**Research Campaign: Use of Nanobubbles for Enhanced Gas-Liquid Contacting in Spaceflight Environments**

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**1.**   **Introduction**

The concept of dissolving gases into liquids on Earth is straightforward. Mixed phase sparging relies on buoyancy effects found in gravitational environments to move pockets of gas through liquid media. This process, dependent on mass transfer at the liquid/gas interface, can dissolve gases in aqueous media according to Henry’s law to advance chemical reactions and biological processes [1]. Gas sparging is resource and energy-intensive, as gas delivery efficiency into aqueous media depends on the short bubble residence time in water given buoyancy [2]. The gas transfer becomes inefficient in microgravity environments with the lack of density-driven bubble movement. Thereby, restricting sparging applications to terrestrial environments. Reducing the bubble size while increasing bubble concentration for a given liquid volume, can drastically increase the gas-liquid interfacial area and mass transfer potential, even in microgravity environments [3].

Ground-based experiments in controlled environments have shown the ability to produce metastable nanobubbles that remain in solution for days to months. Negligible bubble buoyancy due to bubble size means Brownian motion dictates mass transport and eliminates the reliance on gravity [4]. Preliminary results show carbon dioxide (CO2) nanobubbles increase buffering capacity by 10-fold when compared to macrobubble sparging [1] [2] [4]. Biological systems (fish, mice, plants, microorganisms) benefit from oxygen (O2) availability in the form of nanobubbles [5,6]. However, these initial bubble stability characterizations were limited to quiescent storage environments, using deionized water with air, carbon dioxide, oxygen, or nitrogen for bubble production [4,5]. Using different combinations of gases and liquids expands the scope of these experiments and benefits the applications reliant on controlled concentrations of dissolved gases (e.g., wastewater remediation, medical applications, aquaculture, horticulture, and photobioreactors) [6]. Results of these experiments have the power to increase overall system efficiency, reduce power consumption and physical footprint. Conducting nanobubble characterization in dynamic storage environments, such as vibrational loading and uncontrolled temperatures, reflects launch and spaceflight conditions and provides an initial assessment in system competitiveness. Moreover, theoretical bubble models are further refined by empirical bubble measurements with various liquid viscosities and gas species.

This white paper briefly highlights the opportunities for nanobubbles to overcome limitations/challenges of current gas-liquid contacting approaches, the evidence of nanobubble production and stability, spaceflight applications potentially benefiting from nanobubbles, and the research necessary to model and diversify the application of nanobubbles. An experimental program aimed at both ground-based and orbital experiments elucidates nanobubble formation and stability. Thereby, extending the inclusion of sparging systems within various gravity environments, including microgravity, and advancing Earth-independence in human spaceflight.

**2.**   **Current Limitations of Gas-Liquid Contacting**

Buoyancy-dominated and diffusion-dominated are the two categories for terrestrial approaches to mass transfer in gas-liquid systems. As previously presented, those technologies relying on density gradients (bubble columns, packed columns, spray towers) are inappropriate for a microgravity environment. Membrane contactors are diffusion-dominated and have demonstrated their mass transfer capabilities in limited microgravity studies [7–10]. Nonetheless, the contactors are rate limited due to membrane permeability, diffusion coefficients, and Henry’s law, regardless of contactor design or membrane material. These membranes have some selective capability due to gas-specific permeability. However, all available, diffusible gases can pass through the membrane. Thereby, potentially requiring additional systems or operational considerations for minimizing or reducing unwanted mass transfer (water vapor, CO2/O2, etc.). Furthermore, membrane materials provide ideal scaffolding for biofilms and other precipitating materials. Porous membranes suffer degradation in bulk mass transfer due to buildup on contacting surfaces and pore wetting; while nonporous membranes typically have inherently low permeability coefficients, regardless of surface treatments [11]. System designs using membranes can quickly balloon in size to overcome limitations in mass transfer but are a standard for gas-liquid contacting in microgravity.

**3.**   **Questioning the Existence of Nanobubbles**

Nanobubbles are small gaseous structures in liquid solutions with size distributions below 1000 nm; that have high surface energy and gaseous-carrying capacity [6,12–14]. One of the most groundbreaking properties of nanobubbles is their outstanding stability without coalescing for weeks to months [15–17]. Nanobubbles are not affected by buoyancy forces and their transport in solution is explained by Brownian movement. The sustained presence of nanobubbles in solution explains the drastic increase of gas availability in solution. However, the metastable existence of these nanointerfaces contradicts the traditional Young-Laplace bubble catastrophe theory and the Epstein-Plesset theory of bubble dissolution and growth [18].  According to the Young-Laplace equation (Eq 1), theoretically, nanobubble should dissolve due to the high Laplace pressure ΔP (*Pinterior* - *Poutside*) [19–21].

Pinterior=Poutside + 2 𝛾/R                                                       (1)

Here, *Pinterior* and *Poutside* are the pressure inside (vapor phase) and outside (liquid phase) of the bubble, respectively, *R* is the radius of a spherical bubble, and 𝛾 is the surface tension. For example, a nanobubble with *R* = 100 nm, 𝛾water = 72 mN m-1 and atmospheric pressure in the surrounding water = 105 N m-2, results in pressure inside the nanobubble of approximately 14×105 N m-2. According to Epstein and Plesset's theory, the bubble should dissolve within approximately 10 μs [22]. The contradiction between the theories mentioned above and experimental observations of nanobubble existence and stability is an exciting challenge for scientists. Various concepts were proposed, explaining the stability of the bulk nanobubbles, including stabilization due to minor contamination, liquid-gas interface surface charge, significantly different surface tensions, or gas densities compared to their macroscopic counterparts [23–29]. Experimental evidence and nanobubble characterization by different techniques demonstrate that nanobubbles exist and are an untapped way to increase gas delivery for gas-starved systems and other gas dependent technologies. Applications of nanobubbles can emerge from fundamental understanding of physical-chemical properties under both gravity and microgravity conditions.

**4.**   **Nanobubbles Improve Mass Transfer to Liquids**

Due to the high internal pressure of the nanobubbles, a high mass transfer driving force of gasses into solutions is demonstrated. In addition, the small diameter of nanobubbles results in a higher surface area per unit volume. The combination of the mass driving force and higher contact surface area yield a higher volumetric liquid side mass transfer coefficient [16]. This increased mass transferability of nanobubbles has enhanced environmental processes such as wastewater treatment, microbial cultures, seed germination, and plant growth [17,30–34]. Aeration plays a significant role in delivering O2 to sustain life for organisms and initiate oxidative pollutant degradation in these processes. In most of these processes, conventional aeration techniques do not offer sufficient mass transfer efficiency. Approximately 45-75% of a wastewater treatment plant’s total operating cost is electricity powering aerators or diffusers, but O2 transfer efficiency in these plants is only about 6-10% [35]. The volumetric mass transfer coefficient and O2 utilization rate for nanobubbles is 2-fold greater when compared to conventional aeration, due to their low buoyancy diffusing O2 into the surrounding water [35,36]. Increased O2 mass transfer efficiency makes aeration/oxygen supply approaches more cost-effective. Li et al. confirmed that water infused with nanobubble aeration for 15 minutes had a 120% higher O2 concentration than water at equilibrium with air [37]. Comparable results were reported when comparing macrobubbling versus nanobubbling. Under identical operation conditions and reactor settings, the mass transfer coefficient (kLa) for CO2 gas was 11 times greater for nanobubbles [4]. Given large interfacial area, lack of buoyancy, and bubble residence times, increased mass transfer, and sustained gas availability in solution of up to months, nanobubbles will become a paradigm shift for enhanced gas-driven processes.

**5.**   **Impact of Improved Gas-Liquid Contacting on Spaceflight Applications**

Low gas solubility and membrane permeation rates force environmental control and life support system (ECLSS) designs to incorporate technologies relying on toxic materials or hazardous operations to reduce system mass [38,39]. Sustained diffusion of gases by increasing the longevity and stability of bubbles in solution would benefit terrestrial life but also extended to long-duration human spaceflight applications.

*Waste Remediation*

     Current wastewater remediation systems on the ISS (current long-duration human spaceflight standard) rely on an energy-intensive and hazardous process for water reclamation. Water remediation at the ISS includes chemical stabilization of waste, vapor compression distillation, polishing step with ionic exchangers, catalytic oxidation, and iodine dosing for long-term storage [38,39]. Nanobubbles can supplement under-performing catalytic oxidation units, enable biological treatments, and minimize physical footprint of reactor units for the ISS. Terrestrial research is also interested in accelerating wastewater treatment with nanobubbles. Heavy metal reduction rates were 300% faster in nanobubble treatment systems when compared to identical systems sparged with macrobubbles [6]. Additionally, these studies observed radical oxygen species (ROS) formation on the outside of O2 nanobubbles, providing bactericidal effects. A final water “polishing” step before distribution, or a less hazardous method for stabilizing long-term water storage than iodine by using ROS is hypothesized [12]. Bacterial biofilms have an increased resistance to antibiotics, disinfectants, and environmental stresses in microgravity [40,41]. The surface cleaning properties of these nanobubbles combat this increased bacterial persistence issue. Thereby, benefitting components most likely to succumb to biofouling (i.e., of any wetted surface). Proof-of-concept results demonstrated higher removal of organic load in aerobic biological treatments when using nanobubbles [3] [4] [42,43].

*Food Production*

Nutrient composition will degrade in packaged food during Martian and extended Lunar missions, calling on the need for fresh produce during flight [38,39]. Therefore, continued operation of NASA's Veggie system, since its installation on the ISS in 2014, validates the capability of growing higher plants in microgravity for nutritional supplementation [44]. Expanding the scope to include sources of protein, NASA, ESA, and Roscosmos conducted initial experiments using tilapia and other fish on the Space Shuttle and FOTON-M3, respectively. However, wide-spread fish death and early experiment termination was a result of insufficient O2 supply in these closed-loop systems [45,46]. In terrestrial studies, increasing the dissolved O2 concentration in the water of both the plant and fish systems significantly increased crop and fish yield, and potentially resistance to pathogens [47–50]. Incorporating nanobubbles into the Veggie system or future fisheries may increase overall production and increase the production to volume ratio. Thereby reducing the required system size to feed an entire crew over months to years.

*Air Revitalization*

Propelling humans farther from Earth without addressing carbon loop closure in air revitalization will result in launching excessive consumable and waste parasitic mass. Bioregenerative technologies, particularly algal photobioreactors, can close this gap by fixing respired CO2, providing O2, and edible biomass [51,52]. Both the NASA 2015 Technology Roadmap and 2020 Technology Taxonomy identified CO2 removal (closed loop) with O2 recovery as a significant technical challenge for sustained spaceflight, while suggesting solutions in the metabolic processes of algae and bacteria [38,39]. Algal studies conducted in microgravity and terrestrially for air revitalization utilized membranes for CO2 provision [53–55]. The result of these studies observed positive algal growth rates. However, these cultures experienced constrained photosynthetic rates, as diffusion through the membrane was unable to support culture metabolism. Providing enhanced CO2 delivery to algae will further augment the functionality of this bioregenerative system [46,56,57].

**6.**   **Future Research Needs and Recommendations**

We recognize the potential impact of nanobubbles on increasing the endurance of human existence in space. To take advantage of these benefits, the following research questions and objectives need addressing. Our comprehensive research plan leverages insight gained from terrestrial characterizations to develop empirical models and inform subsequent microgravity experiments.

*Terrestrial Experiments*

     It is necessary to expand the experimental matrix beyond oxygen, carbon dioxide, and nitrogen dissolved in water. Varying gas species used to produce nanointerfaces and the saturated solution further develops the understanding of the sensitivity of nanobubble stability to gas nature and physical-chemical properties (e.g., inert gas, gas with acid-base properties, gas with redox character, etc.). Nanobubble characterization methods need standardization of nanobubble quantification and identification. Establishing these methods will enable studies on nanobubble stability and formation. Subjecting nanobubble-saturated solutions to vibrational and dynamic temperature environments, reflective of launch environments, will test bubble cohesion and their appropriateness for flight.

     Previous studies investigated characteristics and stability of bulk nanobubbles for simplified biphasic systems. However, surface nanobubbles require research to comprehend the intermolecular interactions between gas bubbles and solid surfaces. This investigation would delineate the role of reactor and storage vessels for fundamental behavior of nanobubbles in pertinent space applications.

     Laplace Theory realizes surface curvature and electrostatic interactions to describe the stability of gas-liquid interfaces. Recent literature, however, often omits those parameters and simplifies the bubble internal pressure to bubble size only (Eq 1). However, at the nanoscale, the bubble curvature term, as well as charge density, become significantly influential for nanobubble stability. The empirical affirmation of Laplace Theory could be possible with recent developments in bubble characterization and quantification. This approach could enable a fundamental breakthrough in the mathematical modeling of bubble behavior.

*Microgravity Environment Experiments*

It is imperative we conduct nanobubble characterization experiments in sustained microgravity environments. If the longevity of these bubbles in microgravity reflects the current terrestrial results, experiments may need to be in orbit for weeks to months to capture significant bubble degradation. The first flight experiment would include samples of fluids, saturated with nanobubbles pre-flight, monitored to capture the sustainment of these bubbles, but also empirically validate terrestrial models. After understanding these longevity periods, the second spaceflight experiment would verify the ability to produce these bubbles in the microgravity environments. In-flight production is essential if the biological life support technologies constantly consume these gas products, creating a demand on the system. Only after validating these characteristics can we start to use the bubbles in the previously mentioned applications, including enhanced plant or algae growth and waste remediation. The third flight test uses the Veggie system as a testbed, where we can characterize enhanced plant growth in spaceflight-proven systems.

**7.**   **Conclusion**

     Nanobubbles in liquid solution are considered to behave as incompressible monophasic liquids which will revolutionize gas provision in microgravity environments. Experiments conducted both terrestrially and in spaceflight must characterize gas species/liquid interactions, bubble stability in launch environments, generation in microgravity, and biological system response. Experiments that demonstrate those benefits are essential stepping-stones for nanobubble implementation in multifunctional ECLSS.

**8.**   **References**

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