magnetic field. In this case, the production of bulk NBs can be attributed to the increased surface polarization caused by the external electric/magnetic field at the gas- liquid interface which causes cavitation at the liquid interface.

All the aforementioned generation methods are ideal for theoretical laboratory studies. However, very few of them can produce NBs at a high enough rate for industrial applications. For engineering applications such as froth flotation and wastewater treatment, hydrodynamic or acoustic cavitation are the most popular method to produce nanobubbles on a large scale (Oliveira et al., 2018). A large number of microbubbles cause aqueous solution to turn milky during the generation of nanobubbles via hydrodynamic cavitation. Shortly after cavitation stops, the solution becomes clear again as microbubbles rise buoyantly and burst at the top of solution. Therefore, the search for NBs production methods that can generate high concentrations of bulk NBs, are affordable, simple to scale up, and have process control is ongoing to address certain industrial demands.

1.4. *Measurement of NBs*

Measurement methods based on light scattering like NTA (Nanoparticle Tracking Analysis) and DLS (Dynamic Light Scattering) are the most commonly used for determining the size and concentration of nanobubbles in water (Eklund et al., 2021; Jadhav and Barigou, 2021). Nanobubbles like other nanoparticles scatter light and present Brownian motion. Brownian motion refers to the random movement of very tiny particles in a liquid or gas; it was named after botanist Robert Brown, who researched the phenomenon extensively (Nelson, 2020). Both NTA and DLS methods measure Brownian motion and relate it to a hydrodynamic diameter equivalent, with the motion of smaller nanobubbles being more exaggerated. Nanoparticle Tracking Analysis uses image analysis to measure Brownian motion by tracking the movement of each nanobubble, this movement can be related to nanobubble size. Dynamic Light Scattering observes the time dependent variations in scattering intensity resulting from the relative Brownian movements of the nanobubbles within a sample. The DLS method does not visualize the nanobubbles (Eklund et al., 2021). The average nanobubble size can be calculated from time-dependent variations in light intensity. The NTA method tracks particles inside a specified volume and the size distribution obtained is a number distribution, which can be used to calculate relative nanobubble concentrations. The intensity distribution obtained by DLS can be transformed to a volume distribution. Because this conversion is based on a variety of assumptions, number distributions generated using DLS are often regarded as inaccurate.

2. Environmental applications

2.1. *Water quality control and management*

Water quality refers to the chemical, physical, and biological characteristics of water based on the standards of its usage. The compliance to regulations and legislation is crucial to preserve human and environmental health. Water sources vary including creeks, rivers and groundwater following a multi-stage treatment system before it is delivered to the water network. Drinkable water is treated prior to be channeled to our households most of the times by implementation of a muti-barrier approach. This approach recognizes that although each barrier by itself may not completely remove or prevent pollution, all the barriers together provide greater protection that the delivered water will be safe to drink (Mohr, 2020).

Water pollution has many sources such as solid matter sedimentation, pesticides and chemicals from farming and industry, nutrients such as phosphorous and nitrogen from fertilizers and detergents, toxic algae as result of eutrophication, pathogens disease causing micro-organisms etc (Manasa and Mehta, 2020). Remediation methods of each previously mentioned pollution sources have been developed over last decades including floatation, flocculation, aeration and other treatment technologies (Qadri et al., 2020). To this end, NB technology is reported to be implemented as a standalone method or it is utilized as assistance to the conventional technologies.

2.1.1. *Sediment Decontamination and Lake Management*

Excessive external pollutants (agricultural, domestic sewage, industrial wastewater, etc.) discharged into lakes and rivers, can migrate to the sediment-water interface (SWI) and contaminate the sediments. These deposited contaminants (phosphorus, nitrogen, iron, manganese, methyl-mercury, hydrogen sulfide, etc.) can be released from the sediments under specific conditions and re-contaminate the water column (O'Callaghan et al., 2019). Furthermore, released nutrients like nitrogen (N) and phosphorus (P) can cause water eutrophication and subsequently induce harmful algal blooms (HABs) which cause hypoxia and anoxia (Lytle, 2015). In addition, the decomposition of the precipitated dead algal biomass exacerbates the hypoxic/anoxic conditions due to the high oxygen demand (Nürnberg, 2019). Therefore, the need for efficient oxygen delivery technologies is important for mitigating hypoxia and anoxia and restoring eutrophic waters. Conventional oxygenation methods, like mechanical aeration have been reported to be effective to some extent. However, these techniques cannot effectively be applied in large-scale due to high cost and high energy consumption (He, 2019).

Oxygen and ozone NB technology has been developed to address the SWI aeration issue in a cost effective and environmental friendly way. In order to overcome the hypoxia/anoxia problem, NBs are used due to their long time stability and their high mass transfer efficiency. NBs can either in situ generated or delivered into the SWI by natural porous minerals loaded with oxygen (Wanninayake, 2021).

Wang et al. (2018) developed a novel method to quantify the amount of oxygen nanobubbles loaded in porous material. They evaluated different materials and their oxygen-loading capacities were found to be as follows: activated carbon (AC) > zeolite > biochar > diatomite > coal ash > clay. These differences were mainly attributed to differences in the specific surface area and surface wettability.

Zhang et al. (2018), in their pioneer work, experimentally studied the mitigation of hypoxia/anoxia caused by oxygen nanobubble modified natural particles. In their simulated column experiment, they used oxygen nanobubble modified zeolites (ONMZ) and local soils (ONMS) and they found that the dissolved oxygen (DO) levels were increased in the water column and that anoxic conditions at the SWI were also reversed for several months (Fig. 6).

In a similar study, Yu et al. (2019), developed oxygen nano-bubble-modified minerals (ONBMMs) based on

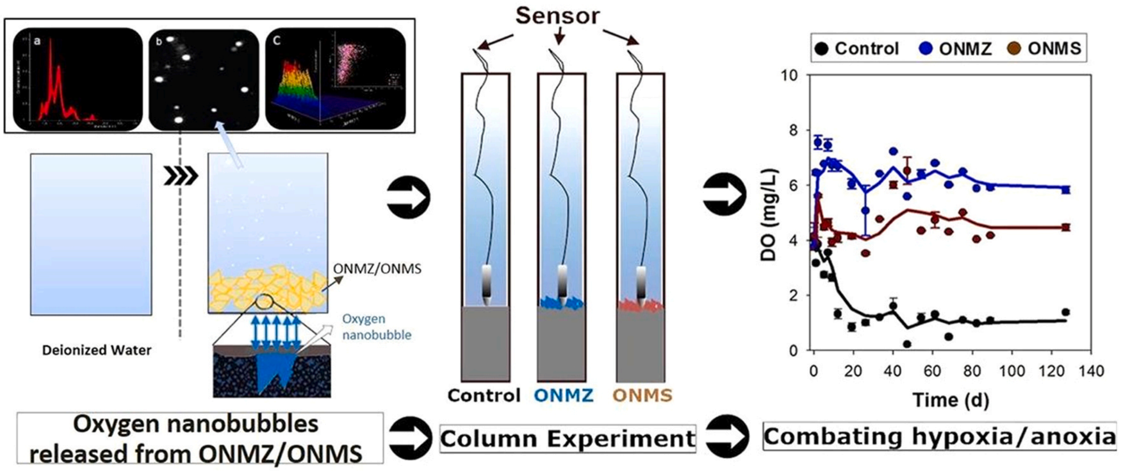

Fig. 6 – Schematic representation of experimental process for combating hypoxia/anoxia by oxygen nanobubbles. Reprinted from (Zhang et al., 2018) with permission from Elsevier.

Table 1 – Water quality changes in treated ornamental pond at Emirates Golf Club.

Parameter (Unit)	25/12/2017	31/03/2018
Dissolved oxygen (mg/L)	4	6.1[a]
Chemical oxygen demand (mg/L)	352	84
Phosphorous (mg/L)	0.33	0.31
Sulphate (mg/L)	5500	2500
Total nitrogen (mg/L)	36	28
Calcium (mg/L)	2500	888
Potassium (mg/L)	850	328
Chloride (mg/L)	15000	11000
Sodium (mg/L)	8500	4920
Salinity (PSU)	40	27
Fecal coliforms (MPN/100 mL)	> 200	< 1.8
Escherichia coli (MPN/100 mL)	> 200	< 1.8

Reprinted from (Kalogerakis et al., 2021) with permission from John Wiley and Sons.

[a] The dissolved oxygen ranged from 6.0-9.7 mg/Lthroughout the test period.

muscovite mineral particles. It was found that (ONBMMs) can effectively increase DO at the SWI and decrease the released amount of phosphorus (P) from the sediments. In another recent work, Tang et al. (2021), observed in their simulated mesocosm experiments that the levels of dissolved arsenic (As) were reduced and also converted to less toxic species after oxygen nanobubble treatment against algal-induced hypoxia/anoxia. Ji et al. (2020), in their experimental study, collected samples of surface sediment and overlying water from a typical algae accumulation zone to perform laboratory tests. They found that the application of oxygen nanobubbles significantly reduced methylmercury (MeHg) levels produced by Hg contaminated sediments and also remediated hypoxic and anoxic conditions. Li et al., in their recent work, developed oxygen-carrying sediment-based biochar (O-SBC) as a novel oxygen nanobubble carrier, using sediment samples from an urban tributary. They reported that (O- SBC) performed long-lasting re-oxygenation for anoxic waters and also observed a microbial community response mechanism for anoxic water restoration.

Aluthgun Hewage et al. (2020) developed an environmentally sustainable on-site method for remediation of river sediments contaminated with organic pollutants and heavy metals, combining ozone nanobubbles and ultrasound technology. In this process, ultrasound energy was used to detach contaminants from the sediments and release them in the water column while ozone nanobubbles were used to degrade organic contaminants and to oxidize the desorbed heavy metals. It was reported that with the application of the specific technique, a contaminant removal efficiency of 91.5% for polycyclic aromatic hydrocarbons (PAHs) was achieved.

Kalogerakis et al. (2021) recently published the results of five field applications of nanobubble technology for ecosystem restoration of large water bodies like lakes and ponds at industrial scale, at several locations world-wide. In this work, a commercial nanobubble generator was used for the production of air nanobubbles with concertation of 2.2×10^8 bubbles/mL at 25 L/min flowrate and mean NB diameter of 97 nm. The results indicated improved water quality parameters with significant reduction in chemical oxygen demand (COD) and pathogens as outlined in Table 1, while Fig. 7 shows the results of on-site application.

Soyluoglu et al. (2022) addressed the issue of geosmin and MIB after blue-algae metabolism affecting odor and taste of water treatment plants using surface water. This study applied oxygen NBs as advanced oxidation technique concluding that the volatilization and thus removal of geosmin and MIB increased by 40% and 20%, respectively. In addition, they find that in real environment, natural organic matter suppressed the performance of this process depending on various water characteristics.

2.1.2. *Soil and groundwater remediation*

These days, contaminated soil and groundwater are gaining greater attention as serious environmental issues. Groundwater and soil contamination provide serious threats to the environment and people, especially in industrialized and urban regions, where the problem is extensive. Human activity is the primary cause of the contamination (Demlie and Wohnlich, 2006; Amin Al Manmi et al., 2019). There are many different types of pollutants found in groundwater and soil, including persistent organic pollutants, organochlorinated pesticides (Persson, 2007), synthetic organic chemicals (Kumari et al., 2008), petroleum hydrocarbon compounds (Cao et al., 2021), pathogens (Shaharoona, 2019), radionuclides (Akortia, 2021), heavy metals (Panwar and Ahmed, Aug. 2018) and xenobiotics. Therefore, remediation

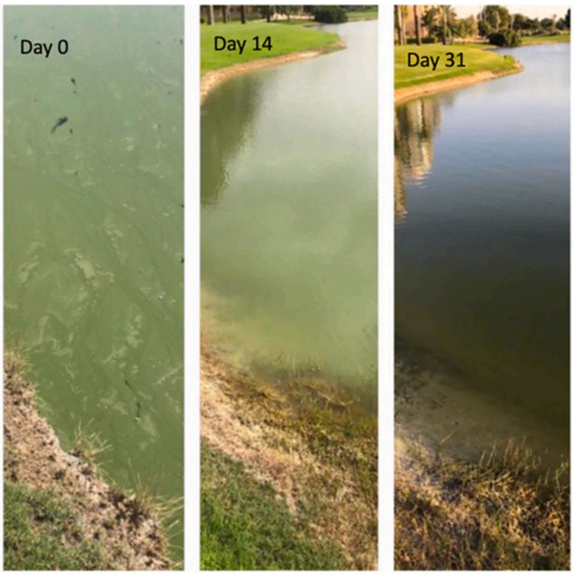

Fig. 7 – The effect of nanobubbles on green algae elimination.
Reprinted from (Kalogerakis et al., 2021) with permission from John Wiley and Sons.

technology for the contaminated sites is urgently needed. Depending on where the treatment takes place, remediation techniques may be divided into in-situ and ex-situ categories. *Ex-situ* remediation involves recovering contaminated soil or groundwater from the subsurface and either treating it on the same site or moving it to another location for treatment (Sharma, 2021; Gerhard et al., 2020). In-situ remediation, on the other hand, is the process of cleaning up contaminated soil or groundwater right in the subsurface (Marschalko, 2022). Due of its lower cost compared to ex-situ remediation, in-situ method is usually preferred.

Air sparging is an in-situ remediation method where a gas, usually air, is pumped into a region underneath the water table using a system of injection wells (Li, 2021). Deep groundwater is forced to circulate by the air injection, and the aeration of the water helps to remove volatiles. Furthermore, air sparging increases the levels of dissolved oxygen in the water enhancing the degradation of pollutants by aerobic microorganisms (Wang et al., 2019; Xu, 2021). Nanobubbles promote bioremediation due to their high mass transfer efficiency, increase the zone of influence (ZOI) due to their high longevity in water and generate free hydroxyl radicals with strong oxidizing ability when collapsing, therefore they are ideal for the purpose in interest (Yao, 2020). Additionally, ozone nanobubbles perform great oxidizing ability in decomposing difficult to decompose organic pollutants. Nanobubbles can also be used as sustainable eco-friendly enhancers instead of chemical ones, in remediation of heavy metal contaminated soil.

Hu and Xia published a series of papers investigating the potential application of ozone Micro-Nanobubbles (MNBs) in organics contaminated groundwater remediation. In one of their studies (Hu and Xia, 2018), laboratory tests were conducted for both surface water and groundwater remediation to degrade methyl orange, which was selected as the representative pollutant. In order to simulate highly permeable soil, glass beads were used. It was reported that ozone MNBs effectively degraded methyl orange while oxygen MNBs showed no oxidation ability. In another study by the same authors (Xia et al., 2019), the mass transfer behavior and the remediation efficiency of ozone MNBs were investigated by both laboratory column tests and trichloroethylene (TCE)-contaminated site field tests.

In another study, Jeong et al. (2017), investigated the potential use of nanobubbles as a remediation on heavy metal contaminated soil. According to the results of this study, the observed increase in efficiency in copper removal, was attributed to the large surface area and high zeta- potential of nanobubbles. In a similar recent work (Kim and Han, 2020), Kim and Han also demonstrated the hydrogen nanobubbles efficiency in remediation of copper contaminated soils. Based on this study, nanobubbles proved to be an eco-friendly enhancer for electrokinetic remediation of heavy metals.

2.2. *Wastewater treatment*

Wastewaters from industry, agriculture and domestic sectors are a severe environmental pollution source in the modern society (Manasa and Mehta, 2020). Wastewater treatment and cleaning is mandatory, especially when hazardous materials are involved (heavy metals, toxic substances etc.). The modern approach to this is a five stage system, applied in all sectors but with adjustments, comprised of a (i) pretreatment stage, (ii) primary treatment, (iii) purification or secondary treatment, (iv) final treatment and (v) sludge management (Crini and Lichtfouse, 2019). From this scheme, the steps of high importance are (ii), (iii) and (iv), where contaminants are removed by various techniques involving intense chemical processes; resulting to be costly. The effect of nanobubbles on every step of wastewater treatment has been investigating showcasing a great alternative to traditional techniques.

2.2.1. *Chemical precipitation*

Chemical precipitation is common method in the secondary treatment for at least the past three decades. Precipitation involves the removal of ions by the addition of chemical compounds of counter-ions in order to change the solubility of the target pollutant (Zhang et al., 2019). The solid particles formed, after this chemical process, are collected by flotation or sedimentation. The challenge of this technology is to overcome the (i) overuse of chemicals, (ii) production of toxic by-products and (iii) high operational cost in an efficient manner.

According to available literature, the investigation of NBs on precipitation contribution has been developed recently; no documentation before 2010 with the higher hype recorded after 2017. The removal of Fe ions by $Fe(OH)_3$ precipitation followed by flotation was studied by Etchepare et al. (2017). They investigated two cases of dissolved air flotation (DAF), microbubbles (MBs) accompanied with NBs and NBs after isolation. The outcome of the study indicated that when MBs are present, the process is faster but the efficiency of NBs solely is greater regarding the separation of formed nanoparticles. Overall, for both cases, the initial Fe concentration played a crucial role; the higher it was the better removal was gained. The latter is attributed to the fact that higher concentration results in better entrapment of NBs as it is described by the authors.

Froth flotation is a process where hydrophobic matter is separated from hydrophilic with the use of various chemicals used as frothers or collectors (Nguyen, 2013). The role of a collector is to make the surcafe of mineral particles hydrophobic. To this end, NBs are reported to assist this purpose resulting in minimizing collector concentration while maximizing the efficiency.

Zhang et al. (2021) present the effect of NBs on the flotation of rutile particles with benzhydroxamic acid (BHA) as collector. The experimental procedure included microflotation tests, BHA adsorption capacity, UV and zeta-potential measurements while the interaction of particles with bubbles was investigated macroscopically with the use of a high-speed camera. The results showed that both in recovery and adsorption capacity, the combination of BHA and NBs performed better than BHA solely, preserving though the trend. This outcome has two ways of interpretation; it indicates that when NBs are present, the concentration of the used collector can be reduced without altering the overall efficiency or that with the same amount of collector the efficiency can be increased by at least 10%.

Nanobubbles in carrier flotation process for fine coal separation was investigated by Zhou (2020). Polysterene was used as collector (carrier) while NBs were generated by hydrodynamic cavitation. The aim of this work was to outline the mechanism of NBs in particle separation by flotation. Authors suggested that NBs served as “bridges” between the hydrophobic PS and fine coal aggregates increasing the flotation efficiency by 43%. The most interesting outcome is the proposed mechanism (Fig. 8) of the effect of NBs supported by the use of E-DLVO theory.

In a subsequent study, Zhou (2022), investigated the effect of aeration rate, preparation time and aging on produced bubble size. The results revealed that preparation time affects NB concentration; as the process continues the concentration of nano size bubbles increases. The most interesting outcome of work is regarding flotation enhancement where quartz particles were used in order to examnie the impact of MBs presene. The result indicated that when MBs are combined with NBs, the recovery of fien grained particles was higher. However, aging experiments showed that after 3 min only NBs remains in the solution. Liu et al. (2021) also refers to “bridging” of particle aggregates by the presence of NBs. Properties such as apparent viscosity and shear stress were also examined for the different hydrophobicity levels. As reference sample, DI water was used with the same amount of hydrophobic reagent to form the three coal hydrophobicity levels (low, medium and high designated as LHC, MHC and HHC respectively). From the conducted measurements, HHC with NBs presented the best performance followed by HHc without NBs. For the rest hydrophobicities, the trend was that the lower the level the less the difference between with and without NBs.

Phosphate ore fine powder flotation was examined with solution enriched with NB and a solution where NBs were absent (Pourkarimi et al., 2021). The conducted study proved the existence of NBs by various direct and indirect measurements such as zeta-potential, FTIR and HPLC. Both FTIR and HPLC showcased that in presence of NBs the adsorption of the collector was lower than in the NBs absence. The measured separation performance presents an increase of ~ 40% in the separation efficiency when NBs are present. Authors attribute this to the particle surface nature alteration due to NBs while suggest that NBs functions as a strong collector.

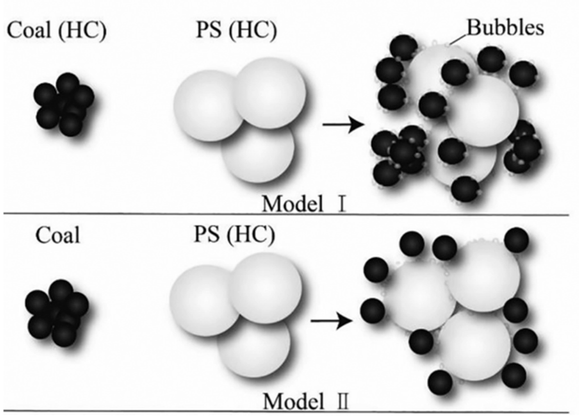

Fig. 8 – Proposed interaction models of NBs and particles during hydrodynamic cavitation; Model I represents proposed interactions at pre-treatment stage and Model II when polysterene particles are used as carriers. Reprinted from (Zhou, 2020) with permission from Elsevier.

In another work, Etchepare with a different team this time, applied NBs in flotation-flocculation process for removal of emulsified oil from saline water (Etchepare et al., 2017). The injection of NBs in the stage of flocculation enhanced the hydrophobicity of the oily aggregates providing better adhesion to NBs leading to an increased efficiency in both processes; by 11% and 3% respectively.

2.2.2. *Adsorption*

One important issue to address in the environmental technologies is the removal of organic pollutants (Karpińska and Kotowska, 2019). The main sources of organic contamination are petrochemical, pharmaceuticals, dye and paper industries (Mandal et al., 2016). Many technologies are proposed for efficient extraction of those compounds depending on the source. The involved principles vary from photocatalysis to oxidation of the organic compounds (Shumbula, 2021).

Adsorption is one the most promising technologies in the field of water/wastewater treatment (Rashid et al., 2021). Pollutants such as heavy metal ions, dyes, phosphates and others are removed efficiently by the use of various adsorbents (Crini et al., 2019). The implementation of adsorption is versatile and cost-effective method due to structural and physicochemical properties of the used materials. Some of the common adsorbents are activated carbon (Kyzas et al., 2016), activated alumina (Tan, 2019), zeolites (Shi, 2018), chitosans (Mohammadzadeh Pakdel and Peighambardoust, 2018) and others (Miricioiu and Niculescu, 2020; Behboudi et al., 2018; Trikkaliotis et al., 2020). Wastewater treatment by adsorption has a two-fold purpose; removal of hazardous contaminants or extraction of valuable compounds such as pharmaceuticals and valuable metals (de Andrade et al., 2018; Asadollahzadeh et al., 2021). Nanobubbles are very promising for assisting the adsorption process by (i) reducing contact time, (ii) increasing contact surface and (iii) deliver pollutant efficiently.

Kyzas (2019), at their pioneer study, investigated the effect of the nanobubbles on dissolved heavy metal adsorption with activated carbon. It was reported that nanobubbles accelerated 366% the adsorption process acting as lead ions carriers as shown in Fig. 9.

A subsequent work of the same team (Kyzas et al., 2020), examined NBs performance with and without agitation on a batch mode adsorption experiments for Pb^{2+} removal by activated carbon. Four cases were evaluated; two with NBs with

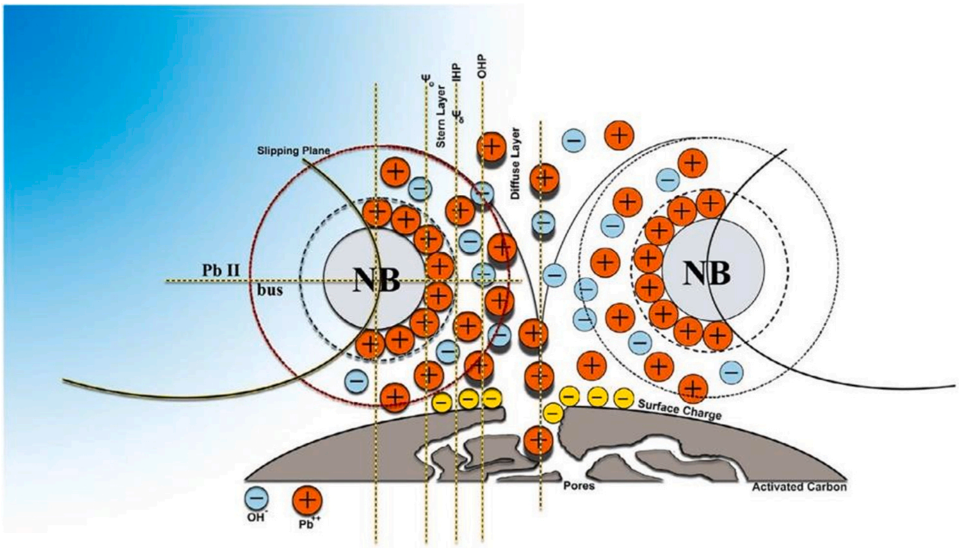

Fig. 9 – Illustration of the nanobubble mechanism of Pb^{2+} adsorption on activated carbon. (Kyzas, 2019).

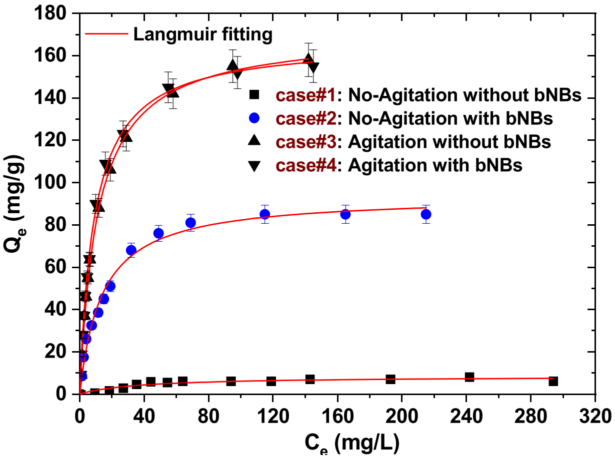

Fig. 10 – Isotherms (Langmuir fitting) for the adsorption of Pb^{2+} onto activated carbon (with and without NBs) in deionized water (with and without agitation) at 25 °C. (Kyzas et al., 2020).

and without agitation and two without NBs at same conditions. The results showed that when NBs where present in the pollutant/adsorbent solution the removal efficiency without “shaking” was even higher as if without NBs but intense agitation (225 rpm). It was concluded that this outcome indicates faster kinetics (Fig. 10) in a fixed bed columns.

Removal of Congo-Red by coal was investigated by He (2021). Again, NBs are presented as “carriers” of the targeted pollutant inside the solution resulting in a increase in contact probability of the contaminant with the surface of the adsorbent. Here, the presence of NBs accelerates Congo-Red removal by 250%; in 1 m minute the concentration reduced to the same level as in 25 min achieved without NBs. The most interesting outcome is the decolorization by 25% even before the introduction of the adsorbent in the solution. Authors suggest that this happened due to oxidation by the free radicals NBs produce.

2.3. *Membrane Defouling*

A membrane is a semi-permeable active or passive boundary that allows certain components of a mixture to flow through while preventing the passage of others. Membrane technologies are extensively used in environmental applications like drinking water treatment, wastewater treatment and desalination (Obotey Ezugbe and Rathilal, 2020). Fouling is the reason for the decrease in membrane performance. In particular, fouling is the phenomenon that causes a membrane to function poorly when suspended or dissolved substances are deposited on its outside surfaces, at its pore openings, or inside its pores. In order to control membrane fouling, various techniques have been developed including chemical cleaning, feed pretreatment, membrane surface modification, flow manipulation, ultrasonic treatment and gas sparging (Chang et al., 2019). Chemical cleaning raises operating costs, might degrade the membrane surface, and discharges hazardous cleaning agents into the environment. High process efficiency and less environment degradation owing to the lack of cleaning chemicals make gas sparging a relative advantage over other methods (Yalcinkaya et al., 2020; Nqombolo et al., 2018; Banerjee et al., 2018).

Nanobubbles have a beneficial impact on gas sparging due to their special properties that distinguish them from larger air bubbles including negative surface charge, increased surface area, high mass transfer efficiency, long stability in water and generation of hydroxyl radicals when collapsing. Nanobubbles introduce turbulence in the liquid environment due to repulsive forces, causing agitation and removal of particles from the membrane surface (Zhou, Aug. 2022). Moreover, NBs mitigate salt precipitation by attracting counterions in their surface and also prevent biofouling by introducing oxidizing hydroxyl radicals that degrade organic contaminants. Therefore, NB technology holds potential as an innovative sustainable technology for membrane fouling control.

Ghadimkhani et al. (2016), investigated the effect of air nanobubbles in ceramic membrane defouling. In this study, humic acid was used as organic foulant for membrane clogging and air NBs were fed to clean the membrane surface. Atomic Force Microscopy (AFM) scans revealed that the permeate flux completely recovered after air NB treatment indicating successful unclog of the membrane pores. It was concluded that air NBs could be protentional used as an economical and sustainable alternative to chemical antiscalants. In another study, Dayarathne et al. (2017),

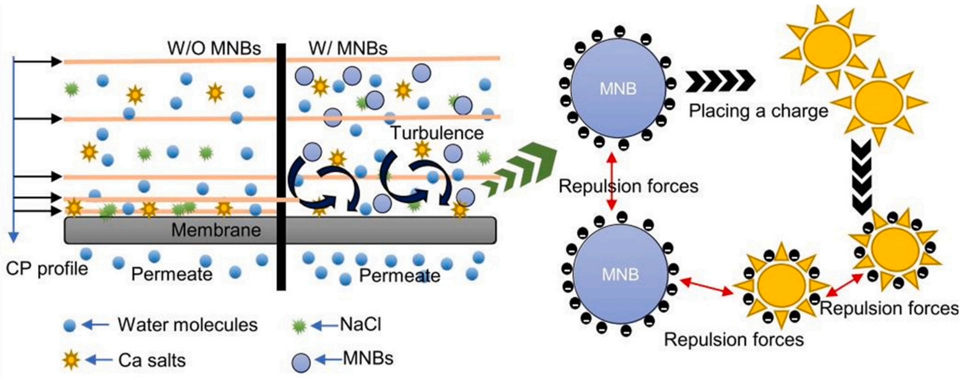

Fig. 11 – Graphical abstract of "Chemical-free scale inhibition method for seawater reverse osmosis membrane process: Air micro-nano bubbles".
Reprinted from (Dayarathne et al., 2019) with permission from Elsevier.

experimentally demonstrated similar results for reverse osmosis desalination process. In this work, lab-scale and pilot-scale experiments were performed in rder to evaluate the impact of air MNBs on membrane defouling. Again, it was reported that the permeate flux was significantly improved and that MNBs can effectively prevent the development of the Concentration Polarization (CP) layer on the membrane surface.

The same authors published a second paper regarding seawater reverse osmosis (SWRO) process (Dayarathne et al., 2019). In this paper, MNBs were evaluated as chemical-free antiscalants in comparison with conventional ones. It was found that MNBs successfully inhibited the formation of salt crystals on membrane surface with superior performance than commercially available chemical antiscalants (Fig. 11).

In another recent work, Farid (2022), studied the role of nanobubbles as a physical cleaning agent in forward osmosis (FO) system for aquaculture wastewater treatment and reuse (Fig. 12). It was reported that the addition of NBs in the feed water significantly improved the performance efficiency and lifetime of the FO membrane. In particular, NBs effectively prevented the accumulation of oxytetracycline (OTC) and other foulants on the membrane surface. The same researchers, in another study, investigated the use of nanobubbles as a chemical-free scale inhibitor in membrane distillation (MD) process. Again, it was observed that the addition of air NBs in the saline feed, successfully reduced membrane fouling and prolonged the effective MD operating time. Moreover, it was suggested that the improved performance can be attributed to the increased surface shear forces at the membrane surface due to the turbulent flow introduced in the feed by the NBs and the electrostatic attraction of counterions on the NBs negatively charged surface.

Analogous encouraging results are reported recently by Ali et al. (2022), who investigated the effect of air MNBs on gypsum scaling on a lab-scale RO membrane. The results of this study showed that the presence of air NBs in the flow significantly reduced gypsum scaling on the membrane surface and also decreased the CP effect.

3. Agriculture applications

3.1. *Horticulture*

Horticulture is one of the most important sectors of agriculture dealing with the cultivation of fruits, vegetables, flowers, medicinal and aromatic plants etc. (Gaur et al., 2018).

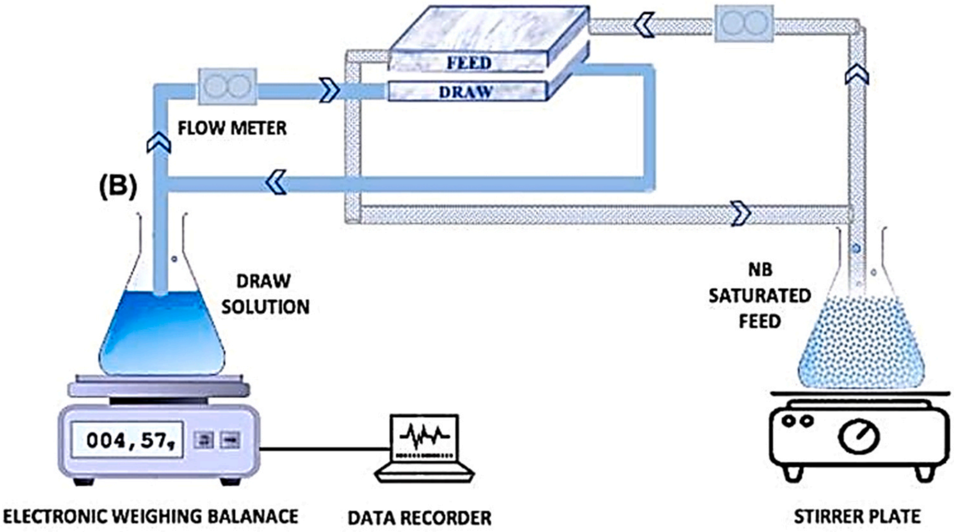

Fig. 12 – The NB-assisted forward osmosis (FO) process.
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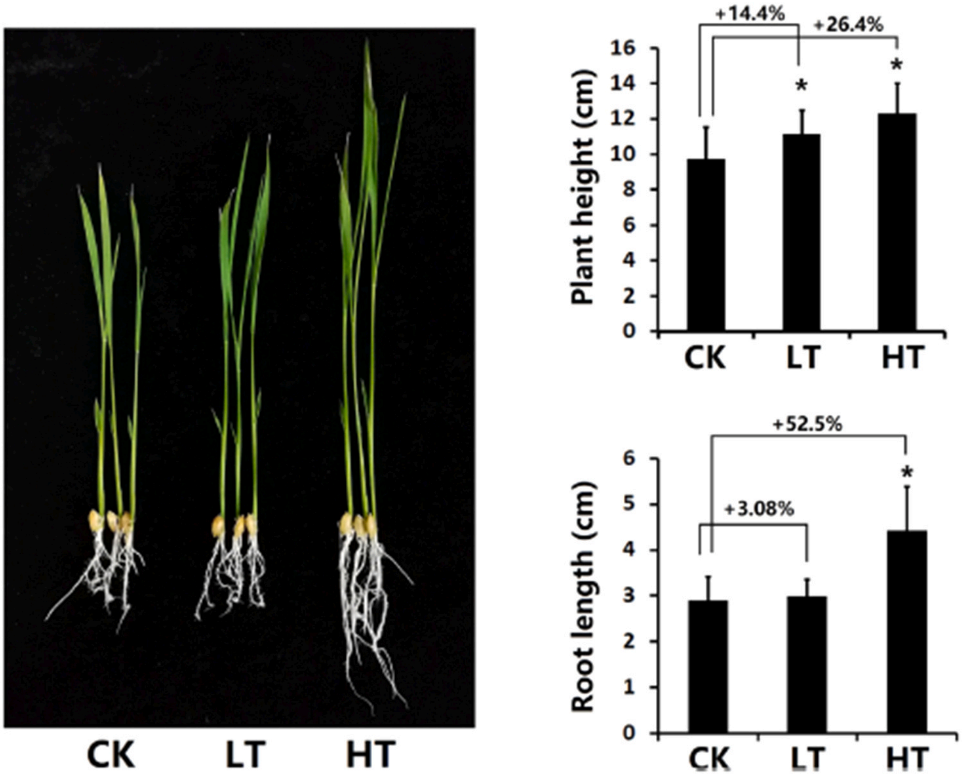

Fig. 13 – Left; Image of representative growth phenotype of three-leaf-stage rice seedlings selected from the three groups: non-treatment control (CK), low-frequency treatment (LT) and high-frequency treatment (HT). Right; Growth trait analysis of rice seedlings in the three groups: plant height (top) and root length (bottom).
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The life cycle of a plant comprises seed germination, sprouting and plant growth (Leopold, 1964). All stages have their requirements for the maximum crop yield and it depends in factors such as soil fertility, water efficiency, diseases and pests (Amaducci et al., 2000). Nanobubbles technology has shown remarkable results in this sector, attributed to the enhancement of the microbial community, encapsulating nutrients, retaining soil humidity etc. (English, 2022; Sha, 2020).

Wang et al. (2020) applied NBs on rice cultivation both in lab and pilot scale. The scope of this study was to minimize the amounts of fertilizers needed as well as to stimulate growth hormones and nutrient absorption genes. Nanobubble generation was done by pressurization method. The experimental procedure was conducted on three sets; without NB as control (CK), low frequency NB (LT) (10 min once a day) and high frequency NB (HT) treatment (10 min every 2 h). The incubated plants were analyzed for growth during 14-day period while after harvesting, they were analyzed for gene expression, hormone content and RNA sequencing. To study the need in fertilizing, in the field study, the group of control (no NBs) and one of the two groups watered with NBs (at same frequency) received the subscribed amount of fertilizer while for the other NB group received the 75% of this amount. The results of the lab experiments are shown in Fig. 13 while the findings from the field study showed that fertilizer can be reduced at least by 25% to achieve the same results regarding height, chlorophyll content etc.

The effect of micro-nanobubbles (MNBs) on drip irrigation of cucumber and tomato was compared to simple water oxidation by Liu (2019). The cultivation was conducted in a greenhouse environment and crop yield as well as fruit quality were examined for all cases. From the analysis it was found that both cucumber and tomato yield was increased by the implementation of MNBs at the same irrigation regime and fertilizer amount. In addition, water use efficiency and quality were increased greatly with the highest effect on tomato observed in soluble sugar (39.2%) and for cucumber the greater difference was in water use efficiency (22.1%).

A more recent study, that of Zhou (2022), investigated effects and mechanisms of different oxygen concentrations in NB enriched water. Crop yield of tomato, soil fertility and bacterial community were studied for two seasons irrigating with NBs water. The total oxygen concentration was the independent parameter ranging from 160 to 280 mg/L. From the obtained results it was shown that there is a threshold of oxygen concentration (indicates the entrapped O_2), for the specific crop, soil and conditions, the most efficient NB oxygen concentration was 220 mg/L while concentrations above that seem to minimize the effect.

In another study conducted by Wu (2019), tomato was used again as the examined species for evaluation of growth and productivity increase due to oxygen containing NBs. Lab scale experiments conducted in soil incubators while pilot scale was conducted in soil columns under greenhouse conditions. Except the control group, group with simple aeration was also established in order to estimate better the effect of NBs. From the results it is shown that NBs oxygenation has the highest performance compared both to aeriation and control; 5% and 23 higher, respectively. Finally, from the soil analysis, it was found that the remaining nutrient and organic content in soil from NBs is greater even than simple aeration. Overall, the study showed that simple aeration and NB utilization have similar results regarding organoleptic properties but NBs promote the growth rate for and showcase better total yield.

Ahmed (2018) also investigated the influence of NBs on seed germination and plant growth. Their study involves various gases (oxygen, air, nitrogen and carbon dioxide) on an extended species including carrot and lettuce. The NBs were generated by gas pressurization through a ceramic membrane after 90 min of gas injection at constant flow rate. The impact of reactive oxygen species (ROS) after NB treatment of tap water was essential for the purpose of this work characterized by photoluminescence technique. Germination test was conducted on lettuce, carrot and bean seed for 6–10 days while for plant growth test carrot and tomato was used in garden soil. In both sets, tap water was used for the control group. Stem and leaves were characterized to examine the effect on the organoleptic properties of tomato, carrot and bean. The study concluded that nitrogen NBs had the best performance in all stages of the plant growth. However, it was obvious that different gas affect its step differently. For example, nitrogen NBs show the highest increase in germination while carbon dioxide NBs grow more enhanced plants. Authors attribute those differences on the different NB properties regarding zeta potential and electrical interphase charging.

Similar differentiation was observed also by Khan et al. (2022) where CO_2 and Air NB were evaluated for Amaranth green cultivation. One additional parameter under investigation was the NB production time at 5, 10 and 15 min. Again, air NB seem not to have as significant effect as CO_2 NBs have both on seed germination and plant growth reaching 90% increase compared to the control group. Regarding time, interestingly for the air NBs it had negative effect on the efficiency while the opposite is observed for CO_2.

3.2. Hydroponics

Another cultivation method is hydroponics where plants are cultivated without soil, instead the roots are immerged in a water/nutrient solution. Hydroponics are characterized by low water consumption and faster growth (Sambo, 2019). Hydroponic systems can vary in applied technique; the most common are static and circulated solution culture, aeroponics (Eldridge et al., 2020) and fogponics (Rakib Uddin and Suliaman, 2021). The nutrient is important as well as its distribution to the water system. Nanobubble application in hydroponics has raised a debate regarding the positive or negative impact of NBs in such systems. However, the truth lies somewhere in between; NBs in plant growth promotion cultivated hydroponically depends on various factors. It has proven that NBs regulate DO in the solution, promote leaf and stem growth (Kobayashi and Yamaji, 2022) achieve better morphological and physiological responses compared to conventional aeriation but only under a specific threshold point above which NBs inhibit plant development (Wang et al., 2021).

Soybean seedling was examined hydroponically under different concentration of nutrient (zero, low and high level of nutrient) and to investigate the effect of NBs (in the under discussion study called ultra fine bubbles) (Iijima et al., Jul. 2020). Deionized water was used as control while N2 and air were used for NB generation. Additionally, conditions of stirring and no stirring were conducted for both control and NB solutions. Interestingly, the NB maximum contribution was observed in zero nutrient group for both stirring and no stirring/ In contrast, NBs even suppressed the high nutrient level regarding shoot/ratio of the crop where control group performed better.

Due to the controversial results of different studies regarding promotion or inhibition of hydroponic culture, the same group of Iijima et al. (2022), conducted a repetitive study adding this time 5 more species while the NB concentration was also examined. Biomass and elongation rate of soybean, cowpea, adzuki bean, wheat, rice and maize. The three first are in the cereal family and the rest three in legumes. The study concluded that there is positive effect of NBs in plant growth but there are several limitations that are more pronounced in hydroponic cultivation. This is attributed to the presence of nutrient during the plant growth and the inhibition (or neutralization) of some components when NBs passes a threshold of concentration. Another work presented at The Third International Tropical Agriculture Conference (TROPAG 2019) by Li and Cave (Li and Cave, 2019), suggests that NBs are a promising method for retaining DO levels in outdoor hydroponics especially in high temperature climates. Specifically, they examined the stability of air NBs at different nutrient solutions and pH conditions compared to NBs in distilled water up to 40 °C temperature. Other studies also investigated the interaction of NBs with the electrical conductivity (Sritiontip, 2022), the plant's nutrient uptake and total dissolved solids (Hasta Pratopo et al., 2021).

3.3. Aquaculture

Aquaculture is a sub-sector of agriculture and comprises all the activities related to breeding, raising and harvesting fish, shellfish and aquatic plants (Bostock, 2010; Tacon, 2020). The aforementioned activities are performed in open sea, rivers, lakes, artificial or natural ponds and indoor facilities (Ghobadi et al., 2021). The challenges in aquaculture depend on the purpose of use (i.e. shelfish or fish cultivation). When aquaculture refers to shrimp or oyster farming, there is a high mortality issue arisen from low quality water, diseases, feed and others (Diwan et al., 2022; Joffre et al., 2018; David et al., 2019; Botta et al., 2020). In fisheries, the main problem is fish poor health due to factors such as fish overpopulation, insufficient nutrients, infections in stationary systems etc (Wanka et al., 2018; Føre, 2018; Chu et al., 2020). Although among the first applications of NB technology was reported in oyster and shrimp cultivation by private sector and various organizations, there is lack of scientific research on that aspect.

One of the most important parameters for fish cultivation is the well oxygenated environment. Mahasri et al. (2018) studied the influence of NBs on oxygen diffusion in nile tilapia, species that requires high oxygen concentration. Dissolved oxygen (DO) decrease rate was measured with and without fishes. From the results, authors concluded that NB enhances DO level and the decrease rate is slower.

As mentioned previously, DO level plays significant role in fish good health; the better the environmental conditions the more enhanced immune system and the less bacteria cultivation. To this direction, Dien (2022) studied oxygen and ozone NBs in combination with a bacteriophage. The phage remained intact after oxygen NB treatment but eliminated by ozone ones. After that, phage adhesion on fish skin and gills were also studied. The results showed an up to ~ 5 times better phage uptake in the presence of oxygen NBs compared to the untreated group. Bacterial reduction was evaluated after treatment with ozone NBs in freshwater fish cultivation by Jhunkeaw (2021) Ozone NB treatment showed a 99.99%

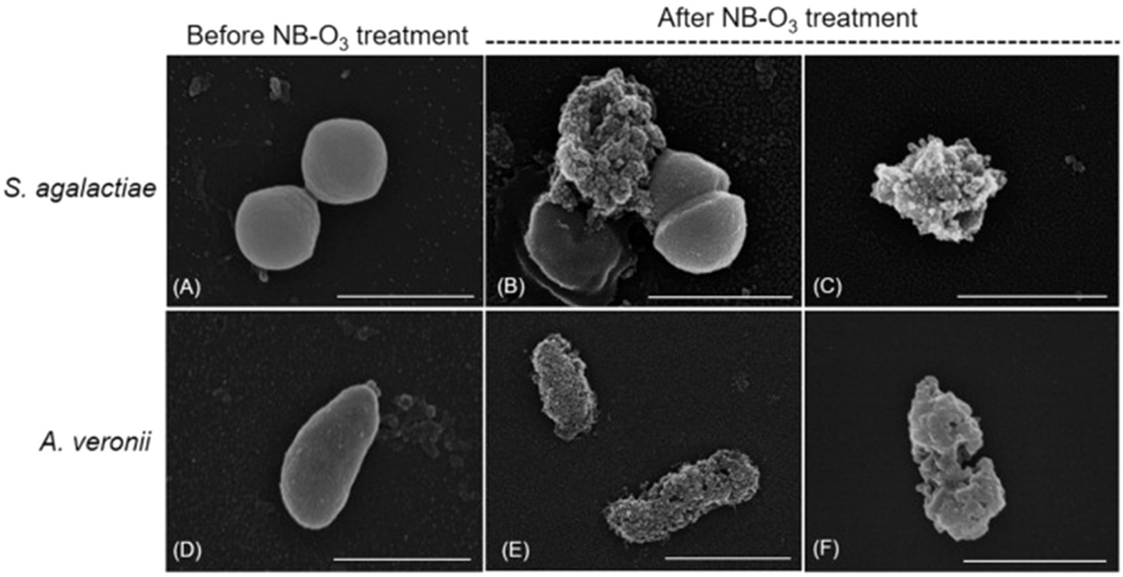

Fig. 14 – Scanning electron micrographs of S. *agalactiae* (A-C) and *A. veronii* (D-F) before and after treatment with NB-O3 for 10 min. Bacterial morphology was normal before treatment while cell destruction was observed after treatment with NB-O3. Scale bar, 1 µm.
Reprinted from (Jhunkeaw, 2021) with permission from Elsevier.

reduction of bacterial concentration by bacteria cell disruption mechanism (Fig. 14). This value was lower when organic matter was present (mucus, feces etc.). In addition, fish morphology after exposure to the treatment was examined as well. Although a short time exposure seems not to affect fish, exposure frequency alters the result.

4. Medicine and bio-medicine application

4.1. *Ultrasound Imaging*

A well-established clinical tool for the diagnosis and treatment of a variety of diseases, biomedical ultrasound (US) imaging is employed in medicine. Using sound waves over 20 kHz, US imaging produces images depending on how the sound waves interact with the surroundings. The tissue and boundaries between tissues, such as skin and bone, will reflect, scatter, or absorb the US waves produced by a transducer (1–20 MHz range). The tissue location and scattering properties may then be determined using the timing and intensity of the returning echoes (Averkiou et al., 2020). However, due to the soft tissues' closely matched acoustic impedances, US imaging can experience decreased contrast (Foley et al., 2013).

Microbubbles (MBs) consisting of a stable shell and a gas core, are employed as ultrasonic contrast agents (UCAs) because of their high impedance mismatch and substantial scattering of nearby tissue. Bubbles go through volumetric oscillations when exposed to an ultrasonic field, scattering ultrasonic energy at the same frequency as the driving US and enhancing contrast (Zahiri et al., 2021). However, microbubbles have limited clinical applicability due to their size, which causes them to stay in the vasculature (Krishna et al., 2018). Nanobubbles, on the other hand, are smaller than typical clinically used MBs and may easily cross through such tough biological barriers as the blood–brain barrier (BBB) (Deprez et al., 2021), tumor vasculature (Wu et al., 2019), and cell membrane (Jugniot et al., 2022). Thus, NBs as contrast agents can produce more detailed US visuals and also provide more precise information to accurately diagnose malignant tumors (Exner and Kolios, 2021).

Recently, targeted NBs have been developed by surface functionalization for increasing the sensitivity of the target detection. Specific targeting ligands like antibodies and biomarkers have been used to achieve tumor selectivity. These ligands enable identifying certain receptors at the target location, which improves tissue selectivity.

Gao (2017), developed CA-125 targeted nanobubble US contrast agents. Overexpression of cancer antigen CA-125 characterizes ovarian cancer. In this work, NBs surface was functionalized with CA-125 antibody. It was reported that targeted NBs exhibited increased tumor retention and prolonged echogenicity compared to untargeted NBs. Perera et al., in the same concept, created PSMA targeted NBs for real time US molecular imaging of prostate cancer. Antigen PSMA is over expressed in prostate cancer incidents. Again, it was demonstrated that tumor accumulation and tumor retention were selectively enhanced by the PSMA targeted NBs, resulting in improved prostate cancer detection and real time imaging. In a similar work, Wang (2020), constructed multimodal targeted NBs for prostate cancer by encapsulating the photoacoustic contrast agent indocyamine green (ICG) in the NB lipid shells and coupling PSMA-binding peptides to the NB surfaces. Enhanced US, photoacoustic and fluorescence imaging of PSMA-positive tumors were again achieved.

In another study, Yu (2020), based on the same principle, developed anti-G250 nanobody- functionalized targeted NBs to enhance US molecular imaging of renal cell carcinomas. Antigen G250 is expressed in renal cell carcinoma cells. It was shown that the targeted NBs managed to enter the tissue space through tumor blood vessels and bind specifically to tumor cells, enhancing the US imaging. Liu (2018), in their recent work, constructed T lymphocyte- targeted NBs to detect T lymphocyte infiltration in acute rejection after cardiac transplantation. T lymphocyte-targeted NBs (NB_{CD3}) and control NBs (NB_{CON}) were characterized with zeta potential analyzer, optical and fluorescence microscopy while flow cytometry analysis was also performed. For heart

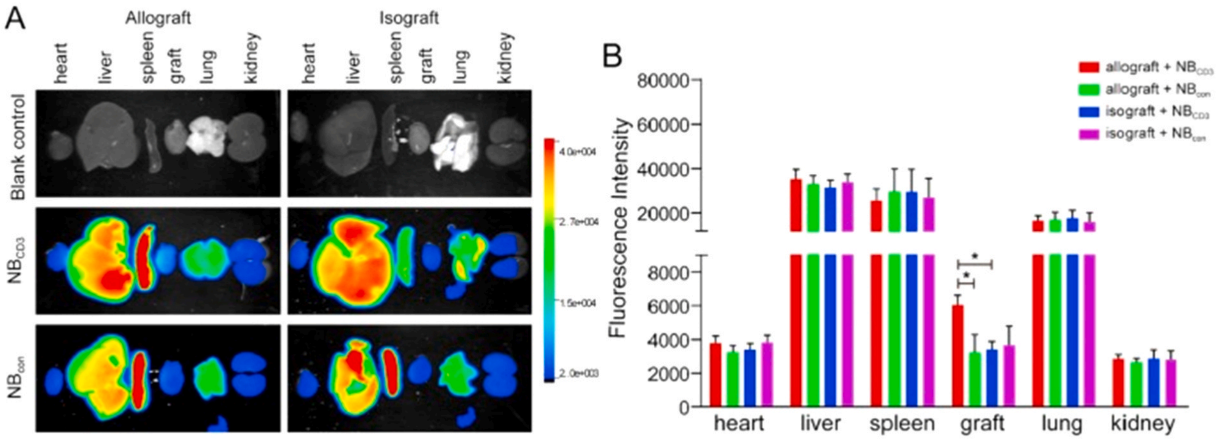

Fig. 15 – The biodistribution of the NBs in rats. (A) *Ex vivo* images of the major organs 4 min after injection of NBs. The NBs were mainly distributed to the liver, spleen and lung tissues. (B) Quantitative analysis of the fluorescence intensity in different organs. The amount of NBCD3 accumulated in the allograft heart was higher than that of the NBcon, and also higher than that of the isograft heart.
Reprinted from (Liu, 2018) with permission from Elsevier.

transplantation simulation, two species of rats were used; BN rat to Lewis rat as allograft and Lewis to Lewis to Lewis rat as isograft group. Target biodistribution was observed in *ex vivo* measurements, Fig. 15 shows the biodistribution after 4 min of NB injection. It was reported that the application of targeted NBs could be beneficial for monitoring the occurrence, progress and outcome after immunosuppressive therapy of acute rejection.

Zhang et al. (2020), in another recent publication, successfully investigated the potential use of lipid NBs in molecular imaging of atherosclerotic plaque. The results confirmed that targeted NBs conjugated with anti-VEGFR-2 ligands could facilitate site-specific recognition of atherosclerosis using ultrasound imaging.

4.2. *Drug/Gene delivery for cancer therapy*

Chemotherapy is one of the primary treatment options for cancer malignant tumors. However, the majority of chemotherapeutic medications target the tissues non-selectively, which has negative side effects on healthy tissues (Bellotti, 2021). Targeted nanobubbles can be used as ultrasound-triggered drug delivery systems for tumor specific targeting. Due to their small size, extravasation from blood vessels into the surrounding tissues is feasible (Cooley et al., 2023). They also have increased stability and prolonged retention time in systemic circulation. Targeted nanobubbles are surface functionalized with certain targeting ligands which allow adsorbing to specific cancer cells and also preloaded with chemotherapeutic drugs or genes (de Leon et al., 2018) (Fig. 16).

Moreover, they collapse under high intensity US acoustic pressure and efficiently release their content to cancer cells. Additionally, collapsing NBs create cell membrane pores by intense jet streaming, a phenomenon known as “sonoporation” which increases cell permeability (Nishimura et al., 2021).

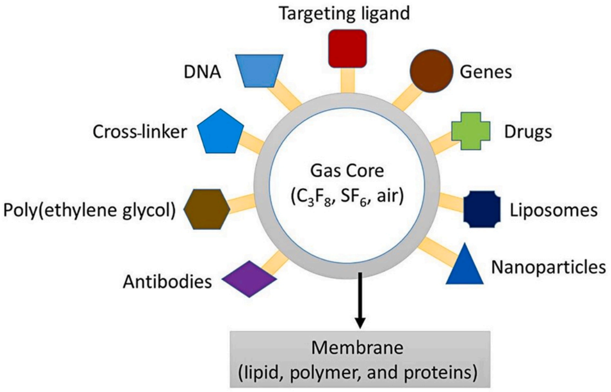

Fig. 16 – Schematic representation of possible MBs and NBs mechanism as ultra sound agents.
Reprinted from (de Leon et al., 2018) with permission from Elsevier.

Drug or gene carrier NBs consist of two main components: outer shell and inner core. The outer shell forms a protective layer around the gas core which reduces the gas diffusion. The shell usually consists of surfactants, lipids, polymers or proteins. The inner gas core can contain oxygen or other insoluble gases such as perfluorocarbons (PFC) and sulfur hexafluoride (SF6) for increased stability (Lu et al., 2022). The gas core and the outer shell also determine the acoustic response of NBs in US fields (Zhang et al., 2021). Drugs may be encapsulated in the core of the nanobubbles, within the shell of the nanobubbles, or just under the shell. A different method of loading the medicine is by encapsulating it in a nanoparticle and then adhering it to the surface of the bubble (Raikwar, 2021).

Wu (2018), developed targeted FoxM1 siRNA-loaded cationic NBs for gene therapy of prostate cancer. It was shown that the produced NBs combined with US mediated nanobubble destruction can effectively inhibit tumor growth. Bessone (2019), synthesized curcumin loaded polymeric NBs for prostate cancer treatment. It was reported that this therapy successfully prevented metastatic spreading in prostate cancer cells.

In another work, Shen (2020), constructed AMD070 and ICG-carrying NBs for breast cancer therapy. These targeted NBs successfully lead to tumor cell apoptosis. Zhang et al. (2018), developed a US-NB gene delivery platform for the treatment of hepatocellular carcinoma. Again, it was confirmed that the therapy inhibited the growth of cancer cells

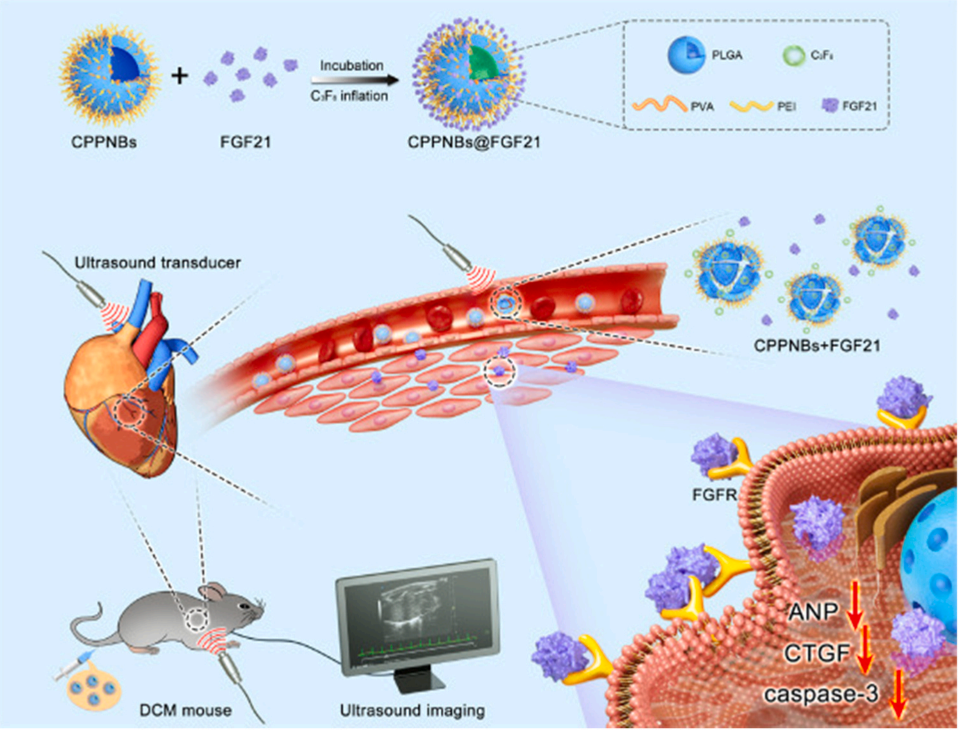

Fig. 17 – Schematic illustration of the fabrication and mechanism of CPPNBs@FGF21 coupled with low-frequency ultrasound in diabetic cardiomyopathy (DCM) therapy.
Reprinted from (Gao, 2021) with permission from Elsevier.

and induced cancer cell apoptosis. In a recent work, Chan et al. (2020), synthesized a micelle-type NB decorated with fluorescein-5- isothiocyanate-conjugated transferrin, with encapsulation of paclitaxel for lung cancer treatment. This method selectively targeted lung cancer cells and significantly inhibited tumor growth via paclitaxel release.

Recently, NB enhanced US imaging and NB targeted drug delivery have been combined in one diagnostic and therapy approach known as “theranostics” (Huang et al., 2020) for effective real time imaging- guided cancer therapy. Li et al. (2017), developed glycine/PEG/RGD-modified NBs loaded with doxorubicin (DOX) as an anti-cancer drug and perfluorohexane (PHF) as an ultrasound probe. This technique exhibited high quality US imaging and significant cancer therapeutic efficacy. In another work, Prabhakar and Banerjee (2019), successfully prepared NB-paclitaxel liposome complexes for US imaging and US triggered drug delivery in cancer cells. In this study, modified NBs exhibited great echogenic stability and enhanced cellular permeability performing great anticancer efficacy as a theranostic platform.

4.3. *Hypoxia treatment*

Most solid tumors exhibit hypoxia, which is typically a result of poor vascular density, irregular vascular geometry, and an imbalance between oxygen supply and demand at the tumor site. When HIF-1a (hypoxia-inducible factor 1a) is over-expressed in these hypoxic areas, angiogenesis and metastasis are induced (J. M. B. T.-M and Brown, 2007). Any cancer treatments that can be applied are usually hindered by this. Radiotherapy, chemotherapy, and other oxygen-consuming treatments like photodynamic therapy are less effective on hypoxic cells. To increase the effectiveness of present cancer treatments, a solution must be developed to mitigate this circumstance for lowering tumor hypoxia. There are two main methods for oxygenating tumors: either enhancing blood oxygenation broadly or improving oxygen delivery at the tumor site. Oxygen NBs can be applied to reverse the hypoxic state and provide more oxygen to cancerous tissue (Gao, 2021; Qiao 2022). The two primary strategies for oxygen delivery using MNBs is (i) injecting MNBs intravenously, followed by the use of high- intensity ultrasound to break the MNBs (Khan, 2018) and (ii) to permit oxygen diffusion along the concentration gradient (Bhandari et al., 2017).

Iijima (2018), experimentally studied the potential use of oxygen NB water, without any additives, against cancer cell hypoxia. The process was evaluated by measuring HIF-1a levels in breast and lung cancer cells. It was reported that oxygen NB water proved to be a promising agent in overcoming the hypoxia induced resistance of cancer cells to radiation. Khan (2019), developed lipid-shelled oxygen NBs by employing a sonication method to mitigate hypoxic conditions generated in a specially designed hypoxic chamber. A significant decrease in the expression of HIF-1a on breast cancer cells was observed, indicating that oxygen NBs were able to achieve the reversal of hypoxic conditions. Song (2020), synthesized protein-shelled oxygen carrier NBs, surface-modified by a liposome layer to increase their stability and half- life. In this study, a substantial improvement of oxygen concentration in hypoxic tumors and enhanced photodynamic therapy efficacy were observed. In another work, Bhandari et al. (2018), produced oxygen encapsulated carboxymethyl cellulosic NBs to inhibit tumor hypoxia and growth. Again, it was confirmed that oxygen NBs successfully delayed tumor progression and improved survival rates in mice models.

4.4. *Other biomedical applications*

Due to its unique properties, NBs are suitable for use in a variety of other therapeutic applications such as in the

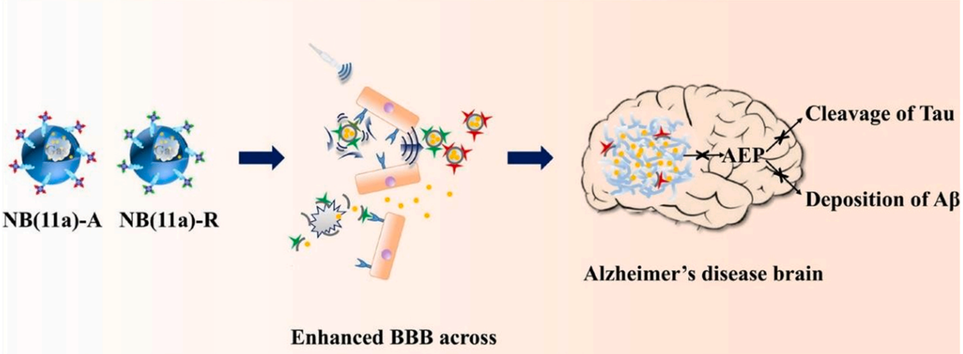

Fig. 18 – Graphical abstract of "Asparagine endopeptidase-targeted Ultrasound-responsive Nanobubbles Alleviate Tau Cleavage and Amyloid-β Deposition in an Alzheimer's Disease Model" by Mi et al.
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treatment of diabetes, atherosclerosis, thromboembolic diseases, Parkinson's disease and Alzheimer's disease. Gao (2021), synthesized perfluoropropane (C3F8) and polyethylenimine (PEI)-doped poly (lactic-co-glycolic acid) (PLGA) nanobubbles in order to improve the therapeutic efficacy of fibroblast growth factor 21 (FGF21) in diabetic cardiomyopathy (DCM); Fig. 17 illustrates the mechanism. The results of this study indicated that the application of the formulated NBs successfully prevented the myocardial hypertrophy, apoptosis and interstitial fibrosis of DMC mice. LaMour et al. (2021) developed a topically-applied hydrogel emulsion consisting of oxygen and carbon dioxide NBs for the symptomatic treatment of diabetic peripheral neuropathy (DPN). It was reported that there was significant relief of DPN pain symptoms, after the NB hydrogel emulsion application.

In another study, Li et al. (2022), experimentally investigated the effect of NBs on the reduction of the size of the atherosclerotic plaque. An alternating magnetic field NB generator was used to enrich Ringer's solution with NBs. The results confirmed that plaque volume significantly reduced in the presence of NBs. Ma (2020), developed cRGD-targeted nanobubbles to enhance the dissolution of thrombus in human vascular thromboembolism. It was reported that higher thrombolysis rates were observed indicating significant thrombolytic efficiency.

In a recent study, Yan (2021), fabricated curcumin-loaded lipid-PLGA NBs for drug delivery through blood-brain barrier (BBB) to treat Parkinson's Disease (PD). The results showed that the combination of the formulated NBs with low-intensity focused ultrasound successfully locally delivered curcumin into diseased brain regions in the Parkinson mice model. Mi (2022), in a novel study, developed US-responsive NBs loaded with RR-11a inhibitor against asparagine endopeptidase (AEP), for targeted intracerebral Alzheimer's disease (AD) treatment (Fig. 18). The results indicated successful drug delivery through BBB to the AD lesion and restored cognitive function of Alzheimer mice model.

Another field of biomedical NB application is dermatology. Nanobubbles have been reported to have anti-inflammatory (Yoshida et al., Oct. 2020), antimicrobial (Shawli et al., 2020), sterilization (Horiuchi, 2021) and healing (Gupta and Shende, 2022) properties, therefore their application for various skin diseases seems very promising. An interesting study of Horiuchi (Horiuchi, Nov. 2020) suggests ozone NBs for palmoplantar pustulosis treatment applied as an oral rinsing on periodontal area. The conducted study involved 6 patients to whom a two-minute rinsing every night was prescribed. After 3–4 months of application the patients had visible results in skin lesion suppression. Another reported application is for topical oxygenation therapy for wound healing (Sayadi, Jun. 2018). Ischaemic wound healing by oxygen NB as topical oxygenation therapy was investigated by Aoki et al (2022). Two groups of rats were examined; one as a wound healing model group and the other with ischeamia. To the latter group, additional wound was created. For the group of wound healing model, no differences were observed between O_2NBs, purified water and saline solution. However, for the case of ischeamic wounds, O_2NBs showed a healing time decrease by 5 days compared to other two treatments.

Other dermatological applications of NBs have been reported by Sayadi (2021, 2019) for burn injuries healing where healing time was significantly reduced while ischeamia prevention was suggested, vapor NBs were used for better diffusion of antimicrobial agents through biofilms promoting wound care (Teirlinck, 2019), ozone NBs in saline solution for dental and anal fistulae (Horiuchi, 2021) and more.

5. Other industrial and non-industrial applications

Nanobubbles have a very wide range of application due to different properties according to the trapped gas, production conditions, solution environment etc. In some cases, NB technology can be considered mature (i.e. water and wastewater treatment). However, there are applications that have emerged only recently such as heavy industry, domestic and personal care products.

An example of NB application in heavy industry is scale formation inhibition and/or treatment. Aikawa et al. (2021) examined NB inhibition on low-carbon steel. Air NBs inhibited corrosion by 50% compared to conventional methods. This study suggest that NB can serve both as cleaning and coating different metallic surfaces preventing corrosion from fluids of various chemical compositions. Similarly, Tagomori (2022), propose air NBs for retarding calcite crystal growth on a substrate. Although the effect decreases as temperature is increased; minimum NB efficiency is 33% at 88 °C while the optimal temperature indicated to be 20 °C where an impressive 53% was obtained.

Harsh environments and operation conditions affect industrial equipment. Brine is one of the most challenging fluid to treat while salts are deposit on any surface coming in contact. An indicative example is membrane, where brine clogs the pores and reduces the efficiency. Farid et al. (2022) conducted a study of scaling inhibition on membrane distillation (MD) of high salinity water. The results showed a significant improvement of MD process retarding salt deposition and prolonging membrane lifetime for more than 100 h; without NB assistance MD was totally clogged after 13 h of operation while with NBs the performance was reduced at 63% after 98 h.

Nanobubbles utilization in geothermal energy systems reduces heat capacity and promotes heat transfer. Soltani et al. (2022) reviewed NB application in energy production by geothermal implementation and concluded that among others, viscosity, volume fraction, density, specific heat capacity, mass flow rate, nanoparticle shape and size, Brownian motion, pressure drop, and friction factor are the parameters affecting the geothermal system efficiency. To this end, NBs improve the performance attributed to the need of lower volume fraction of suspended nanoparticles.

In food industry, NBs can be utilized for fruit and vegetable preservation, efficient cleaning and viscosity reduction in produced liquids (i.e. juices) (Yang and Chen, 2022). Babu and Amamcharla (Babu and Amamcharla, 2022) applied NBs in dairy spray drying process. One of their work objectives was to examine the effect of MNBs on microstructural and functional properties of milk protein concentrates. Rheological measurements showed that MNB treatment reduced viscosity by 65% due to disruption of large aggregates while overall microstructure differentiated from the control samples. Lastly, rehydration test indicated that MNB treated milk protein concentrates were rehydrated easier and more efficiently.

Nanobubble assistance in Fuel performance has been also investigated. Senthilkumar et al. (2022) used air NB mixed with hone oil biodiesel and TiO2 additives. The performance was analyzed regarding break-specific fuel consumption (BSFC), break-thermal efficiency (BTE) and exhaust emission. The presence of NBs reduced BSFC by 15%, enhanced BTE by 3.1% and reduced emissions of hydrocarbons, carbon monoxide and nitrogen oxides (15%, 19% and 14%, respectively). In another study, NBs were introduced in gasoline fuel and the performance was estimated theoretically (Sharif, 2019). Again, NBs reduced emissions while enhanced the performance of the engine due to sufficient oxygen content in fuel/air mixture.

Hydrogen NBs were used for wrinkle reduction studies (Tanaka and Miwa, 2022). Five individuals from 49 to 66 years old were subjected to facial treatment of a towel soaked in H_2 NB solution for 11–98 days. The results indicate that NB are promising for anti ageing and skin care products. Other reports also support this outcome, claiming that NBs increase skin permeability of nutrient compounds in skin products.

6. Conclusions

This work provides a thorough literature review of the most recent developments in NB technology and its application. The reviewed aspects include environment with focus on water and wastewater treatment by NBs, agricultural sector divided in (i) hericulture, (ii) hydroponics and (iii) aquaculture, medical and bio-medical sector where studies regarding theranostics and drug delivery are mainly presented. Other sectors such as energy and intense industrial processes were also discussed briefly. The review was limited only to research articles since 2017 up to date. A search in Scopus database using “nanobubble technology” as keyword and restricted to research articles, returns back 1843 results with the first published work reported in 1999. When the year of publication is set to the range from 2017 to 2023, the total of articles is 1154; more than 60% of the ever published research on NB technology. Those illustrative results outline that although NB technology can be considered as the horizon of many challenges to overcome, it can be claimed that it is in immature stage yet. Therefore, a continuous monitoring of NB development on every sector is critical in better understanding of this technology and the mechanisms involved.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.cherd.2022.11.013.

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