

Review

Review of Micro- and Nanobubble Technologies: Advancements in Theory and Applications and Perspectives on Adsorption Cooling and Desalination Systems

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Abstract: Adsorption refrigerators are a compelling ecological alternative to compressor refrigerators; global warming forces us to constantly look for alternative sources of energy and cold. Cold production in adsorption chillers is based on the use of heat generated by other processes running in the company. Waste heat from production processes, which has, until now, been irretrievably lost, is a potential source of energy for generating cold via an adsorption unit producing chilled water. Cooling optimizes the use of the heating network in summer and can lead to increased electricity production while reducing heat supply losses. Thus far, attempts to implement adsorption refrigerators for widespread use have not been successful as a result of the low efficiency of these devices; this is directly related to the poor heat and mass transfer conditions in the beds and heat exchangers of adsorption refrigerators. The solutions used so far, such as new working pairs, glued beds or modifications to the structure or cycle length, are still not strong enough for these devices. Therefore, it is necessary to look for new solutions. Using micro- and nanobubbles as media to increase mass and heat transfer in refrigerators is an innovative and pioneering solution. Thus, this document describes the most important features of micro- and nanobubble technology applications in adsorption refrigerators. This article is an introduction and a basis for the implementation of further research, consolidating the existing literature as a review.

Keywords: adsorption cooling and desalination systems; heat transfer; mass transfer; nanobubbles; microbubbles; nanobubble stability; sustainability; energy efficiency; net-zero emission



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1. Introduction

Over the last few decades since the first industrial implementation of fluidization, it has found wide application in various fields of science. Fluidization is used in both physical and chemical processes. Examples of fluidized physical processes are transporting, drying and separating bulk materials and numerous applications in heat exchange. Chemical processes carried out in the fluidized bed can be divided into processes for processing material grains (e.g., gasification, combustion of fuels) and gas processing processes (e.g., production of acetylene, ammonia) [1,2].

Among many technologies used in industry, solid fluidization is distinguished by its good development of the contact surface of the solid and liquid phases and by its mixing of the components of both phases. This ensures an equal temperature throughout the suspension and very good heat, mass and momentum exchange between the grains and the gas. These features make the fluidization process used in many chemical and food industries [3,4]. Fluidized bed devices are also used as heat exchangers in fluidized bed boilers and food freezers [5,6].

The idea of bubble technology dates to the 1950s, when the Epstein–Plesset theory was developed to predict the lifetime of a single bubble as a function of its radius and saturation [7]. Over time, the technology and knowledge regarding nanobubbles (NBs) have continued to evolve and they are now gaining more and more applications, such as in water treatment [8], agriculture [9] and drug delivery [10]. Due to their unique properties, such as large specific area, high mass transfer efficiency and low rising velocity in liquid, nanobubbles have become popular research objects [11–14]. A model presented in [15] shows promising results for the use of cavitation to enhance heat transfer, as cavitation can thin the thermal boundary layer, reduce thermal resistance and improve heat transfer. The authors also presented that the cavity bubbles generated a stronger turbulence flow, significantly thinning the flow and viscous bottom layers. Acoustic cavitation was also used to improve the performance of CPU water-cooling systems [16]. The authors of [15–19] agreed that using cavitation to reduce the boundary layer in heat exchangers effectively enhances heat transfer. These studies have shown that using bubbles in fluids could be suitable for intensifying heat transfer processes. Therefore, the papers mentioned above suggested the idea of using micro/nanobubbles to enhance heat transfer in adsorption chillers. While using nanobubbles to improve the efficiency of adsorption chillers has not been proposed in the literature, it is a promising and innovative approach that still requires research and empirical knowledge. However, this exciting concept can contribute to developing adsorption cooling and desalination systems, particularly heat transfer between the adsorption bed and heat exchanger. The discussed issues fit into implementing a net-zero emission strategy, as adsorption cooling and desalination systems adsorption chillers are in line with achieving zero emissions. Improving heat and mass transfer conditions increases the energy efficiency of cooling and desalination systems. Micro/nanobubble technologies have been used in many different industries and are also emerging in the context of heat and mass transfer. According to the literature review, there are no existing applications of bubble technology in adsorption and desalination systems. Therefore, we decided to prepare this short review as an introduction to future work on the implementation of micro- and nanobubble technology in adsorption refrigeration and desalination systems, which are innovative solutions.

2. Nanobubbles: Explanation and Properties

Over the last twenty years, there has been great interest in nanoscopic bubbles from both academia and industry. Industry interest stems from the idea that dynamically stable and small bubbles will avoid typical buoyancy collapse, enabling them to intervene in applications ranging from water treatment, lake remediation, foam production, cleaning of contaminated surfaces and disease treatment [13]. The term nanobubbles covers at least two types of bubbles: the first is surface nanobubbles and the second is bulk nanobubbles. Surface nanobubbles are gas-filled pockets/sacs on surfaces that form spherical caps. Surface nanobubbles have spherical shapes with a height of 10 to 100 nm and the diameter of their contact lines ranges from 50 to 500 nm. Bulk nanobubbles, on the other hand, are described as spherical cavities filled with gas, obtained in the liquid phase, with a diameter of less than 1000 nm. The term nanobubbles often refers to micropancakes that are a few nanometers high and stretch the sides up to several micrometers, but their gaseous state is a debatable issue [20–22].

The curvature of the nanobubble contact surface influences the internal Laplace pressure. This pressure increases the solubility of the gas in the surrounding solution, which leads to the bubbles shrinking by diffusing the gas into the solution. According to calculations, the lifetime of a nanobubble should be much shorter than a second, which is 3–5 orders of magnitude shorter than the observed lifetime [23]. Therefore, additional theories regarding the stability of nanobubbles have been proposed. One theory is the presence of a layer of impurities that may inhibit gas transport or cause surface pressure that counteracts surface tension. However, studies have shown that in most cases such

contaminants are not present and bubble stability is maintained even after the addition of a surfactant to remove any contaminants [23–25]

Bubbles are thermodynamically unstable structures due to the energy cost of the interface. For traditional bubbles (i.e., MBs), there are three main causes of instability: thermodynamic instability, buoyancy failure and coalescence [26]. The total interfacial area of bubbles decreases as they fuse on a surface or with another bubble. The paper presents the relationship between wettability and surface energy, which, according to the Young–Laplace equation, causes a decrease in capillary forces [27].

Reducing the surface area is also achieved by compressing the gas inside the bubble, ultimately reducing bubble size. However, in the case of a bubble, a decrease in size is always accompanied by an increase in pressure inside the bubble [13]. The relation between the pressure within a bubble and its immediate surroundings is described by the Young–Laplace equation:

$$\Delta P = \frac{2\gamma}{r}, \quad (1)$$

where γ is the surface tension for the bubble interface and r is the bubble's radius.

The authors of [28] suggest that Laplace pressure plays an important role in analyses regarding the stability of nanobubbles. They posit that bubbles are unstable in size unless the gas inside them is in equilibrium with the gas dissolved in the solution. If this balance is maintained, the follicle can be expected to maintain a constant size. Laplace's equation describes the increase in pressure inside the bubble compared to the immediate surroundings. However, the solubility of a gas in solution increases with pressure, which means that Laplace pressure increases the solubility. The smaller the bubble, the greater the solubility of the gas. For the gas in the bubble to be in equilibrium with the gas dissolved in the solution, the solution must be supersaturated relative to the external pressure. Therefore, a bulk bubble can only be in equilibrium if the solution surrounding it is not in equilibrium. It is impossible for both to be in balance at the same time. The authors of [28] consider a single bubble in equilibrium with an infinitely larger reservoir of solution surrounding it, with a small perturbation resulting in the bubble becoming marginally larger. This will reduce the Laplace pressure and the solubility of the gas in the solution surrounding the bubble. In response, more gas will enter the bubble. However, this increases the size of the bubble and thus further reduces the Laplace pressure and the solubility of the gas in the solution surrounding the bubble. A positive feedback loop is created in which the bubble grows and can never reach equilibrium with the reservoir. Conversely, if we choose an initial imbalance such that the bubble becomes marginally smaller, then the solubility of the gas in the solution surrounding the bubble increases. Thus, even if equilibrium is initially established between the bubble and the reservoir, a slight disturbance will destroy the equilibrium and cause the bubble to either become very large and be removed from solution by buoyancy or to cease to exist. The authors suggest the name “Laplace's pressure bubble catastrophe”; according to them, this is of key importance when considering the thermodynamic stability of bubbles. From a practical point of view, this issue is important as it plays a significant role in the mass transfer intensification. A detailed consideration of the solid–liquid interface can be found in [29].

According to Henry's law, a decrease in the size of the bubble leads to a decrease in the gas concentration in its immediate vicinity, which in turn causes the gas to diffuse from the solution into the bubble and create a feedback loop. Similarly, when a bubble bursts, its volume initially decreases, which increases the gas concentration around it, leading to a greater influx of gas and further expansion of the bubble. This phenomenon is important in the context of the stability and control of micro/nanobubbles because even small disturbances can lead to significant changes in their size and properties [30,31].

2.1. The Issue of Micropancakes

Micropancakes have been considered a form of nanobubble for a long time. In the 1990s [32], the beginning of the period of consideration of the existence of nanobubbles and of research into their properties within the world of science, there was a distinguished group of researchers convinced of the ease of producing surface-attached nanobubbles [20]. However, during routine research, the authors of [33] discovered that what was previously considered certain may turn out to be contradictory. Until then, it was assumed that all bubbles on the surface were made of gas, including micropancakes, but the research results showed what were originally considered to be surface nanobubbles have the so-called shape of micropancakes. Residue from lubricants may be found on the barrels of the disposable syringes used in nanobubble research. In response to this information, research was carried out confirming the thesis that micropancakes can be created from the remains of solvents used to produce nanobubbles [22]. This information cast a shadow over previous research on nanobubbles, which used disposable equipment rather than Teflon or glass. Quoting the authors of [20], "...two of the authors of this review are physicists who learned some painful lessons about careful experimental preparation". These studies indicated the need to create procedures that can maintain the highest possible purity of research samples, in order to avoid repeating such mistakes in the future. It is worth mentioning that not all works published before [33] are burdened with false data and some of them have stood the test of time well; however, the information indicates the need to be careful in interpreting the results regarding surface nanobubbles.

2.2. Zeta Potential

Zeta potential is defined as the electric potential at the interface of the slip plane of the NB electrical double layer. A higher zeta potential increases the stability of nanobubbles in suspension due to the repulsion between the bubbles. Reduced zeta potential results in less stability and coagulation. The zeta potential of bulk NB suspensions is a function of electrolytes, surfactants in solution, pH and gas type. Nanobubbles have been reported to be negatively charged at pH values from 4 to 12 and zeta potential values from -4.3 to -62 mV, respectively. At neutral pH, the measured zetapotential values were between -20 and -30 mV [34–36].

In [37], the author writes about the influence of zeta potential on the behavior of nanobubbles. Takahashi et al. [38] reported that the negative zeta potential increased as pH increased, and that the zeta potential values were positive for acidic solutions with a pH below 4.5. Therefore, increasing the pH of the solution with a high concentration of OH^- ions will increase the zeta potential. Moreover, it will increase the hydrogen bonds around the bubbles and help increase the stability of the bubbles. Based on the experimental results, the lowest zeta potential value was obtained for a solution with a pH of 4. Therefore, the reduction in the negative zeta potential can be easily attributed to the high concentration of H^+ ions in the solution or the reduction in the concentration of OH^- ions. The results of tests conducted in [37] showed that stable bubbles were formed both at neutral solution pH and at solution pH above 7. Although the nanobubbles in high pH NaOH solutions showed a highly negative zeta potential value at the time of generation, it quickly decreased to values close to the zeta potential of nanobubbles generated at neutral solution pH.

The results also showed that nanobubbles in acidic solutions were difficult to produce and zeta potential values were usually positive. This may indicate that the surface charge of nanobubbles is strongly related to the concentration of OH^- ions.

2.3. Stability of Nanobubbles

Both SNB and BNB have excellent stability and long holding times. They do not tend to combine or dissolve. In the case of mass formations of nanobubbles, the solubility of gases in water is higher; these systems show very high zeta potential values. Systems containing nanobubbles also have low buoyancy, which contributes to the long-term stability of BNB [39]. Surface nanobubbles are easier to detect compared to bulk nanobubbles.

Atomic force microscopy is mainly used to identify them. Bulk nanobubbles are evenly dispersed in solutions and do not tend to stick to the surface. However, their small size, low number density, homogeneity and high degree of dispersion make it difficult to detect and distinguish bulk nanobubbles from other dispersed nanostructures or contaminants. A high number of OH^- ions can reduce the value of the zeta potential and generate smaller bubbles in solutions with higher pH levels. The density of the bubbles per unit volume of fluid is also noted as a factor to consider in preventing bubble aggregation [40].

Bulk nanobubbles are small, spherical gas cavities that are transferred into a liquid solution. Since the pressure inside bubbles is inversely proportional to their diameter, nanobubbles should have extremely high internal pressure according to Laplace's equation. Tan et al. [40] described a model explaining the long-term viability of bulk nanobubbles based on experimental studies. Their study showed that submicron-sized vesicles are stable over a wide range of Debye lengths in both concentrated electrolytes and deionized water. The authors proposed a modified Epstein–Plesset model that takes into account electrostatic pressure. Assuming that the charge is conserved in the initially contracting bubble, this increases the charge density at the surface, increasing the electrostatic pressure. This is because the chemical equilibrium between the gas–water interface and the mass of the curved bubble is different compared to the flat phase surface. On the submicron scale, the chemical potential of charged ions at the curved contact surface of spherical bubbles is lower adjacent to this surface, which allows for a higher charge concentration compared to a flat surface, which in turn leads to a significant accumulation of zeta potential. As a result, the charge accumulation mechanism at the interface significantly improves the stability of bulk nanobubbles.

However, there are some inconsistencies in this argument. The curvature at the nanoscale is not large enough to cause such significant differences as described in the presented model [13].

Xiao et al. [41] observed an increase in the concentration of generated nanobubbles after introducing gas into solutions with the addition of nanoparticles. The prior addition of titanium dioxide nanoparticles significantly increased the concentration of nanobubbles. This is because heterogeneous nucleation during the generation process is much easier than homogeneous nucleation. Additionally, the presence of titanium dioxide nanoparticles in the solution promotes the stability of the nanobubbles [41]. Moreover, to generate massive nanobubbles, Olszok et al. [42] injected filtered compressed air into colloidal suspensions of platinum nanoparticles and polystyrene beads. The study showed that the nanobubble formation process is much more effective in the presence of nanoparticles, compared to a background bubble solution without nanoparticles [42].

Surface Nanobubbles

The proposed aspects of nanobubble stability are not the only parameters that can be regarded as crucial for achieving optimal stability. The authors of [43] presented an experiment to observe nanobubbles in various temperature regimes. It was found that the size of the nanobubbles changed with temperature. The growth or shrinkage of bubbles was independent of their size and the environment in which they stayed; namely, growths were strongly dependent on the proximity of other bubbles. Moreover, the authors put forward a thesis regarding the potential thermodynamic equilibrium of the bubbles; this was confirmed by the reaction to slow adiabatic temperature changes according to the Kelvin equation. Figures 1–5 show the properties of nanobubbles in different temperature regimes.

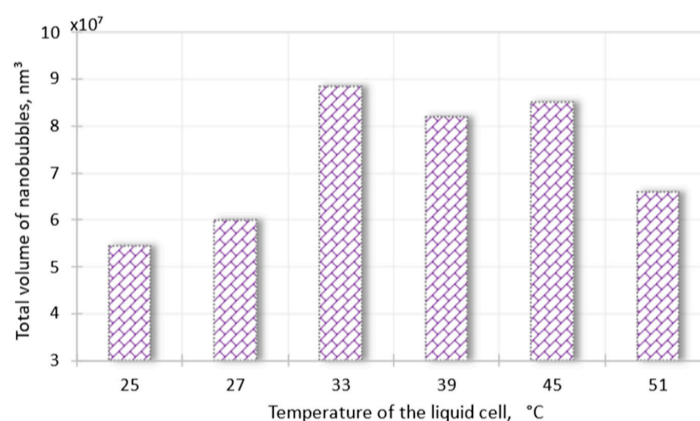


Figure 1. The overall nanobubble content at various temperature levels. The nanobubble volume grows when the temperature is lowered to 45 $^{\circ}\text{C}$ and decreases as the temperature is further reduced to 27 $^{\circ}\text{C}$, reaching its peak at 33 $^{\circ}\text{C}$.

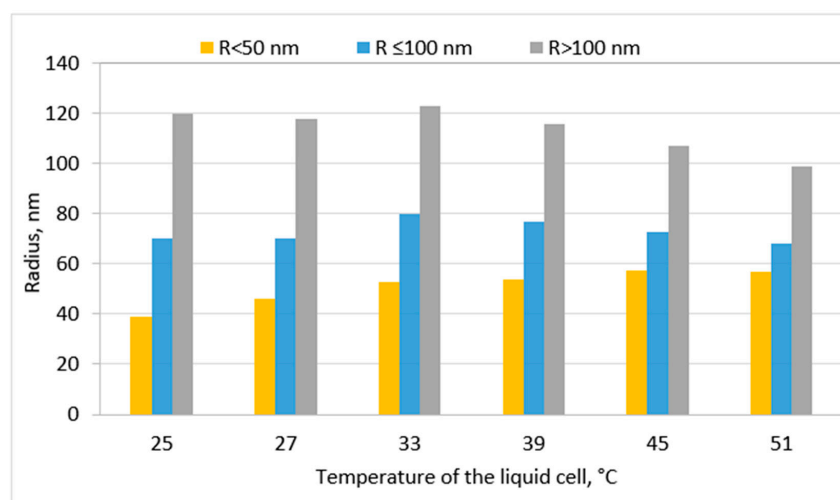


Figure 2. Temperature dependence of the nanobubble radius of the tracked nanobubbles as the temperature decreased from 51 $^{\circ}\text{C}$ to 25 $^{\circ}\text{C}$.

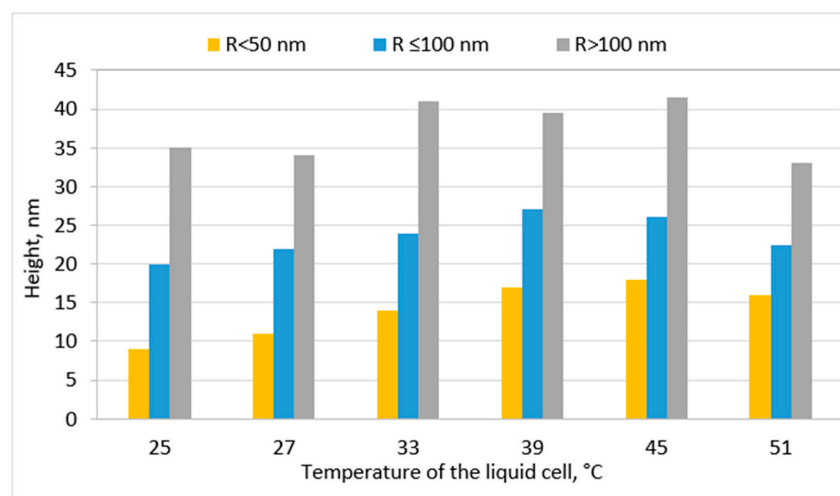


Figure 3. Temperature dependence of the height of the tracked nanobubbles as the temperature decreased from 51 $^{\circ}\text{C}$ to 25 $^{\circ}\text{C}$.

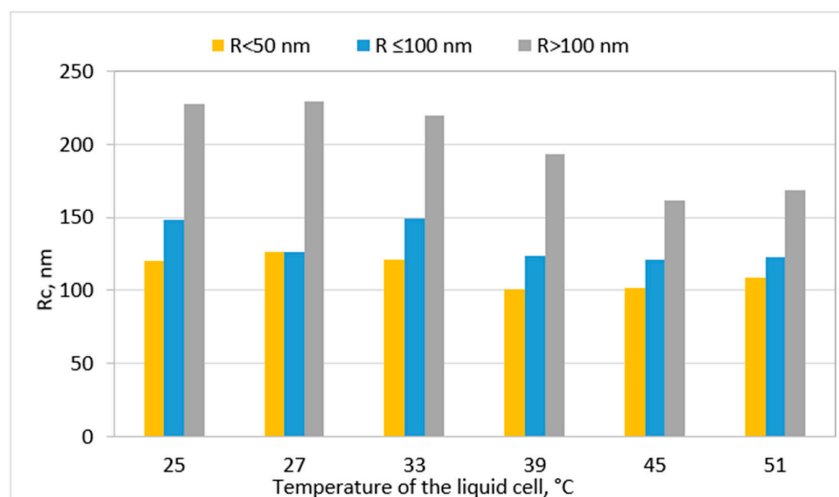


Figure 4. Temperature dependence of the radius of curvature of the tracked nanobubbles as the temperature decreased from 51 °C to 25 °C.

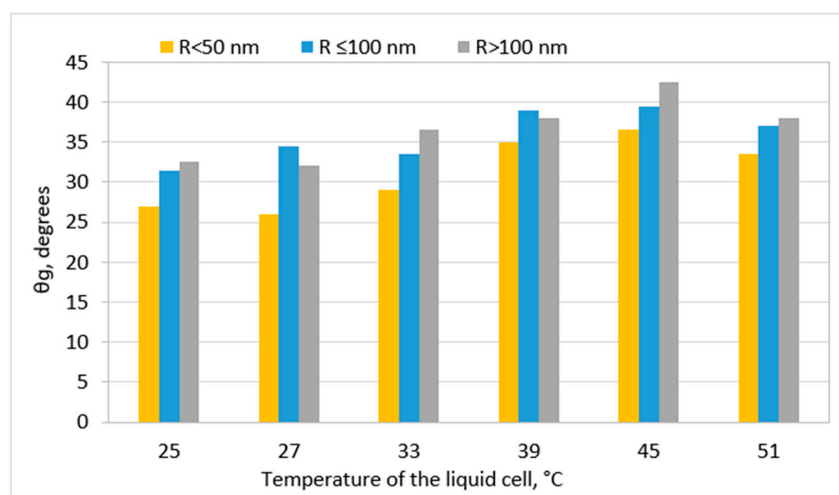


Figure 5. Temperature dependence of the contact angle of the tracked nanobubbles as the temperature decreased from 51 °C to 25 °C.

It is common practice to use atomic force microscopy when measuring a contact angle. Nanobubbles, due to their microscopic scale, require advanced measurement techniques; atomic force microscopy is ideally suited in this context. This method not only allows the imaging of the structure of nanobubbles with nanometer resolution, but also the measurement of intermolecular forces on their surface. This, in turn, allows for precise determination of the contact angle, which is crucial to understanding the properties of wet surfaces at the nano level. This took place in the cited study [44]. Moreover, the main fractions of contamination were organic compounds originating from the packaging in which the cantilevers were stored. At the same time, the study showed that sample contaminants can significantly increase the contact angle at the nanoscale.

Sun et al. [45] cite many theories and models regarding nanobubble stability, such as the contamination (impurity) theory, dynamic equilibrium theory, Knudsen gas theory, modified dynamic equilibrium theory, contact line pinning effect and nanobubbles internal pressure theory. However, none of them answer all the questions relating to the existence and stability of nanobubbles [45]. According to classical thermodynamic theories, nanoscale bubbles in water should dissolve rapidly, but many research results in recent years indicate that nanobubbles can exist stably at the solid–liquid interface. One of the theories that partially answers the question about the stability of nanobubbles is the impurity theory. It

states that the stability of the bubbles results from the presence of a layer of impurities at the gas–liquid interface, which causes a decrease in the surface tension of the nanobubbles and a decrease in the contact angle of the gas phase.

In this way, the internal pressure of the nanobubbles, i.e., the driving force for gas diffusion, is reduced. Moreover, a layer of insoluble impurities hinders the diffusion of gas inside the nanobubbles, consequently increasing the nanobubbles' stability.

The dependence of the contact angle and Laplace pressure on the pollutant coverage fraction at the liquid–gas interface was analyzed in [44]. The tests showed that the contact angle of the gas phase and the internal pressure decreased significantly with the increase in contamination coverage, which was consistent with the contamination theory. However, it was suggested in [46] that the pressure drop might be too small to explain the exceptional stability. It is also believed that the presence of impurities can reduce the surface tension at the gas–liquid interface, but this does not affect the time scale of gas diffusion; this suggests the conclusion that surface tension is not related to the stability of nanobubbles [47]. Based on their research, the authors of [29] report that the speed at which nanobubbles coalesce is high, and thus that the existence of insoluble impurities limiting diffusion at the gas–liquid interface is probable. This is another argument against the pollution theory.

Another theory regarding the existence of nanobubbles is the dynamic equilibrium theory [48]. According to this concept, the gas inside the nanobubbles diffuses under the influence of surface tension, and the dispersed gas is compensated by the gas entering the nanobubbles at the same time, thus achieving a state of dynamic equilibrium. The gas inside the nanobubbles continuously dissolves in the water, driven by surface tension and the dissolved gas thus concentrates on the hydrophobic surface. Then, the gas with increased concentration passes into the nanobubbles from the three-phase contact line to compensate for the gas diffusion. Some research results confirm the validity of this theory. It has been shown that in degassed water, the thickness of the depleted density layer is reduced, which reduces gas supply. As a result, the nanobubbles in the degassed water may shrink or dissolve. Additionally, some facts confirm the validity of this theory. According to [49,50], the addition of electrolytes does not affect the parameters of the dynamic equilibrium theory, which means that it does not affect the morphology and stability of the nanobubbles according to the Brenner and Lohse theory. However, in the work of Meegoda et al. [37] the relationships between the numbers of OH^- ions and the properties of nanobubbles, including their stability, are presented. This is one reason why one might question the theory of dynamic equilibrium. Zhang et al. [51] found the presence of nanobubbles on the mica surface. Mica is a completely hydrophilic material, and the contact angle is approximately 0° . No gas accumulates at the solid–liquid interface and no gas enters the nanobubbles from the three-phase contact line to compensate for the gas outflow. Therefore, according to the dynamic equilibrium theory, nanobubbles cannot exist stably or disappear instantly on the mica surface.

Zhang et al. [52] reported that negatively charged gold nanoparticles, when mixed with bulk nanobubbles, performed differently depending on their size. In the case of a solution of 60 nm gold nanoparticles with nanobubbles, the larger gold nanoparticles interact strongly with the nanobubbles; the dynamic light scattering data obtained during the study no longer showed either the peak of pure gold nanoparticles or the original nanobubbles. Instead, a new peak of intermediate size is formed, smaller than the original bulk nanobubbles but larger than the original gold nanoparticles. Aikawa et al. [53] present the possibility of using nanobubbles as corrosion inhibitors in acidic environments. In [54], water electrolysis was used to generate negatively charged nanovesicles that interact with liposomes with a controlled surface charge. It was confirmed that nanobubbles interact with both positively and negatively charged liposomes.

3. Nanobubbles for the Surfaces Cleaning

It is well known that surface cleanliness can be achieved without the use of detergents or surfactants by using mechanical methods. In this approach, the fight against lichens can

be effective thanks to nanobubbles, which play an important role as a mechanical barrier against the adsorption of pollutants on the surface [55]. A safe approach would be to cover the surface with nanobubbles before using them in a process exposed to contaminants, which would protect the surface from their effects. However, it should be noted that although this process is effective, the surface is not fully covered with nanobubbles. On the other hand, if the production of nanobubbles takes place on an already contaminated surface, the entire number of contaminants can be removed [55].

In [55], both hydrophobic and hydrophilic surfaces were examined for cleaning and prevention. Nanoparticles prepared by water electrolysis were used, which creates surface nanobubbles associated with the supersaturation of dissolved gas. The commercially available protein bovine serum albumin (BSA) was used as the contaminant. It was found that for already contaminated hydrophilic surfaces, the nanoparticles could remove the contaminant, but not for hydrophobic surfaces. The membrane fouling process plays a key role in the surface cleaning problem. Due to the widespread use of membrane separation technology in desalination, wastewater treatment, dairy production, biomass/water separation and rare metal recovery, there is a need for cheaper and greener antifouling technologies. One of the proposed alternative and promising techniques is the use of nanobubbles, which can significantly reduce the phenomenon of membrane fouling. Studies have shown that nanobubbles inhibit the growth of CaSO_4 and CaCO_3 crystals on the surfaces of ceramic membranes. Additionally, they support seawater reverse osmosis operations without the need to use chemicals. This makes it possible to reduce the operating costs of the installation and the negative impact on the environment by eliminating the use of chemicals. Nanobubble technology offers a promising path to achieving the goal of cleaner and more effective cleaning of membrane surfaces [56,57]. Figure 6 depicts the process of nanobubble formation during gas flow through the membrane.

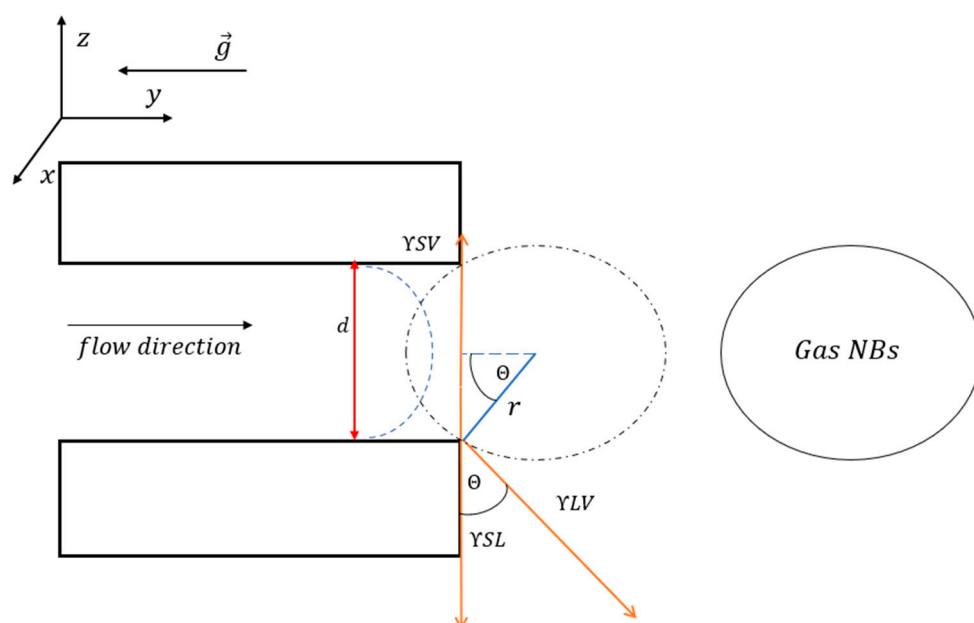


Figure 6. Simplified scheme of nanobubble formation as a result of flow through a ceramic membrane, where YSV is the solid–vapor interfacial energy, YSL is the liquid–vapor interfacial energy, θ is the contact angle, D indicates pore size, r is a radius of the NBs.

It is worth mentioning the existence of nanobubbles stabilized with a coating. NBs can be fabricated from various gas core and shell components, thereby influencing NB stability, biocompatibility and acoustic response. Phospholipids are usually commonly used for stabilization. Because their amphiphilic nature facilitates the rapid self-assembly of a monolayer at the liquid–gas bubble interface, thereby increasing stability and providing enhanced biocompatibility, this increases the applicability of nanobubbles in medicine [26].

Nanobubble therapy is an innovative therapeutic strategy that attracts researchers' interest due to its ability to deliver targeted and prolonged release of drugs with increased efficacy and minimal side effects. Compared to traditional methods such as chemotherapy, photodynamic therapy (PDT) and photothermal therapy (PTT), nanobubble therapy has several potential advantages, with specificity being the main one. Nanobubbles also have a minimally invasive nature and the ability to be customized for targeted therapy, opening new possibilities in cancer treatment. Despite existing gaps and uncertainties related to nanobubble therapy, extensive research can lead to the discovery of a potentially effective therapy that will provide better cancer treatment in the future [58].

Gao et al. [59] developed US contrast agents targeting CA-125 nanobubbles. Overexpression of the CA-125 tumor antigen characterizes ovarian cancer. In this work, the NB surface was functionalized with the CA-125 antibody. It was reported that targeted NBs showed increased tumor retention and longer echogenicity compared with untargeted NBs. In another study, Yu et al. [60] developed targeted NBs functionalized with anti-G250 nanobodies to improve molecular imaging of renal cell carcinomas in the US. The G250 antigen 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 specifically bind to tumor cells, improving ultrasound imaging. In their recent work, Liu et al. [61] constructed T-cell-targeted NBs to detect T-cell infiltration in acute heart transplant rejection. T-cell-targeted NBs (NBCD3) and control NBs (NBCON) were characterized by zeta potential analyzer, optical and fluorescence microscopy, and flow cytometry analysis was performed. Zhang et al. [62], in another recent publication, successfully explored the potential application 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 diagnosis of atherosclerosis using ultrasound imaging. Targeted nanobubbles have a surface functionalized with certain targeting ligands that enable adsorption to specific cancer cells and are also preloaded with drugs or chemotherapeutic genes [63].

Wu et al. [64] developed targeted cationic NBs loaded with FoxM1 siRNA for prostate cancer gene therapy. It has been shown that the produced NBs combined with the destruction of nanobubbles using ultrasound can effectively inhibit tumor growth [65], Curcumin-loaded polymeric NBs synthesized for prostate cancer treatment. This therapy has been reported to be effective in preventing metastatic spread in prostate cancer cells.

4. Technological Advancements in Adsorption Refrigeration: Improving Performance and Efficiency

Technologies related to adsorption refrigeration are an intensively developed branch of industry and science. In recent years, researchers have proposed a number of improvements to increase the ability of these devices to produce cold and make them more and more competitive with compressor refrigerators. Bujok et al. [66] provided a comprehensive discussion on critical design factors of heat exchangers influencing their performance. Plate–fin heat exchangers and fin–tube heat exchangers were discussed. The experience in operating of the three-bed adsorption chiller with the desalination functionality, equipped with a plate–fin tube heat exchanger, was also considered by Sztékler et al. [67].

Interesting solutions include the use of heat pipes [68], metal foam [69] and coated heat exchangers [70]. Another option is the optimization of operating parameters to achieve the best possible performance of the system [71]. Nikbakhti et al. [72] proposed incorporating adsorption into the absorption cycle. This allows the adjustment of the generation/adsorption pressure and enhances the system's performance. An integrated adsorption–absorption (AD–AB) cooling system was used to study two solar system layouts, one with and the other without a hot water storage tank [73,74]. The proposed novel system achieved a higher performance than conventional systems. The thermal capacitance ratio of the adsorbent and inert material of the sorption element, overall thermal conductance ratio of the sorption element and evaporator and silica-gel mass were analyzed in [75,76]. The increase in adsorbent mass resulted in the increase in

sorption properties, which improves the cooling capacity of the considered reheat two-stage adsorption chiller. Moreover, the increase in the overall thermal conductance ratio for the evaporator to sorption element resulted in an increase in cooling capacity. Since the poor thermal properties of adsorption-packed beds of adsorption chillers are responsible for the low coefficients of performance, the influence of metal particle and carbon nanotube additives on the thermal diffusivity of a silica-gel-based adsorption bed was analyzed by Kulakowska et al. [77] and Pajdak et al. [78]. Despite the increased thermal diffusivities of the prepared modified adsorption beds, the addition of CNTs to SG resulted in a decrease in the sorption capacity. Another interesting option is the use of fluidization adsorption beds [79–85]. This is a new concept, in which fluidized adsorption beds replace packed beds in conventional adsorption chillers. Another promising method of improving heat and mass transfer uses micro- and nanobubble technologies for adsorption cooling and desalination systems.

5. Advanced Adsorption and Desalination Systems Using Ultrafine Bubbles

An interesting concept that could find applications in further development of adsorption cooling and desalination systems is the use of micro- and nanobubble generators [86,87]. The unique properties of nanobubbles have already found applications in many engineering fields. Nanobubbles dissolved in the material cause an increase in porosity, which consequently changes the material's original properties, such as density or fluidity. Bubbles smaller than 1 μm exhibit exceptional stability in a solution due to their size. Such tiny bubbles do not rise to the upper surface of the solution and do not burst due to their high surface-to-volume ratio. Bubbles created through cavitation seem to be a promising solution for using heat transfer fluids. It is also worth mentioning that the unique properties of nanobubbles, such as their size, sizeable interfacial space and long residence time, stimulate further research into their potential applications [88]. From a heat exchanger design perspective, maintaining turbulent flow through the heat exchanger tubes is a desirable effect [66]. Therefore, introducing micro/nanobubbles into the circulation of water flowing through the exchanger can help increase the heat transfer between the fluid and the exchanger walls.

Research presented in [89] suggested the increase in heat transfer processes in the presence of micro- and nanobubbles. An experiment in [89] involved observing the time required to heat and cool water in a tank. Nanobubbles were dissolved in one of the tanks, while the other was left as a control. Both tanks were placed on a hotplate, and an experiment was conducted to determine the time required to reach the desired temperature. The temperature ranges used in the study were 304.15–343.15 K and 304.15–353.15 K for heating and, inversely, 353.15–343.15 K and 353.15–304.15 K for cooling, respectively. The experiment showed that the time required to heat the water was longer during trials with water containing dissolved bubbles. However, when cooling the water from the specified temperatures, the water with dissolved nanobubbles achieved significantly faster results for longer cooling trials. Shen et al. [18] mention that in the case of heat transfer enhancement by nanofluids and acoustic cavitations, the smaller size of the bubbles improves heat transfer, but the temperature generated by the bubbles during their collapse may increase the fluid temperature [18].

The change in the rate of fluid cooling is a favorable phenomenon in the context of adsorption refrigerators. Installation of devices that generate nanobubbles before entering the adsorption bed, due to the generated bubbles, will increase the convective heat transfer coefficient from the water to the heat exchanger wall, improving the overall heat transfer coefficient. A schematic diagram of the proposed setup is given in Figure 7.

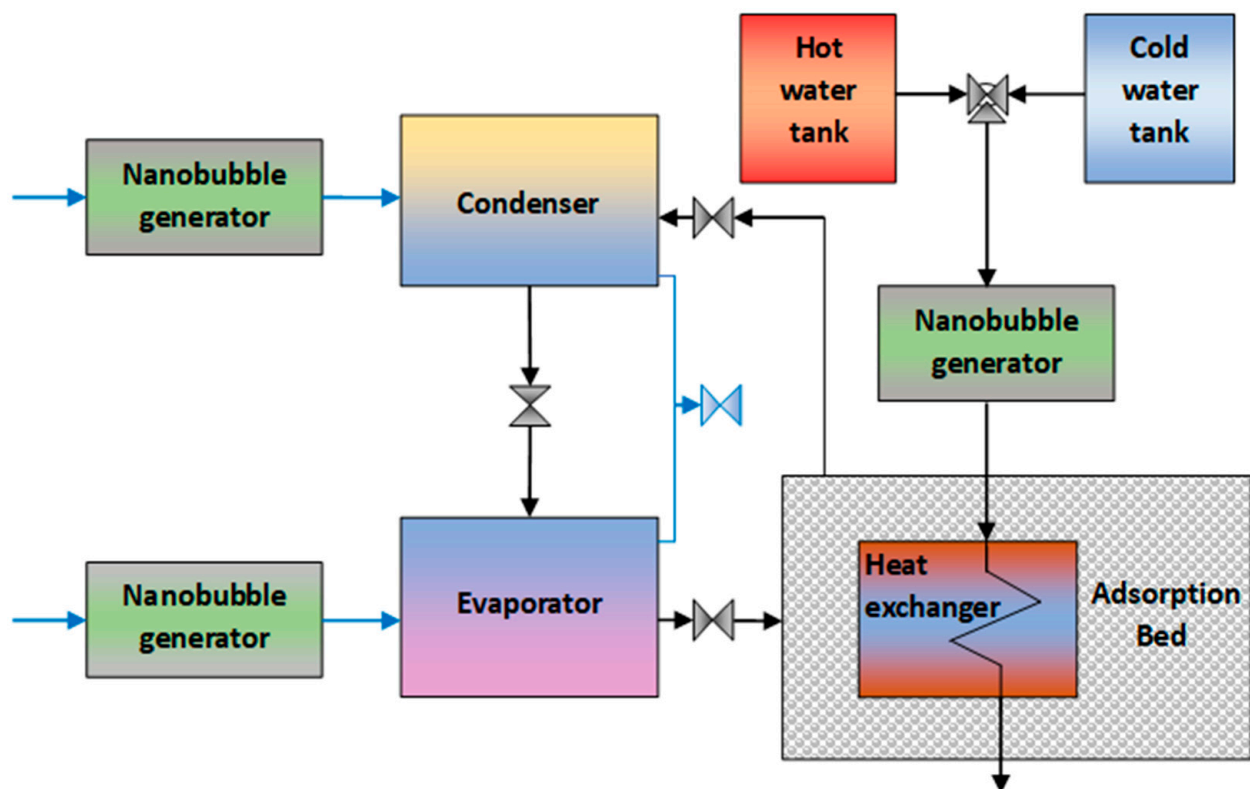


Figure 7. A schematic diagram of the proposed idea.

The adsorption chiller is composed of heat exchangers in the form of an evaporator and condenser and an adsorption bed. In the adsorption bed, mass and heat exchange systems are combined to create a refrigeration device. Cold water is supplied to the bed in the adsorption phase to cool it, and hot water in the desorption phase to warm it. The valves connecting the evaporator with the bed in the adsorption process are open, so the water vapor produced in the evaporator is absorbed by the bed. After completing a single adsorption cycle, the valve connecting the bed with the evaporator is closed. The bed's ability to absorb is limited, so after a certain time it requires regeneration, which is carried out using hot water. While the bed is heating, the valve connecting the bed and the condenser is open and allows the refrigerant to condense. An innovative approach is the introduction of a nanobubble generator to the adsorption refrigerator, which is intended to intensify the heat transfer process.

The author of [90] presents a process to produce a nanofluid via acoustic cavitation to investigate the effect of dissolving copper nanoparticles on heat transfer. However, acoustic bubbles were not the only medium responsible for heat transfer, and the author emphasizes their importance in improving heat transfer in the apparatus he presented. Furthermore, the author highlighted that the bubbles reduced the thickness of the boundary layer and concludes that the nanofluid behaves more like a fluid than the conventional solid–liquid mixtures, while, however, still retaining mixture characteristics such as Brownian motion, gravitation and frictional forces [90]. The nanobubbles' size and coalescence prevention are crucial aspects for enhancing heat transfer using micro/nanobubbles. Meegoda et al. [37,91] suggest that the gas used to create the nanobubbles and the environment in which they are produced play essential roles in keeping the bubbles small and preventing coalescence. Moreover, in the case of hydrodynamical cavitation presented in [92], the authors also conclude the dependence between the content of different molecules in a liquid and the size and stability of nanobubbles. The presence of hydrophilic nanoparticles could have positive and negative effects, depending on the particle's concentration, while the hydrophilic particles, even in small numbers, sharply increase the median size of the nanobubbles [92].

The research presented in [93] proved the influence of pH on decreased nanobubble size. The authors of that study also assumed that the developed double layer plays a vital role in the formation and stability of nanobubbles. The high negative potential zeta provides a repulsive force which prevents interbubble aggregation and coalescence of bubbles [93]. The amount and type of gas in which nanobubbles are made play an essential role in the behavior of the whole fluid and its thermal properties [94]. Nanobubble-forming gases are characterized by low thermal conductivity; thus, too many bubbles dissolved in fluid cause a decrease in the ability to transfer heat between the fluid and the wall of the heat exchanger [95].

6. Discussion

It can be concluded, based on the cited literature, that the topic of bubbles in the context of mass and heat transfer remains something of a mystery, despite the passage of time since their discovery. There are also many discrepancies between the theoretical knowledge about nanobubbles and the research experiments. Therefore, it seems important to improve our knowledge in this area, as all researchers are aware of the unique properties of micro- and nanobubbles. However, despite learning about their wide range of applications, there is still a gap in knowledge as to what gives them their unusual values. This provides motivation for further exploration of this topic. Considering the cited publications in the context of the article's subject, the main advantage of using bubbles is their potential to increase mass and heat transfer.

Even though, according to the abovementioned studies, nanobubbles can improve heat transfer, studies such as [96] should not be omitted because, according to their authors, nanobubbles do not significantly contribute to improving heat transfer. These authors claim that only larger bubbles can introduce turbulence significant enough to have a real effect. Based on the comparison of these two positions, the question arises of how large the bubbles should be in order to significantly impact a fluid's thermal properties. Moreover, in this case, it is also necessary to consider whether increasing the size of the bubble will have a negative or destructive effect on the heat exchange system in adsorption refrigerators. This is undoubtedly a field for interesting research aimed at finding an optimal solution combining the required bubble size, their stability and appropriate enhancement of the fluid's thermal properties flowing through the heat exchanger. In the context of using micro/nanobubbles in adsorption chillers for heat exchange, one cannot forget about the process conditions that influence the addition of bubbles to the coolant. The appropriate selection of the flow speed and the number of bubbles that enter the heat exchanger (bubbles' concentration in the fluid) plays an important role. To improve heat transfer conditions, reaching an equilibrium point where the number of bubbles is large enough to cause flow disturbances and increase turbulent motion while destroying the boundary layer is important. However, the number of bubbles must be small enough to allow the refrigerant flow to freely absorb the excess heat generated by adsorption processes in the bed. Similar principles apply to the behavior of fluidized beds used in refrigerators and dehumidification systems, where inlet velocity plays a key role in heat transfer [83,84,97].

Introducing a certain number of micro- and nanobubbles into the water circulation would eliminate the problem of difficult heat conduction in the boundary layer of the heat exchanger. Moreover, it should be noted that at the nanoscale, the effect of bubbles on heat transfer was not recognized. Moreover, improving ecological refrigeration technologies in a time of increasing energy prices and global warming necessitates progress in refrigeration technologies. The lack of a unified theory describing each aspect of nanobubble behavior makes work on this topic difficult. The small size of the tested objects introduces additional difficulties in the form of the need to eliminate any contaminants from the tested samples, because particles found in the tested fluids may be incorrectly classified as nanobubbles, thus affecting the assessment of their properties.

The presented theories exemplify the fact that there are still contradictions in the research process of nanobubbles. Theoretically, nanobubbles should dissolve within

1 ms, while in reality there is evidence that they can remain stable for up to several days. Another important element is that the pressure inside the bubbles calculated using the Young–Laplace equation should be significantly different from the atmospheric pressure. However, there are also some discrepancies because, according to the author of [98], the pressure is slightly higher than atmospheric. Therefore, this subject is still awaiting examination.

7. Conclusions

Micro/nanobubble (mNB) technology involves creating extremely small gas bubbles in a liquid. The smaller the diameter of the bubbles, the more stable they are. Bubbles with a diameter of about 100 nm can remain in liquid for a very long time, even up to several weeks, depending on external conditions. This and other unique features of mNBs make them a powerful tool in many industries. The use of small bubble technology increases the efficiency of chemical reactions by multiplying the interfacial surface in liquid–gas reactions (the surface expands rapidly due to the bubble explosion). In specific applications, it is necessary to develop new or modify existing devices that will use gas saturation of liquids.

Micro- and nanobubbles have the potential to contribute to major growth in many areas of science. They can be applied to other systems, such as dehumidification systems or auxiliary mass extraction systems. However, the prerequisite for this application is that the system has an installation that allows the injection of micro- and/or nanobubbles.

The issue of nanobubbles is still not fully resolved: their stability. In this case, the following question remains: “What makes them so stable?”. Despite the passage of time, this seemingly simple question remains without a certain answer. Despite the creation of many theories, there is still no uniform answer, especially in the context of bulk nanobubbles. Due to the authors’ field of interest, the main path to develop nanobubble technology is their use in the cooling and desalination sector. The use of nanobubbles in adsorption refrigerators will support the development of the field with new research. According to the authors, the experience gained in the field of interaction between nanobubbles and heat exchangers will bring a completely new value. This article can be treated as an introduction to future research on the possible potential of nanobubbles. In the context of heat transfer, nanobubbles can provide a cheap and environmentally friendly additive that improves heat transfer, hence the interest in this technology.

Apart from the directions we have already mentioned, the use of nanobubbles in cancer therapies is certainly a very interesting research direction. Although there is still not enough research confirming the possibility of use in clinical conditions, the potential of nanobubbles may shed new light on cancer therapy. Of course, we cannot forget about other applications, such as surface cleaning, water aeration and soil purification; nanobubbles can also be used in many aspects in environmental engineering applications. It is also worth recalling the work showing the auxiliary role nanobubbles play in chemical processes. In the case of horticulture, hydroponics and aquaculture there are also many applications. Returning to the topic of medicine, nanobubbles have the potential to help fight cancer and they have also proven to be useful in biomedical applications. In relation to the first part of the question, we are limited by our knowledge regarding the physical properties of bubbles, their stability and reactions at the nano scale with other particles and pollutants. However, we are convinced that the potential breadth of their applications will certainly reward the effort put into developing theoretical issues related to them. The use of nanobubbles can introduce a new method for improving mass and heat transfer in refrigerators. Micro- and nanobubbles allow the creation of a completely new type of improvement to these devices. The work collected for this purpose focused on other aspects of refrigerators, such as the construction of the bed or the use of new types of admixtures. In addition, the use of various types of gases and process conditions will certainly contribute to better understanding and even greater popularization of ultrafine bubbles.

This paper also presents an innovative solution that opens a new field of interest and research for the authors. Adsorption cooling and desalination systems use low-temperature

renewable and waste heat energy sources, being part of the area of activities related to sustainable development. The impact of the use of micro- and nanobubbles on heat transfer in heat exchangers opens a new research area and constitutes another step towards understanding the unique properties of nanobubbles.

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Nomenclature

NB	Nanobubble
MB	Microbubble
mNB	micro/nanobubble
SNB	surface nanobubble
BNB	bulk nanobubble
PDT	photodynamic therapy
PTT	photothermal therapy
γ_{SV}	solid–vapor interfacial energy
γ_{SL}	liquid–vapor interfacial energy
P	pressure [Pa]
r	bubble radius [m]
γ	surface tension [Pa]
Δ	change
θ	contact angle [°]
D	indicates pore size [m]

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