## HIGH PRESSURE WATER SCRUBBER FOR CONTROLLED ATMOSPHERE STORAGE

by

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#### ABSTRACT

Ph.D.

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# HIGH PRESSURE WATER SCRUBBER FOR CA STORAGE

A theoretical model of a high pressure water scrubber (HPWS) was developed in order to evaluate the performance of such an apparatus in reducing the  $O_2$  pull-down period in commercial controlled atmosphere storage facilities for fruits and vegetables. A simulation study indicated that the HPWS could be designed to meet most specific requirements.

Mass transfer considerations in the design of the HPWS required: 1) a non-intrusive technique to sample bubble clouds for accurate estimation of bubble physical dimensions generated at the bottom of a water column, this, in order to evaluate available bubble generators; and, 2) of a simple method to determine the solubility of gases in pure and contaminated water since existing methods were deemed time-consuming and expensive.

The digital image analysis technique of bubble measurement was refined by: (i) the development of a discriminatory parameter permitting identification of those bubbles within a known focus range such that errors could be held to reasonable magnitudes; and, (ii) the development of a mechanical bubble sampler enabling characterization of high density bubble clouds.

A simple and inexpensive pressure apparatus was developed to determine gas solubility in water of various degrees of purity. Results for  $CO_2$ ,  $O_2$  and  $N_2$  solubilities compared favourably with published data.

The HPWS was designed, constructed and its performance in controlling  $CO_2$  and  $O_2$  was tested under typical CA storage conditions. It was effective for  $CO_2$  removal, however, it did not reduce enough the  $O_2$  concentration of the CA storage chamber to justify the initial capital investment of this equipment.

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#### RÉSUMÉ

Ph.D.

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Agricultural Engineering

## LESSIVEUR À L'EAU À HAUTE PRESSION POUR L'ENTREPOSAGE AC

Un modèle théorique d'un lessiveur à l'eau à haute pression (LEHP) a été développé pour en évaluer les capacités à réduire la période de mise in régime d'entreposage à atmosphère contrôlée (AC). Une simulation a démontré qu'un LEHP peut être conçu pour répondre à la majorité des conditions d'entreposage AC.

Tenant compte du transfert de masse, la conception d'un LEHP requiert: 1) une technique de mesure non intrusive permettant l'estimation des paramètres physiques de nuages de bulles générées au bas d'une colonne d'eau afin de caractériser les générateurs de bulles disponibles; et 2) une méthode simple de mesure de la solubilité d'un gaz dans l'eau distillée ou contaminée, les méthodes existantes étant dispendieuse et laborieuse.

Une méthode de mesure de bulles basée sur l'analyse d'images digitalisées a été améliorée par: (i) la mise au point d'un paramètre discriminatoire permettant d'identifier les bulles qui sont à l'intérieur d'une distance focale correspondant à une erreur de mesure acceptable; et (ii) la mise au point d'un échantillonneur mécanique de bulles permettant la caractérisation de haute densité de population de bulles.

Un appareil simple et peu dispendieux a été mis au point pour déterminer la solubilité d'un gaz dans l'eau de différentes qualités. Les résultats obtenus lors de la mesure de la solubilité du  $CO_2$ ,  $O_2$  et  $N_2$  dans l'eau distillée sont comparables avec à ceux citées dans la littérature.

Un LEHP a été conçu, construit et mis à l'essai sous des concentrations de gaz normalement rencontrées dans l'entreposage AC. L'élimination du  $CO_2$  a été réalisée, toutefois le LEHP n'a pas suffisamment réduit la concentration de l' $O_2$  pour justifier l'investissement d'un tel appareil.

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#### FORMAT OF THESIS

This thesis is submitted in the form of original papers suitable for journal publications. The thesis format has been approved by the Faculty of Graduate Studies and Research, McGill University, and follows the conditions outlined in the "Guidelines concerning thesis preparation, section 7, Manuscripts and Authorship" which are as follows:

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Although all the work reported here is the responsibility of the candidate, the project was supervised by Dr. G.S.V. Raghavan, Department of Agricultural

Engineering, Macdonald Campus, McGill University, Montréal. The image analysis software for bubble population characterization and the solubilimeter were developed by the candidate under the supervision of **Dr. B. Panneton**. The entire research project was conducted at the Agriculture Canada Research Station, Saint-Jean-sur-Richelieu.

Dr. G.S.V. Raghavan is the co-author in the first manuscript. The second, third and fifth manuscripts are co-authored by Dr. B. Panneton and Dr. G.S.V. Raghavan. The co-authors of the fourth manuscript are Dr. B. Panneton, Miss V. Orsat and Dr. G.S.V. Raghavan. Co-authors of the sixth manuscript are Dr. G.S.V. Raghavan and Dr. R.L. Granger. The fifth paper, concerning the development of a solubilimeter, has been submitted for patent consideration and will not be published until it is authorized. All the other manuscripts were submitted to the Canadian Agricultural Engineering Journal. The first manuscript has been published and the second is in press.

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#### I GENERAL INTRODUCTION AND LITERATURE REVIEW

#### 1.1 General Introduction

The storage life of a number of fresh fruits and vegetables has been extended by storage in controlled atmospheres ranging from 0 to  $10\% O_2$ , 0 to  $30\% CO_2$  and the balance as N<sub>2</sub> (Kader, 1985). Recent studies on a number of apple varieties have shown that keeping quality is inversely related to the time taken to bring the storage atmosphere from ambient to controlled conditions, or O<sub>2</sub> pull-down period (Sharples and Munoz, 1974; Lau, 1983). Many systems for maintaining specific desired atmospheric compositions have been developed and a number of methods for rapid O<sub>2</sub> pull-down exist. However, none of the former incorporate rapid O<sub>2</sub> pull-down and the latter are either expensive or make use of expensive or hazardous input materials.

The research to be presented in this thesis concerns the development of a high pressure water scrubber (HPWS) for application in a controlled atmosphere (CA) facility for the long-term storage of fruits and vegetables. This apparatus was conceived as an improved alternative to existing methods of arriving at and maintaining the specific atmospheric compositions required in a CA storage setting, the perceived advantages being: 1) the potential to reduce the  $O_2$  pull-down period, 2) independence of expensive or dangerous input materials, and, 3) flexibility with respect to changes in type of stored commodity. Although the idea of using water or brine-scrubbers in storage applications is not new (Palmer, 1959; Smock et al., 1960), the conceptual modifications inherent in the HPWS were expected to lead to a more acceptable technology.

The HPWS is based on gas separation by solubilization in water, as is the conventional water scrubber. In a conventional storage scrubbing system, gas-liquid contact for mass transfer is produced by spraying water droplets through the storage air, capturing the droplets and circulating them to the outside. Solubilization rates are limited by the differential gas partial pressures, gas solubilities, droplet dimensions and water circulation rate. The process could, in theory, be rendered more flexible if the storage room pressure could be adjusted; however, such an approach is impractical at the commercial scale.

The HPWS concept is to bring storage air to a pressurizing container and produce the required mass transfer by bubbling the pressurized air through a water column outside the storage room where captured gas may be released. The main advantage of this conceptual modification is the ease of control of the degree of pressurization of a small quantity of storage air at a time so as to increase or decrease solubilization rates as well as adjust the overall system mass transfer ratio of O<sub>2</sub> to CO<sub>2</sub>. In principle, the HPWS can reduce the O<sub>2</sub> pull-down period and thereafter maintain the desired CA by scrubbing excess  $CO_2$  while limiting the entry of O<sub>2</sub> to the storage room.

The HPWS concept was formulated mathematically and its operation in a commercial-scale storage facility was then simulated. Results indicated that it should be possible to significantly reduce the  $O_2$  pull-down period in a commercial storage facility for apples and to maintain the desired CA thereafter. There were two major obstacles to the design and construction of the HPWS. These were: 1) that bubble clouds produced by available bubble generators were not parametrized with respect to the physical dimensions relevant to mass transfer, and 2) that there was no rapid and inexpensive method available to measure gas solubilities in water of qualities to be encountered during system testing and operation.

This thesis therefore describes the theoretical formulation, simulation and testing of the HPWS as well as the steps taken to overcome the obstacles mentioned above.

#### 1.2 <u>Review of Literature</u>

The purpose of this literature review is to place the research presented in this thesis in the context of existing agricultural research priorities in general, and to those specific recent developments deemed relevant to the work described. A philosophical basis for CA research is first established, basic principles of CA are then summarized and followed by a brief review of the available technology. The physical bases relevant to the HPWS system are then reviewed.

#### **1.2.1** Rationale for Controlled Atmosphere Research

In Northern climates such as ours, field production of fruits and vegetables is severely restricted. The need and demand for fresh produce has been, for the most part, satisfied by importation (including of locally-produced varieties), at a significant cost to the economy.

Efforts at extending the time of availability of local supplies through the development of the greenhouse industry have been stymied by rising energy costs and an uncertain global economic climate. Year-round production of most fresh commodities is, as yet, an ideal. A variety of commodities do, nevertheless, lend themselves to long-term storage in controlled atmospheres (Smock, 1979; Isenberg, 1979), thus encompassing, in some instances, an end-of-harvest to beginning-of-harvest cycle (9 to 12 months). Post-storage quality is fresh or near-fresh in some cases, thus resolving the year-round availability problem for certain items.

Potential improvements with respect to post-storage quality and storability extension are therefore important in this context. Moreover, improvements in post-harvest technology should also lead to higher quality imported produce and substantial reductions in losses associated with the transportation, wholesale and retail distribution time frames.

#### **1.2.2** Controlled Atmosphere Technology

#### 1.2.2.1 Principles

In the latter half of this century, plant physiological research has led to a firm understanding of the processes involved in post-harvest senescence and decay and has led to the development of technologies aimed at slowing these processes. The principal cause of produce deterioration, given sanitary conditions, is respiration - the oxidative breakdown of complex molecules. Traditionally, cooling has been used to slow respiration. Aerobic respiration rates may be further reduced by limiting the available  $O_2$  and controlling  $CO_2$ levels (Phan, 1975; Bartsch and Blanpied, 1984). Storage in such atmospheres is referred to as controlled atmosphere (CA) storage.

Typical CA's consist of gas composition ranges of 0 to 10%  $O_2$ , 0 to 30%  $CO_2$  (Kader, 1985; Salveit, 1985; Meheriuk, 1985). The balance is  $N_2$  and a variety of trace gases evolved by the produce and resident micro-organisms (Ouelette et al., 1990). Optimal

gas compositions depend on the biochemical and physiological characteristics of the produce. Excessively high  $CO_2$  levels cause injury to many varieties of fruit (Lidster, 1990) while excessively low  $O_2$  concentrations may induce anaerobic respiration and subsequent damage (Phan, 1975).

CA storage combined with refrigeration and high relative humidity offers several advantages to conventional cold room technology. These are: 1) reduction of respiration and transpiration rates, 2) prevention of chlorophyll degradation, 3) retardation of softening and firmness retention, 4) reduction of post-harvest pathogenic activity (Bohling et al., 1977; Smock, 1979), and 5) lower refrigeration costs due to reduced respiration and heat evolution. CA storage has been very successful in extending the storage life of, in particular, pome fruits (Meheriuk, 1985), and cabbage (Raghavan *et al.*, 1984 and Gariepy et al., 1984).

In a commercial setting, CA storage consists of two phases: i) the period between loading and establishment of optimal CA, or  $O_2$  pull-down period, and ii) long-term storage period at optimal conditions. While most of the research and development of CA systems has focused on determining and maintaining optimal conditions for various commodities, recent work on apples has shown that there are advantages to reducing the  $O_2$  pull-down period. Sharples and Munoz (1974) found that a pull-down exceeding 7 days resulted in significantly poorer results for Cox's Orange Pippin apples. Lau et al. (1983) showed that apple firmness loss can be reduced if the pull-down to 2.5%  $O_2$  is achieved within three days of storage room sealing. Bartsch (1986) noted that the best results are obtained if the room is flushed to 5%  $O_2$  within five days. These pull-down periods are much shorter than the 20-day period generally associated with natural pull-down by respiratory activity alone (Anon., 1980, 1981).

In principle, rapid establishment of CA is desirable; however, the sensitivity of commodities to post-harvest storage conditions is quite variable, as are unit weight market values, production levels and marketing distances. These factors weigh heavily in the decisions of whether to adopt CA storage, what type of CA system to be used, whether rapid  $O_2$  pull-down is necessary and whether to use an  $O_2$  or a  $CO_2$  control system. A summary of available technologies is presented in the following sections.

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1.2.2.2 Oxygen Control Systems

If product respiration does not reduce the oxygen level fast enough, one of the following methods should be considered: 1) external burners, 2) liquid or gaseous nitrogen, and 3) gas separator systems.

External burners remove the oxygen through combustion with propane or natural gas to produce a mixture of carbon dioxide and water vapour which is cooled and fed to the storage room. There are two types of burners: 1) the open flame burner, which uses outside air as the air supply and releases the low oxygen exhaust into the CA room, and 2) the catalytic burner, which is usually preferred since it allows for continuous complete combustion with a catalyst, bringing the  $O_2$  to levels as low as 3%.

External burners are inexpensive. The disadvantages are: a) the production of large quantities of carbon dioxide and ethylene, b) the production of effluent gas that is hot, and c) the explosion hazard associated with the combustion process. Consequently, scrubbing and cooling are needed concurrently with combustion.

Liquid or gaseous nitrogen are also an effective means for establishing rapid CA conditions. The quantity of  $N_2$  required to bring the  $O_2$  level down is a function of the desired  $O_2$  concentration and the standard free volume of the storage room. Liquid nitrogen is sprayed into the storage room via spray headers placed in front of the evaporator fans (Bartsch, 1986), thus pulverizing the  $N_2$ . The injection of liquid nitrogen in the storage room provides some refrigeration, but the extra cost for insulating the supply lines to the room and the potential freezing burns to the fruit may offset refrigeration savings (Waelti and Cavalieri, 1990). Gaseous nitrogen could be introduced in the storage room until the desired atmosphere has been reached.

This method of oxygen pull-down is non-explosive, and does not introduce carbon dioxide or ethylene gases. The method has the disadvantage of being expensive unless onsite nitrogen is available.

Three types of gas separators are available on the market. These are: i) the pressure swing adsorption system (PSA) and ii) the hollow fibre membrane separator system (HFMS) and iii) ammonia cracking at high temperature. They can also be used to control the  $CO_2$  level, but other  $CO_2$  scrubbing systems are usually preferred (Bartsch and Blanpied, 1988).

Most PSA systems work on a similar principle (Anon., 1987). A stream of air is compressed, purified by filters and pushed through a molecular sieve which selectively absorbs  $O_2$ . The gas stream that leaves the gas separator mostly contains  $N_2$ . The  $O_2$ retained in the gas separator is released to ambient air by ventilation of the separator. Two sorption vessels mounted in parallel permit the absorption/desorption cycle to take place without affecting the flow of  $N_2$  rich stream. The purity of the  $N_2$  gas output can vary between 90 and 99.9%, depending on pressure, air flow circulating through the PSA and gas temperature. PSA's have proved successful in apple CA storage (Bartsch and Blanpied, 1988). They require air free of oil and water, and the system requires periodic mechanical checks. Initial capitalization is fairly high.

The hollow fibre membrane system is based on different permeation rates for the gases passing through the membrane. Compressed heated air is filtered and forced into a hollow fibre membrane chamber.  $O_2$  and  $CO_2$  gases are separated from the  $N_2$  by their higher permeabilities. The stream rich in  $O_2$  and  $CO_2$  is vented to ambient while the nearly pure  $N_2$  gas is fed to the storage room to purge the  $O_2$  and establish the desired gas concentration. HFMS has been used successfully in apple CA storage (Bartsch and Blanpied, 1988). An HFMS has a high initial cost and the replacement cost of a membrane is higher than for the charcoal beads of a PSA system. Like the PSA system, air must be free of both water and oil. There is less maintenance than with a PSA system.

Ammonia cracking is a process in which anhydrous ammonia is split at high temperature into nitrogen and hydrogen gas. Store room air is circulated through this system and oxygen reacts with the hydrogen to form water. The return air consists of nitrogen and water vapour free of carbon dioxide and hydrocarbons. The system requires ammonia gas which is a dangerous substance to handle. Operating cost is high and the nitrogen air stream requires cooling before entry into the room (Bishop, 1990).

#### 1.2.2.3 Carbon Dioxide Control Systems

There are five reagents commercially available for removal of excess  $CO_2$  from the CA storage room. They are: 1) caustic soda, 2) hydrated lime, 3) activated charcoal and molecular sieves, 4) membranes, and 5) water. The operation of these scrubbing systems

requires routine measurement of the room atmosphere for  $CO_2$ .  $CO_2$  scrubbing is regulated by the flow through the scrubber.

Caustic soda (NaOH) dissolved in water is one of the oldest  $CO_2$  scrubbing procedures in CA storage (Pflug, 1960).  $CO_2$  level is controlled by adjusting the time of caustic soda solution made available to the chamber atmosphere. Caustic soda has been largely discontinued due to the corrosiveness of the mixture. However, new systems using dry caustic soda are now being introduced and appear to be a viable alternative (Bartsch and Blanpied, 1984).

One of the simplest and most efficient methods of regulating  $CO_2$  levels in a CA room is the hydrated lime  $[Ca(OH)_2]$  scrubber. The scrubber consists of an insulated and airtight plywood box (or other material) externally connected to the CA room. The box contains enough lime for the entire storage period. Lime should be replaced if its efficiency drops. Airflow to the scrubber may be by convection or regulated by blowers and dampers. The CO<sub>2</sub> and hydrated lime react in a 1:1 ratio to form CaCO<sub>3</sub> (limestone) and  $H_2O$ . Dolomitic lime which has a high Mg content is not as efficient as calcitic lime. Since the efficiency of the  $CO_2$  scrubbing by the lime is controlled by the surface area of the lime exposed, the following points should be noted. Lime with a smaller particle size (fine mesh) is more efficient than lime with a larger particle size (coarse mesh). The lime should be packaged in 25 kg bags which have no polyethylene liner. To maximize air circulation the bags should be stacked on a pallet with a 10 cm space between layers. Since it has been observed that less than 20 % of the lime in a 25 kg bag may be used because the outer surface hardens during the reaction, more efficiency is obtained if each bag is only partially filled, e.g. 50 %. The lime requirement depends on the storage To keep CO<sub>2</sub> below 2 %, approximately 12 kg of lime/ton of apples is duration. recommended for each 3 to 4 months of storage. Another common practice is to place about 50 % of the lime recommended for the anticipated storage period inside the storage room. The bags can be placed on pallets on the floor or in single layers on pallets placed on the uppermost bins.

Activated charcoal and molecular sieve units consist of a container filled with the absorbent material (activated charcoal or molecular sieve), two blowers and four timer controlled valves. The operation requires two consecutive steps; first, air from the CA room passes through the scrubber where  $CO_2$  is absorbed. Second, the saturated absorbent is reactivated by circulating outside air through the scrubber. When a molecular sieve is used, a heater is required to increase the temperature of the absorbent during the reactivation process. Both systems have low operating costs and absorbers are replaced every five years. These systems, like the water scrubber, return some  $O_2$  to the room because fresh outside air is allowed into the scrubber during the re-activation process.

When fruits and vegetables are packaged in polymeric films, the atmospheric composition is affected by product respiration and by the gas permeation through the membrane. This is a common practice in retail stores and about 40% of the produce is now marketed in such packages (Desrosiers and Desrosiers, 1977). Composition, structure thickness and surface area of the package, as well as temperature and partial pressure difference between the outside and the inside of the package, are the main parameters contributing to the gas exchange across the package envelope which acts as a semi-permeable membrane. Membranes use the concept of selective permeation to separate gases. Each gas has a characteristic permeation rate that is a function of its ability to dissolve and diffuse through a membrane.

Marcellin and Leteinturier (1967) have extended the use of the semi-permeable membranes to large pallet loads and storage rooms. The system creates a CA condition by means of a series of rectangular bags of silicone rubber connected in parallel. Bag number depends on the size of the cold room and on the respiration behaviour of the stored commodity. These units can be installed inside or outside the cold room. When the unit is located outside the cold room (exposed to ambient air), the CA circulates inside the silicone rubber bags. Analysis of the CA composition indicates whether enough bags are in use. To maintain a CA of 5% CO<sub>2</sub> and 3% O<sub>2</sub>, 50 m<sup>2</sup> of silicone membrane are required per 100 tons of fruit when the standard free volume is about 2.9 to 3.6 (Raghavan and Gariépy, 1984). Although this system was initially designed for apples and pears, it has potential for storage of vegetables such as cabbage, leeks, celery and rutabaga (Gariépy et al., 1984).

A modified version of the Marcellin system consists of gas diffusion panels enclosed in an airtight metallic container with two separate air flow paths and a control unit (Raghavan et al., 1984). The gas diffusion panels are made of square frames on which the

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silicone membrane is fixed. They are banked side by side in an airtight arrangement such that they permit the outside air and the cold room air to flow on opposite sides of the membrane without any mixing. Circulation, of both room and outside air, in the diffuser is maintained by centrifugal blowers. Their operations are controlled by a timer on a percentage basis of a complete cycle. Although this system is relatively new in Canada, it has been used successfully in commercial cabbage facilities since 1982.

There are two types of water scrubbers, the brine  $CO_2$  scrubber (Palmer, 1959), and the modified system of Smock *et al.* (1960). In the brine scrubber, brine-water is pumped over the evaporator coils, where it absorbs the  $CO_2$ , and is gravity-fed to a water reservoir located outside the CA room. The water is then pumped from the reservoir to an aerator where the  $CO_2$  is released to the outside air. The corrosion problem resulting from the use of salt water can be avoided when dry evaporator coils are used. The modified system uses two aerators: one located outside and one located inside the storage room. The two water systems efficiently control  $CO_2$  levels and also increase the relative humidity levels inside the storage room. The  $CO_2$  level is maintained by adjusting the water flow rate through the aerators and by the operating time. Since the respiration rate may be higher at the beginning of storage, Bartsch and Blanpied (1984) recommend that some hydrated lime be placed inside the CA room, to help assist control of  $CO_2$  levels. The combination of hydrated lime and water scrubber is a less expensive alternative to a water scrubber having the capacity to remove the large quantities of excess  $CO_2$  respired at the early stages of storage.

The CO<sub>2</sub> absorption system for the CA storage room should be designed on the basis of quantity of commodity in the CA room, the CO<sub>2</sub> production rate of the commodity and the CO<sub>2</sub> level required. Pflug (1960) recommended 100 l of water per hour per ton of apples, for 5% CO<sub>2</sub>, 3% O<sub>2</sub> at a storage temperature of 1°C. This gives a scrubbing capacity of 0.02 m<sup>3</sup> of CO<sub>2</sub> per m<sup>3</sup> of water circulated through the scrubber.

To avoid freezing, water scrubbers should be used in rooms that are held above  $0^{\circ}$ C. However, when the water is aerated outside the room to release CO<sub>2</sub>, it absorbs O<sub>2</sub> which is then introduced into the room. This problem may be overcome in an HPWS as will be described in Chapter 3.

#### 1.2.3 Mass Transfer at a Gas-Liquid Interface

The operational basis of a water scrubber is the mass transfer of gases at a gasliquid interface. In a conventional water scrubber the interface is at the surface of water drops sprayed through the storage air (Fig. 3.1) while in an HPWS the interface is at the bubble surface. Both gas and liquid properties are involved. The diffusion coefficient, D, of a gas at infinite dilution in water is temperature-dependent but varies only slightly with pressures below 10 atm. (Cussler, 1984). In a CA context, temperature may be assumed to be constant and pressure effects may be neglected since compression does not exceed 10 atm. The governing diffusion equation is therefore Fick's Second Law with D constant (Crank, 1975):

$$\frac{\delta C}{\delta t} = D \frac{\delta^2 C}{\delta x^2}$$
(3.1)

from which the mass transfer through the surface under steady-state conditions may be derived:

$$M = \frac{-A \ D \ \Delta C \ \Delta t}{\Delta x} \tag{3.2}$$

The main factors involved are therefore: the quantity of gas in water, the diffusion coefficient, the contact time and bubble dimensions.

Diffusion coefficients of gases at infinite dilution in water at 25°C are given in Cussler (1984). In the design of the water scrubber, the gas solubilities in water must also be known and are given for various temperatures and 1 atm. pressure in Hodgman et al. (1954).

The contact time depends on air bubble movement through a water column and therefore on bubble dimensions. The theoretical development and empirical modifications leading to solution of the mass transfer equation during operation of the HPWS are elucidated in Chapter 3.

#### **1.2.4 Bubble Generators**

The role of bubble parameters in the modelling of mass transfer during HPWS operation was elucidated in the preceding section. In the design process, the choice of bubble generator is therefore important. Moreover, the size distribution of bubbles produced by a given generator should be known. In this section, methods of bubble generation are reviewed. The problem of measurement is discussed in the next section.

The mechanism of bubble formation has been a subject of interest for several decades (Hadamard, 1911) and many techniques for production of microbubbles have been developed. Among these are: multistage bubble fractionation (Leonard and Blacyki, 1978), cavitation by ultrasound (Aymé and Carstensen, 1989), electrostatic atomization (Sato, 1980) and micro-bead packed columns (Bowley and Hammond, 1978).

A simpler approach has been the injection of gas into the liquid through a porous material made by sintering or bonding fine particles of carbon, ceramic, polymer or metal. Such materials can produce bubbles of diameter of the order of  $100\mu$ m or more and can be used as foam generators or for gas dispersion. The size of bubbles produced depends on pore size and the imposed pressure drop. The dispersive effectiveness depends on pore size, structure and surface roughness of the material, the liquid medium and the depth, turbulence and coalescence of the gas bubbles (Fair et al., 1984). Alternatively, one may inject gas into the liquid through capillary tubes of various inside diameters and adjust capillary tube angles to obtain different bubble sizes (Datta et al., 1950).

#### **1.2.5** Determining Bubble Dimensions

Several experimental techniques have been used to measure bubbles. The oldest method was to measure the liquid displacement caused by a given number of bubbles (Haberman and Morton, 1953). More sophisticated methods fall into two categories depending on whether the sensors used are internal or external (Lim *et al.*, 1990). Internal sensors include miniature capacitance/resistance probes, optical probes and static pressure probes. Since these sensors make direct contact with the bubbles bubble behaviour is altered to a greater or lesser degree. Moreover, the characterization of the bubble size and shape from the measurement of the perceived bubble lengths requires extensive data interpretation and calibration. External sensors include X-ray,  $\gamma$ -ray, laser and camera

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systems. These techniques provide good visual observation, thereby improving the qualitative understanding of bubble behaviour (Pal et al., 1988). Novel techniques have also been developed. Image analysis has been classed as one of the best (Tsukada and Horio, 1990).

The development of real-time image analysis, has facilitated the measurement of key bubble parameters in two-dimensional beds (Lim et al., 1990). There is a lack of information needed for analysis in three dimensional setups. Bubble contours may be traced by means of a chain-coding algorithm (Reid and Searcy, 1989) and volumes computed using the First Pappus Theorem (Ayres, 1972). Bubble velocity may also be determined using the difference between bubble position on successive scans. While real-time image analysis is a powerful tool, it has some limitations associated with depth of field in the three-dimensional situation and when dense bubble clouds are the subject of interest.

#### 1.2.6 Measuring Gas Solubility

Accurate knowledge of equilibrium air-liquid partitioning coefficients, or gas solubility, is essential to a variety of applications such as the design of air stripping towers or water aeration processes involved in environment control processes. Current methods for measuring gas solubility usually involve batch air stripping in a bubble column as proposed by Mackay *et al.* (1979). This technique is popular because it only requires the measurement of relative concentrations so that the preparation of standard curves for determining exact concentrations is unnecessary. This technique is, however, limited; equilibrium is assumed to be reached between the gas concentration in the liquid and in the bubbles, and the time available for reaching equilibrium is set by the height of the column.

The EPICS technique (Equilibrium Partitioning in Closed Systems) was developed by Lincoff and Gossett (1984) for gas mixtures. This technique requires only the measurement of relative concentrations of the gas in one phase (gas or liquid). Since this technique uses closed systems which may be equilibrated indefinitely, it is free of mass transfer limitations.

These two techniques require gas chromatography or other gas analysis methods to determine the relative gas concentrations in either or both the gas and the liquid phases. The standard method described in ASTM (1990) has the advantage of avoiding the

utilisation of gas analysis method but involves hazardous operations, equipment and materials such as mercury. This method is also fairly tedious and requires expensive equipment.

Theoretically, pure gas solubility may be measured indirectly by monitoring the pressure drop caused by gas solubilization in a pressurized closed container. In 1803, William Henry observed that the amount of gas that dissolves in liquid at a given temperature is directly proportional to its partial pressure (Gerrard, 1980). Henry's Law is expressed by Eq. 3.3.

$$C_l = \frac{P_{eq}}{H} \tag{3.3}$$

Where  $C_l =$  liquid phase solute concentration at equilibrium, mole of gas•L<sup>-1</sup> of liquid;

H = Henry's constant of the solute, L of liquid•kPa•mole<sup>-1</sup> of gas;

 $P_{eq}$  = partial pressure of the gas at equilibrium, kPa.

There are theoretical reasons for employing mole fraction, but there are no universally accepted units for expressing the solubility of a gas and this unit is not convenient for general use. Since the solubility of a gas is usually determined by measuring the dissolved volume, rather than the dissolved mass, units of volume of gas per volume of liquid are more frequently used (Glasstone, 1946). Eq. 3.4 can be used to transform the  $C_l$  to solubility expressed in terms of volume of gas per unit of volume of liquid.

$$S = \frac{C_l}{\rho} = \frac{P_{eq}}{\rho H}$$
(3.4)

Where S = gas solubility,  $L_s \bullet L^{-1}$  of liquid;

p

= specific gravity of the gas, mole  $L_s^{-1}$ .

The unit  $L_s$  is the standardized litre of gas, that is, the volume occupied by the a mass of gas at 0°C (273.15°K) and 101.3 kPa absolute pressure. The conversion from L to  $L_s$  is performed using the perfect gas law.

The solubility of gas in liquid is inversely proportional to the temperature. Published data of gas solubility in distilled water (Weast and Astle, 1979) for the three gases involved in the separation process of the HPWS will be presented in a later Chapter.

#### 1.2.7 Summary

CA storage is recognized as a viable technique to extend the storage life of fruits and vegetables and potentially guarantee year-round fresh supplies. Rapid achievement of optimal CA conditions is an essential aspect of process optimization for certain commodities. Existing techniques of Rapid  $O_2$  pull-down have not been found to be commercially viable. Theoretically, an HPWS offers a solution which is the topic of the third Chapter.

The development of an efficient HPWS depends on optimization and control of the mass transfer of gases in water, an available and inexpensive substance. The key physical factors involved in HPWS design are bubble dimensions and gas solubility. The measurement of both are important to the design process and improvements in the cost-efficiency of these measurements should reduce the overall research and development costs of a commercially-viable HPWS.

#### **II GENERAL OBJECTIVES**

The main objectives of this research were:

- A) To demonstrate the theoretical feasibility of reducing the  $O_2$  pull-down period in CA storage using an HPWS and to single out the principal parameters affecting the performance of a such a system.
- B) To develop an HPWS and monitor its performance with appropriate instruments.

C) To test an HPWS and demonstrate the possibility of using this system for Rapid  $O_2$  pull-down in CA storage by minimizing the amount of  $O_2$  entering the storage while scrubbing the  $CO_2$ .

The secondary objectives of this research were:

- A) To develop and test a non-intrusive system for measuring bubble size.
- B) To develop a discriminatory parameter permitting the characterization of a cloud of bubbles in a 3-D system.
- C) To characterize available air diffusers and determine the most suitable diffuser for use in an experimental HPWS.
- D) To develop a simple and inexpensive method of measuring pure gas solubility in pure and contaminated water.

## III THEORETICAL MODEL OF A HIGH PRESSURE WATER SCRUBBER FOR CONTROLLED ATMOSPHERE STORAGE

#### 3.1 Introduction

Respiration is the metabolic process defined as the oxidative breakdown of complex materials such as sugars into  $CO_2$ , water and energy. It is possible to slow down this destructive activity to a certain degree by cooling and to a greater degree by also controlling the atmospheric composition of the storage space. A storage system which controls gas concentrations and temperature is referred to as controlled atmosphere (CA) storage. CA methodology seeks to minimize  $O_2$  entry and limit  $CO_2$  accumulation subject to the constraints that anaerobic decomposition and  $CO_2$  toxicity are not induced. Optimal compositions vary according to the biochemical and physiological characteristics of the stored produce.

Optimal gas compositions associated with CA range from 0 to 10%  $O_2$ , 0 to 30%  $CO_2$  and the balance in  $N_2$  (Kader, 1985, Salveit, 1985 and Meheriuk, 1985) and trace gases, such as  $CH_4$  and ethylene, produced by the commodity and resident microorganisms. The time between sealing of the storage room and establishment of optimal CA is referred to as the  $O_2$  pull-down period. In general, the maximum  $O_2$  pull-down period is 480 h (Anon., 1980; and Anon., 1981). Although a shorter period is not required by regulations, post-harvest research has shown that, for some commodities, a shorter  $O_2$  pull-down period results in better post storage quality.

Lau *et al.* (1983) showed that apple firmness loss can be decreased using a rapid  $O_2$  pull-down (2.5%  $O_2$  or less within the 2 to 3 days after sealing the room) compared to a 20 day  $O_2$  pull-down period. In general, the best results are obtained when the room is filled, cooled, and flushed to 5%  $O_2$  in no more than five days (Bartsch, 1986). Although rapid  $O_2$  pull-down may be achieved by a number of methods, such techniques have yet to be used commercially due to high initial investments and/or high operating costs, depending on technique.

The objective of this study is to demonstrate the theoretical feasibility of reducing the  $O_2$  pull-down period in CA storage with a high pressure water scrubber that minimizes

the amount of  $O_2$  entering the storage while scrubbing excess  $CO_2$ . Furthermore, this study aims at demonstrating that, at high pressure, there is a net transport of  $O_2$  out of the CA storage.

#### 3.2 Theory and Design

According to Sive and Reznisky (1977), credit must be given to Palmer of Hilton, New York, who, in 1959, noticed that his brine spray unit was absorbing  $CO_2$  and then releasing it by aeration on its return to the brine tank. Smock *et al.* (1960) improved upon the brine spray unit by using dry evaporator coils to prevent corrosion and Pflug (1960) adapted the engineering and design base for construction of water scrubbers. Pflug preferred a separate absorption system to a combined evaporator-absorption unit.

The description of each component of a simple tower design with full jet spray nozzle for water scrubbing was presented by Smock *et al.* (1960). In the water scrubber shown in Fig. 3.1, the water is circulated through the two sprayers and is used as the medium to transfer the  $CO_2$  out of the storage room. As  $CO_2$  is soluble in water, it diffuses into or out of the water drop to reach equilibrium with its saturation capacity in water. The  $CO_2$  is maintained at a higher concentration inside the storage room than in the ambient air. Thus its concentration at saturation is higher in the water exhausting from the room than in the water entering into the room. This results in a net outflow of  $CO_2$  from the room. However,  $O_2$  and  $N_2$  are also soluble in water and the flow of these two gases in the scrubbing medium must also be considered. To improve the performance of a  $CO_2$  water scrubber, the basic principles of diffusivity and solubility of gases in water must be well understood.

#### **3.2.1 Diffusion Phenomena**

The theory of diffusion (Fick's first law of diffusion) in an isotropic medium states that the flux through a plane of unit area is proportional to the concentration gradient normal to the plane as expressed in Eq. 3.1.



Figure 3.1: Diagram of a conventional  $CO_2$  water scrubber.

$$F = -D \frac{\partial C}{\partial x}$$
(3.1)

The fundamental principle of conservation of substance within a finite volume element V, at any given instant may be stated as follows (Mikhailov and Ozisik, 1984):

$$\begin{vmatrix} Rate & of \\ substance \\ accumulation \\ in V \end{vmatrix} = \begin{vmatrix} Rate & of & substance \\ passing & through \\ the & bounding \\ surface & of V \end{vmatrix} + \begin{vmatrix} Rate & of \\ substance \\ generation \\ in V \end{vmatrix}$$
(3.2) T  
he  
in V he

ndamental diffusion equation in an isotropic medium may be derived from Eqs. 3.1 and 3.2. When diffusion is one-dimensional (gradient of concentration along the x-axis only) and the rate of substance generation (last term on the right hand side of Eq. 3.2) is 0, the diffusion equation (referred to as Fick's second law of diffusion) takes the following form for D constant (Crank, 1975):

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$$
(3.3)

Under steady state conditions, the mass transfer through a surface can be derived from Eq. 3.3 and rewritten as follows:

$$M = \frac{-A \ D \ \Delta C \ \Delta t}{\Delta x} \tag{3.4}$$

The size of the water drops, the diffusion coefficient, the quantity of gas in water at saturation and the contact time are the main factors involved in the scrubbing process. The diffusion coefficient is a measure of the response of a gas to a variation in concentration and is specific to each gas and medium involved. The diffusion coefficient is affected by temperature and by pressures higher than 10 atm. (Cussler, 1984). Since the temperature is fairly stable in CA storage, and pressure does not exceed 10 atm., the diffusion coefficient is considered constant in a water scrubber. The diffusion coefficients of the three gases involved in the process are presented in Table 3.1.

Table 5.1.	1984).		
	Gases	Diffusion Coefficient $(10^{-9} \text{ m}^2 \text{ s}^{-1})$	
<u> </u>			

2.00 2.10 1.92 1.88

Air O<sub>2</sub> CO<sub>2</sub> N<sub>2</sub>

Table 3.1:	Diffusion coefficients of gases at infinite dilution in water at 25°C (Cussler,
	1984).

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#### 3.2.2 Solubility Of Air Components In Water

Accurate knowledge of gas solubility in water is essential to the design of a water scrubber. In 1803, William Henry observed that the amount of gas that dissolves in water at a given temperature is directly proportional to its partial pressure (Gerrard, 1980). The constant of proportionality between the gas partial pressure and the gas concentration in a liquid at equilibrium is now known as "Henry's Constant" and can be expressed by Eq. 3.5 (Lincoff and Gossett, 1984):

$$C_l = \frac{P_s}{H} \tag{3.5}$$

The solubility of gas in water is inversely proportional to the water temperature. It is therefore advantageous to maintain the water temperature as low as possible (just above  $0^{\circ}$ C) since more gas may be dissolved and the air exhausting from the gas separator should be cold. Solubilities of the three gases involved in the water scrubber are presented in Table 3.2 (Hodgman *et al.*, 1954).

#### 3.2.3 Conventional Water Scrubber

The time required to saturate a water drop with  $O_2$  and CO2, as would occur in a conventional water scrubber, was calculated as a function of the diameter of a drop (Fig. 3.2) by applying the diffusion equation (Eq. 3.3). From this time requirement and the fluid mechanics theory describing the terminal velocity of a water drop in air (Clift *et al.*, 1978), the minimum height required to saturate the water drop falling in a still air condition was calculated as a function of its diameter (Fig. 3.3). As the upstream air velocity increases, the minimum height decreases. For small drop diameter, the required height to reach saturation is very similar for both gases and is less than the height usually encountered in a commercial water scrubber system. In order to saturate water with a gas, water drops should be small, but not small enough to be swept by the air stream. The water scrubber capacity is therefore only a function of the water flow and the gas concentration in the water at saturation.

The water scrubber capacity may be increased by increasing either the water flow
Table 3.2:	Gas solubilities in water under normal pressure and at different temperatures
	(Hodgman <i>et al.</i> , 1954).

Gases	Solubility (L/L) at temperature (°C)			
	0.0	5.0	10.0	15.0
O <sub>2</sub>	0.049	0.043	0.038	0.034
CO <sub>2</sub>	1.713	1.424	1.194	1.019
N <sub>2</sub>	0.023	0.020	0.018	0.016



Figure 3.2: The time required to reach gas saturation of the water drops calculated as a function of the diameter of drop.



Figure 3.3: Falling height required to saturate water drop with gas as a function of their diameter in a still air condition.

or the gas concentration in the water at saturation. When the water flow is increased the  $CO_2$  transport increases along with those of  $O_2$  and  $N_2$ . For a given drop diameter,  $O_2$  and  $CO_2$  require approximately the same height to saturate water; the ratio of  $O_2 / CO_2$  transport is determined by the partial pressure of these gases inside and outside of the room and by their solubility in water. It is possible to change this ratio by increasing or decreasing the pressure of the gases; the quantity of a soluble gas contained in water at saturation is a function of its solubility and its partial pressure (Eq. 3.5). The  $O_2$  transport can then be reduced by increasing the pressure of the air in contact with the water spray inside the storage room. This would also have another positive effect since the  $CO_2$  transport capacity of the water would rise, thus reducing the necessary water flow rate for  $CO_2$  extraction. However, increasing the storage room pressure is not practical.

# 3.2.4 Theoretical High Pressure Water Scrubber

Although it is not feasible to increase the pressure in the storage room, it is possible to draw storage room air into a compressing unit and then to diffuse the compressed air as bubbles through a water column outside the storage room. The schematic representation of this idea, the high pressure water scrubber (HPWS), is presented in Fig. 3.4. A compressor is used to pressurize the air coming either from the storage room or from the outside atmosphere and to introduce it into the water stream. There, the air bubbles begin to rise to reach the top of the water column. During their rise,  $CO_2$ ,  $O_2$  and  $N_2$  are absorbed by the water at rates depending on various factors. At the top of the water column, the air is returned to the storage room. During this time, the water is pumped from a reservoir to the top of the water column. As the water flows down to the bottom of the column it absorbs some gases to equilibrate with their partial pressures, and then flows to the water reservoir. Since the water is under atmospheric pressure, it releases or absorbs the three gases to equilibrate at a new condition.

By analyzing the factors which affect the gas diffusion through the air-water interface, one can simulate the performance of a high pressure water scrubber. The rising movement of the air bubble in water creates an internal motion. This spherical vortex is a function of the interfacial tension between water and air, the viscosity ratio of these two fluids, the size and the speed of the bubble. Purity of water is also quite important since



Figure 3.4: Diagram of high pressure water scrubber using gas solubilities in water to separate the gases.

small amounts of surface-active agents can cause the formation of a "skin" around the bubble that effectively prevents gas diffusion between the water and the air bubbles. In an HPWS, it might be important to choose conditions which cause internal motion and thus get uniform internal gas composition due to the mixing effect.

### 3.2.5 Air Movement In Water

Considering a spherical air bubble moving in pure water such that the interface is completely free of surface-active contaminants and the interfacial tension is constant, and assuming that both Re and Re<sub>b</sub> are small (i.e. Re  $\leq 0.1$ ), Eq. 3.6 can be applied to both fluids (Clift *et al.*, 1978).

$$E^4 \psi = E^4 \psi_{\rm b} = 0 \tag{3.6}$$

where  $E^2$  may be written in spherical polar coordinates as:

$$E^{2} = \frac{\partial^{2}}{\partial r^{2}} + \frac{\partial^{2}}{r^{2} \partial^{2}} - \frac{\cot \theta \partial}{r^{2} \partial}$$
(3.7)

Taking a frame of reference attached to the air bubble with the origin located at the centre of the bubble, the boundary conditions are the following:

1) Uniform stream flow in the z- direction at large distances from the sphere:

$$\Psi_{(as \ r \ - \ \infty)} = \left(\frac{v \ r^2}{2}\right) \sin^2 \theta \qquad (3.8)$$

2) No flow across the interface:

$$\psi = \psi_{b} = 0$$
, at  $r = a$ ; (3.9)

3) Continuity of the tangential velocity across the interface:

$$\frac{\partial \Psi}{\partial r} = \frac{\partial \Psi_b}{\partial r}, \quad at \ r = a; \qquad (3.10)$$

4) Continuity of the tangential stress across the interface:

$$\frac{\partial}{\partial r} \frac{\partial \Psi}{r^2 \partial r} = \kappa \frac{\partial}{\partial r} \frac{\partial \Psi_b}{r^2 \partial r}, \quad at \ r = a; \qquad (3.11)$$

where  $\kappa = \mu_b / \mu$  is the viscosity ratio.

5) Continuity of the normal stress across the interface:

$$p_{f} - 2\mu \frac{\partial}{\partial r} \left( \frac{1}{r^{2} \sin \theta} \frac{\partial \psi}{\partial \theta} \right) + \frac{2\sigma}{a} = p_{b} - 2\mu_{b} \frac{\partial}{\partial r} \left( \frac{1}{r^{2} \sin \theta} \frac{\partial \psi}{\partial \theta} \right)$$
(3.12)

where the term  $\sigma/a$  is the pressure increment associated with interfacial tension. The solutions of the Eq. 3.6 when  $p_{0b} - p_0 = 2\sigma/a$ , is presented by Clift *et al.* (1978), (Eqs. 3.13 and 3.14).

$$\psi = -\frac{v r^2 \sin^2 \theta}{2} \left( 1 - \frac{a (2 + 3 \kappa)}{2 r (1 + \kappa)} + \frac{\kappa a^3}{2 r^3 (1 + \kappa)} \right)$$
(3.13)

$$\psi_b = -\frac{\nu r^2 \sin^2 \theta}{4 (1 + \kappa)} \left(1 - \frac{r^2}{a^2}\right)$$
(3.14)

Eq. 3.14 gives the air motion within the bubble. The pressure distribution at the bubble surface  $(p_f)$  is given by Eq. 3.15.

$$p_f = p_0 + \frac{\mu v_{\infty} a \cos\theta}{r^2}$$
(3.15)

This pressure distribution is a function of  $\theta$ , so that the bubble experiences a net pressure force or drag force. Integration of this pressure, the shear stress and the normal stress over the surface of the bubble leads to the overall drag coefficient (f) expression:

$$f = \frac{16}{Re} \tag{3.16}$$

The terminal velocity  $(v_{\infty})$  of a bubble flowing in water in these conditions can be obtained

by equating the total drag to the net buoyancy force (F<sub>s</sub>):

$$F_s = \frac{\pi D_l^3 \Delta \rho g}{6} \tag{3.17}$$

$$v_{\mu} = \frac{2 g a^2 \Delta \rho}{6 \mu} \tag{3.18}$$

The maximum bubble diameter to which Eq. 3.13 and 3.14 apply is 0.06 mm corresponding to Re = 0.1. As the diameter of the air bubble increases, the shape of the bubble changes, Re increases and the solution presented above does not apply any more.

Furthermore, the terminal velocity of air bubbles in water is affected by the water purity and the bubble shape. Experimental terminal velocities of air bubbles were measured by different authors. Fig. 3.5 shows results presented by Clift *et al.* (1978) and Bird *et al.* (1960) for pure water.

#### 3.2.6 Height Of The Water Column

For bubbles of moderate size (0.5 to 10 mm) with spherical and ellipsoidal shape rising in water with no surface-active agents, the gas in the bubble undergoes a toroidal circulation. As the gas circulates, it encounters fresh liquid at the top of the bubble. As the bubble rises, the liquid moves downward in relation to the bubble and leaves when it reaches the bottom of the bubble. The liquid near the interface is usually in laminar flow and appears to maintain its identity. Thus the liquid behaves much like the liquid at the surface of a falling film (Bird *et al.*, 1960). The contact time is certainly short, so that the penetration of the dissolved gas is slight and the following assumptions are valid:

1) The gases in the air bubble are slightly soluble in water and hence do not change the viscosity of water appreciably;

2) The diffusion takes place so slowly in the water that the gas does not penetrate very far into the water, i.e. the penetration distance is small in comparison with the water thickness. This is particularly true when the size of the water column is large compared to the diameter of the air bubble.





Therefore, for a first approximation, Eq. 3.19 can be used to estimate the instantaneous gas diffusion (Bird *et al.*, 1960).

$$M_{1} = 8 a^{2} \sqrt{\frac{\pi D_{ab}}{t_{exp}}} C_{l}$$
(3.19)

The time exposure  $t_{exp}$  is the time required for the liquid to slide along the bubble from top to bottom. This can be approximated by Eq. 3.20.

$$t_{\exp} = \frac{2 a}{v_{\infty}}$$
(3.20)

Hence the instantaneous gas diffusion through the air-water interface is:

$$M_{1} = \sqrt{(32 \pi a^{3} D_{ab} v_{\infty})} C_{l}$$
(3.21)

The solubility of each gas changes with time since the gas concentrations in the bubble change (Eq. 3.5). The diameter of the bubble and the terminal speed  $(v_{\infty})$  decrease as the gases are absorbed in the water. These facts make the mass transfer a transient process. The total mass transfer through the air-water interface can be obtained by numerical integration:

$$M = \sqrt{(32 \pi D_{ab})} \Delta t \sum_{n=1}^{n=N} \sqrt{(a_{n-1}^3 v_{\infty n-1})} C_{ln-1}$$
(3.22)

The solubility of the gas, the radius of the bubble and the terminal speed are adjusted at the end of each step of the numerical integration. The height of the water column required to absorb the CO<sub>2</sub> gas contained in the bubbles can be calculated from the mass transfer numerical integration. Using Eq. 3.22, data were generated for a 0.5 mm air bubble initially containing 10.5%  $O_2$  at a pressure of 2 atm.. These data are shown in Fig. 3.6. Similar curves can be generated for other conditions needed for the design of the scrubber at known capacities.



Figure 3.6: Ratio of gas concentration at any height of the water column to the gas concentration at height zero plotted for different elevations of the water column for a 0.5mm bubble containing 10.5% of  $O_2$  and subjected to a 2 atm. pressure.

# 3.3 Details for Simulation

The first case used for the simulation of product storage consisted of a room volume of 1818 m<sup>3</sup>. The stored product selected was 410 t of McIntosh apples. The space factor was 3.6 m<sup>3</sup> of air per m<sup>3</sup> of stored product. O<sub>2</sub> and CO<sub>2</sub> levels in the room were 2.5% and 5% respectively. The room was assumed to be airtight and the ventilation was operated as required to maintain the O<sub>2</sub> level at 2.5%. The air and water circulation rate through the water scrubber were 16.7 1 s<sup>-1</sup> and were the same for each case studied.

The simulation was performed to evaluate the theoretical behaviour of a high pressure water scrubber using data on apple respiration rate. The simulation is based on the work of Pflug (1960) but did not assume equal  $O_2$  and  $CO_2$  absorption and desorption coefficients. The factors considered for this simulation are listed in Table 3.3. Using the respiration rate data presented by Pflug (1960):

$$r_1 = 45.5 - 30.7 (1 - 0.46^{t/5}) \tag{3.23}$$

it was possible to conduct a mass balance on the storage room. The performance of a proposed HPWS system was compared with a conventional water scrubber system and a hybrid HPWS and  $N_2$  flushing system in order to find out whether there is any advantage with respect to  $O_2$  pull-down period.

#### 3.4 Results and Discussion

The results obtained for the three systems are presented in Fig. 3.7. The first system simulated a conventional water scrubber that operates when the  $CO_2$  concentration inside the storage room exceeds the desired level (5%). The room is ventilated using ambient air when  $O_2$  is lower than 2.5%. The simulation shows that it takes 417 hours from the time of sealing of the storage room to reach 2.5%  $O_2$ . This corresponds to the  $O_2$  pull-down period encountered in commercial apple storage facilities using commodity respiration to decrease the  $O_2$  concentration inside the room.

The Hybrid HPWS and  $N_2$  flushing system uses an HPWS to extract the excess  $CO_2$  from the storage room. The absolute pressure inside the water column of the HPWS is adjusted from 1 to 8.4 atm. as the  $O_2$  level in the storage room decreases from 21% to

 Table 3.3:
 Factors considered for the simulation of a water scrubber.

Factors	Symbols	Units
Storage room volume	V <sub>1</sub>	m <sup>3</sup>
Amount of product in storage	B <sub>1</sub>	m <sup>3</sup>
Space factor	$Z_1 = V_1/B_1$	m <sup>3</sup> m <sup>-3</sup>
Respiration rate	r <sub>i</sub>	$m^3 O_2 m^{-3} day^{-1}$
$O_2$ level in storage	$\tilde{\mathbf{Y}}_1$	$m^3 O_2 m^{-3}$ of air
$CO_2$ level in storage	C <sub>1</sub>	$m^3 CO_2 m^{-3}$ of air
Leakage or ventilation rate	X1	m <sup>3</sup> m <sup>-3</sup> of air day <sup>-1</sup>
Temperature	$T_1$	°C
$O_2$ in normal atmosphere	$\dot{O_2}$	$m^{3} m^{-3}$
$\overline{CO}_2$ in normal atmosphere	$\tilde{O_2}$	m <sup>3</sup> m <sup>-3</sup>
Apple bulk density	$\tilde{A_1}$	kg m <sup>-3</sup>
Time	t	days
$O_2 / CO_2$ transport ratio	$\mathbf{w}_1$	$m^{-3} O_2 m^{-3} CO_2$
Pressure inside the scrubber	P.	atm.



Figure 3.7: Temporal variation of  $O_2$  concentration inside the storage room for different water scrubbing systems.

2.5%. This permits adjustment of the partial pressure inside the pressurized water column to the outside  $O_2$  partial pressure and thus eliminates any  $O_2$  net transport by the HPWS as  $O_2$  concentration decreases inside the room. As  $CO_2$  above 5% concentration is eliminated, it is replaced by  $N_2$  to maintain the volume of gas inside the room at a constant level. This helps to eliminate the  $CO_2$  without allowing any  $O_2$  to enter and results in a 277 hour  $O_2$  pull-down period.

The third system used in the simulation is represented by the curve for the HPWS at 10 atm. pressure (Fig. 3.7), and consists of continuous operation of the HPWS at 10 atm. absolute pressure from the time the storage room is sealed until the sum of the  $O_2$  and the  $CO_2$  concentrations reaches 10.5%. Respiration by the produce decreases the  $O_2$  concentration to 2.5%. After this period the HPWS operation is based solely on  $CO_2$  concentration. Ambient air is used as the make up gas. This process results in a 140 hour  $O_2$  pull-down period.

With the first two systems, only the commodity respiration is used to reduce the  $O_2$  level inside the room. With the last system, the HPWS is also used to reduce the  $O_2$ . With the system being operated at 10 atm., the partial pressure of the  $O_2$  inside the pressurized water column (Fig. 3.4) is higher than the  $O_2$  partial pressure in the water reservoir and this results in a net  $O_2$  transport out of the room. In that process, the air make up (Fig. 3.4) is also processed through the HPWS before being introduced into the room in order to partially eliminate the  $O_2$  it contains.

Since the operating time of the HPWS is based on the  $O_2$  concentration, the HPWS can be designed to meet any  $O_2$  pull-down requirements. In the present case, the water and air flow through the HPWS were chosen to meet the five day  $O_2$  pull-down period recommended by Bartsch (1986) but those parameters can be adjusted to other specific requirements.

### 3.5 Conclusion

From this investigation, it can be concluded that:

- 1) It is theoretically feasible to reduce the net  $O_2$  transport into the CA room, using a high pressure water scrubber leading to a reduction in the  $O_2$  pull-down period.
- 2) The high pressure water scrubber causes a difference in  $O_2$  partial pressures that

results in a net transport of  $O_2$  out of the CA room.

3) The results justify the construction and testing of an HPWS to eliminate the  $O_2$  after a storage room is sealed and to control the  $CO_2$  during the storage period.

# **3.6** Nomenclature<sup>1</sup>

 $\theta$  = angular coordinate, measured away from front stagnation

 $\kappa$  = viscosity ratio,  $\mu_{\rm b}$  /  $\mu$ 

 $\mu$  = water viscosity, M L<sup>-1</sup> t<sup>-1</sup>

 $\mu_{\rm b}$  = air viscosity, M L<sup>-1</sup> t<sup>-1</sup>

 $\rho$  = specific gravity of water, M L<sup>-3</sup>

 $\rho_{\rm b}$  = specific gravity of air, M L<sup>-3</sup>

 $\Delta \rho$  = specific gravity differential between air and water, M L<sup>-3</sup>

 $\sigma$  = interface or surface tension, M t<sup>-2</sup>

 $\Psi$  = Stokes stream function for water relative to air bubble centre, L<sup>3</sup> t<sup>-1</sup>

 $\Psi_{\rm b}$  = Stokes stream function of the air inside the bubble relative to air bubble centre, L<sup>3</sup> t<sup>-1</sup>

a = the radius of the air bubble, L

 $a_n$  = the radius of the air bubble at the time n $\Delta t$ , L

b, c = empirical parameter

A = area of the surface,  $L^2$ 

 $A_1$  = apple bulk density, M L<sup>-3</sup>

 $B_1$  = volume of product in storage,  $L^3$ 

C = concentration of diffusing substance, M  $L^{-3}$ 

 $c_1 = CO_2$  level in storage,  $L^3 CO_2 L^{-3}$  of air

 $C_1$  = liquid phase solute concentration at equilibrium, M L<sup>-3</sup>

 $C_{ln}$  = liquid phase solute concentration at equilibrium corresponding to the gas concentration at the time n $\Delta t$ , mole L<sup>-3</sup>

<sup>&</sup>lt;sup>1</sup> Units are presented in fundamental dimensions: L = length; M = mass; t = time and T = temperature.

CO <sub>2</sub>	= $CO_2$ concentration in normal atmosphere, $L^3 L^{-3}$
D	= diffusion coefficient of a material through its particular environment, $L^2 t^{-1}$
$\mathbf{D}_{\mathbf{ab}}$	= diffusion coefficient of the material a through the environment b, $L^2 t^{-1}$
$\mathbf{D}_{\mathbf{i}}$	= diameter of the sphere or the air bubble, L
Е	= spherical operator
F	= rate of mass transfer per unit area of section, M $L^{-2} t^{-1}$
f	= drag coefficient
F <sub>s</sub>	= buoyant force the water applies on the air bubble, M L $t^2$
g	= gravitational acceleration, $L t^{-2}$
Н	= Henry's constant of the solute, $L^2 t^{-2}$
М	= total amount of gas diffusing through the surface of the bubble, M
$\mathbf{M}_{\mathbf{i}}$	= average of mass transfer through the bubble surface, M $t^{-1}$
Ν	= total number of steps in the numerical equation
n	= step number in the numerical integration
O <sub>2</sub>	= $O_2$ in normal atmosphere, $L^3 L^{-3}$
$\mathbf{p}_{\mathbf{f}}$	= pressure field due to the flow in the fluid f, i.e. discounting hydrostatic pressure
	variations, M $L^{-1} t^{-2}$
$P_s$	= pressure inside the scrubber, M $L^{-1} t^{-2}$
P <sub>i</sub>	= partial pressure of the solute, M $L^{-1} t^{-2}$
p <sub>o</sub>	= reference pressure, M $L^{-1} t^{-2}$
р <sub>оь</sub>	= reference pressure in the centre of the bubble, M $L^{-1} t^{-2}$
r	= distance from the origin in polar coordinates, L
r <sub>1</sub>	= respiration rate, $L^3 O_2$ consumed, $L^3 t^{-1}$
Re	$= \frac{D_l V_{\infty} \rho}{\mu} , \text{ the Reynolds number}$

 $\operatorname{Re}_{b} = \frac{D_{l} V_{\infty} \rho_{b}}{\mu_{b}}$ , the Reynolds number based on bubble properties

t = time, t

 $t_{exp}$  = time required for the liquid to slide along the bubble from top to bottom, t T<sub>1</sub> = temperature, T

- $v_{\infty}$  = terminal speed of the air bubble, L t<sup>-1</sup>
- $v_{\omega_n}$  = terminal speed of the air bubble at the time n $\Delta t$ , L t<sup>1</sup>
- $V_1$  = storage room volume,  $L^3$
- $w_1 = O_2/CO_2$  transport ratio,  $O_2$  added/CO<sub>2</sub> removed
- x = space coordinates measured normal to the normal plane
- $x_1$  = leakage or ventilation rate,  $L^3 L^{-3}$  of air  $t^{-1}$
- $Y_1 = O_2$  level in storage,  $L^3$  of  $O_2 L^{-3}$  of air

 $Z_1$  = space factor,  $L^3 L^{-3}$ 

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# **CONNECTING TEXT**

The design and construction of the HPWS shown in Fig. 3.4 required a system to generate bubbles at the bottom of a water column. The size distribution of the bubbles generated had to be known to determine the most suitable bubble generator for a given specific application.

The three following chapters present the procedure used to characterize the population of bubbles produced by different bubble generators in a 3-D environment. The first step was to choose an existing method of bubble measurement corresponding to the estimated bubble size and speed. This system involved commercially available components. Its precision was determined and its limitations discussed.

# IV REAL TIME IMAGE DIGITIZING SYSTEM FOR MEASUREMENT OF AIR BUBBLES

#### 4.1 Introduction

The study of the theoretical model of a pressurized  $CO_2$  water scrubber demonstrated the importance of the air bubble diameter in the gas separation process. In a bubbling system, bubbles rise from their source and their gas composition changes due to mass transfer through the gas/water interface. The main parameters affecting mass transfer are bubble volume and velocity. Together, these determine bubble shape and the relevant mass transfer coefficient.

Several experimental techniques have been used to measure bubbles. Two- and three-dimensional systems have been used. In general, the measurement techniques fall into two categories depending on the nature of the sensors (Lim *et al.*, 1990). Internal sensors such as miniature capacitance/resistance probes, optical probes and static pressure probes can be grouped in the first category. Such sensors make direct contact with the bubbles. The intrusive nature of these submersible probes is expected to alter bubble behaviour. Moreover, the characterization of the bubble size and shape from the measurement of the perceived bubble lengths requires extensive data interpretation and calibration. The second category consists of external sensors. The X-ray,  $\gamma$ -ray, laser and photographic methods fall into this category. These techniques provide good visual observation, thereby improving the qualitative understanding of bubble behaviour. Unfortunately, the collection of quantitative data requires subsequent frame by frame analysis of the images. This can be time consuming and laborious unless the procedure is automated.

An adequate system should at least be able to measure characteristic lengths along two perpendicular axes so that both bubble shape and size can be assessed. The measurement of bubble velocity is also useful. The preliminary analysis of the scrubbing system established that bubbles of a diameter ranging from 0.25 to 3.5 mm are required for optimum scrubber performance. Bubbles in this range have a maximum rising velocity of about 330 mm s<sup>-1</sup> (Clift *et al.*, 1978). A vision system that meets the above requirements was selected based on commercially available hardware. This paper discusses the software, calibration and overall accuracy of a digital image analysis system for measuring bubble diameter and velocity.

# 4.2 Materials and Methods

# 4.2.1 Hardware

The experimental set-up is illustrated in Fig. 4.1. A  $200(1) \times 200(w)$  and 400(h) mm aquarium was built from 5 mm glass. It was illuminated on one side by a 75 W incandescent floodlight. Two diffusers assured uniformity of light intensity across the field of view of the monochrome Solid-State Charged-Coupled-Device (CCD) camera mounted on a positioner on the opposite side. The camera positioner allowed movement in the horizontal plane. The entire system, including the light source, the aquarium and the camera, was covered with an opaque curtain to minimize the effect of ambient light on the image.

The camera had a 16.9 mm scanline interlaced transfer CCD. It required a minimum scene illumination of  $0.5 \text{ Im} \cdot \text{m}^{-2}$  at F1.4 for effective monitoring. An electronically variable shutter with 9 settings ranging from 0.02 to 0.0001 s and an Automatic Gain Control circuit provided clear images over an acceptable range of light intensities. The distortion associated with the bubble motion was minimized by setting the electronic shutter at the 0.0001 s. The CCD had a resolution of 768(H) x 493(V) pixels. The video output signal had a horizontal resolution of 570 rows.

A 12.5 to 75 mm zoom lens and a close-up kit on the camera permitted a field of view of 13.5 mm across at a distance from the camera of about 20 cm. An interlaced image was formed by first scanning the odd rows after the shutter had been opened and closed. After an interval of 1/60 s, the shutter was opened and closed again and the even rows were scanned. This way, it was possible to observe, on a single interlaced image, the same moving bubble at two different positions (one on the even rows, one on the odd rows) separated by the distance it travelled during the 1/60 s interval. This feature was useful for velocity measurements.



Figure 4.1 Schematic of the experimental system.

The signal from the camera was digitized using an OCULUS-300<sup>1</sup> board installed in an IBM-AT compatible microcomputer. This system digitized 480 by 512 pixel images at 8 bits of resolution per pixel in real time. The board could store 4 full frames at once, and perform real time operations on images such as image subtraction.

## 4.2.2 Software

Software supplied with the digitizing board contained a set of subroutines and variables that controlled all board functions. These could be called from programs written in C language. An interpreter was also supplied which provided access to pre-programmed basic image analysis routines. However, the time required for image analysis and the flexibility of this interpreter were not suitable for this application. Dedicated software was developed to automate the measurement of bubble parameters which are described in the following sections.

# 4.2.3 Parameter Measurement

Fig. 4.2A shows a plot of grey levels as a function of pixel position for a horizontal line of pixels centred in the field of view. Since the grey level profile was fairly flat, no background subtraction was necessary. Fig. 4.2B shows the same data when a bubble was centred on the line. When light is transmitted through a bubble immersed in water, the light rays near the centre of the bubble are transmitted with marginal attenuation and the rays hitting near the periphery of the bubble are completely reflected. This explains the characteristic shape of the signal displayed on Fig. 4.2B. The signal was large and bubble boundaries were well defined. A simple threshold was applied to discriminate between bubbles and background. Its level was the grey level on Fig. 4.2B where a maximum slope was observed, between maximum and minimum grey levels. A threshold level of 90 was used in all cases, resulting in good image repeatability.

To ensure good repeatability, the image was focused by a technique similar to the

<sup>&</sup>lt;sup>1</sup> Mention of specific trade names does not imply endorsement of those products by Agriculture Canada or McGill University. Similarly, omission of product names is not intended as discrimination against those not mentioned.



Figure 4.2 Grey level along the horizontal centre line of A)background; B) a 2.2 mm diameter bubble centred in the field of view of the camera.

one described by Panneton (1989). This technique maximized the slope of the grey level versus pixel position at a well defined transition perpendicular to a scan line. The slope was calculated and displayed on the computer screen. The camera was adjusted toward the object plane until the slope was maximized.

Once an image was grabbed, the system had to detect whether or not it contained a full bubble. A full bubble was defined as a set of two images of the same bubble which did not intersect the frame boundaries. As stated previously, the first image was formed on the odd numbered rows and the second image on the even rows. The image of the bubble formed on the odd rows appeared below the one from the even rows due to the 1/60 s delay.

Pixels on the even rows along a vertical line in the centre of the field of view were compared to the threshold level starting from row 2, and down. If any of these pixels had a grey level lower than the threshold, the image was discarded and the process was repeated with a new image until a pixel with a grey level lower than the threshold was found. The coordinate of this pixel was then stored to identify the image of the bubble on the even rows. The process was repeated using the odd rows but the searching started one row above the one where an image on the even rows had been found. Only when both an image on the even and an image on the odd rows were identified did the analysis proceed further.

The contour of the bubble was traced by means of a chain-coding algorithm (Reid and Searcy, 1989). It started searching from the pixel used to identify the image on the even rows. The algorithm checked the even row pixels surrounding the current pixel in a clockwise direction, starting with the pixel next to the last unsuccessfully checked pixel. When a "good pixel" (pixel having a grey level below the clipping level) was found, the coordinate of the pixel was stored with the corresponding direction code to form a data point. The direction codes shown on Fig. 4.3 refer to the position of the newly found contour pixel with respect to the current pixel. The contour algorithm was repeated until it returned to the starting pixel, thereby completing the contour of the upper image of the bubble. The contour of the lower image of the same bubble was traced in the same way using the odd row data.

The vertical diameter of the bubble was calculated from its two images. It was the



Figure 4.3 Illustration of the chain-coding scheme.

difference between the maximum and the minimum Y coordinates of the pixels forming the contour of each bubble image. The horizontal diameter was calculated similarly by using the X coordinates.

The total surface (S) of a bubble on an image (projected area of the bubble) was calculated by using the following Eq.4.1:

$$S = 2 \Delta x \left[ \left[ \sum_{i=1}^{n} Y_{i} \right]_{dir=6, 7, 8} - \left[ \sum_{i=1}^{n} Y_{i} \right]_{dir=2, 3, 4} \right]$$
(4.1)

where n is the number of pixels on the contour. In the summation,  $Y_i$  was replaced by zero if the direction code of the i<sup>th</sup> pixel was different from the ones given as subscripts (dir = ...).

The central axis was defined as the vertical axis which divided the surface of a bubble into two equal areas. First, the contour data were sorted in an ascending order of the X coordinates, then the Eq. 4.1 was applied but summation stopped when  $X_{i-1}$  was different from  $X_i$  and the surface being computed was equal to or larger than half the total surface. Linear interpolation was used to locate the central axis at a point between the last two X coordinates.

The volume (V) of the bubble was calculated using the First Pappus Theorem (Ayres, 1972):

$$V = 2\pi\Delta x \left[ \left[ \sum_{i=1}^{n} Y_{i} \left( X_{i} - X_{c} \right) \right]_{dir=6, 7, 8} - \left[ \sum_{i=1}^{n} Y_{i} \left( X_{i} - X_{c} \right) \right]_{dir=2, 3, 4} \right]$$
(4.2)

Where the summations follow the same rule as in Eq. 4.1 and  $X_c$  is the X coordinate of the centre of the bubble.

The vertical velocity of the bubble was derived by dividing the difference between the maximum Y coordinate of each image of the bubble by 1/60, the interval of time between the two images.

The pixel size determination was made using both the maximum and minimum useable magnification of the zoom lens. A plastic transparent ruler with 0.0508 mm divisions was used to measure the pixel dimensions along both the vertical and the

horizontal axes.

Bubbles have been produced by both a stationary and a moving bubble generator. In the stationary system, two capillary tubes, 0.305 mm I.D. and 1.15 mm I.D. respectively were used for bubble generation. The capillary tubes were positioned at different angles from the horizontal to the vertical position in order to obtain different bubble sizes (Datta, *et al.*, 1950). Air was supplied to the capillary tube from an airtight reservoir maintained at constant pressure by a water column (Fig. 4.1), varying less than 1 % over a 3 hour test period. Air pressure was measured with a U-tube manometer accurate to 9.8 Pa of water. A flowmeter was connected between the air reservoir and the capillary tube. The complete air supply line together with the flowmeter were immersed in water to facilitate the detection of air leaks. The tip of the capillary was typically positioned at the bottom of the camera's field of view for focusing or below the field of view when bubble measurements were performed.

Smaller bubbles than the ones obtained using fixed capillary tube were produced using a rotating disk system shown on Fig. 4.4 using a 0.305 mm I.D. capillary tube. This system consisted of an 83 mm diameter plexiglass disk, 3.2 mm thick, mounted at the end of a 6.2 mm diameter shaft, 180 mm long. The shaft was driven by a 12 volt DC motor. Three holes, 2.8 mm diameter, were drilled on the disk at 15.7, 27.5 and 37.5 mm from the centre of the disk. The motor was mounted above the surface of the water in such a way that the disk maintained a horizontal position, 20 mm below the field of view of the camera. The capillary tube was inserted through the different holes and its end was maintained 10 mm above the disk upper surface. The tube was fixed at the bottom of the aquarium at a horizontal position coincident with the axis of the motor shaft. A DC power supply was used to control the rotational speed of the disk, which was measured with a hand-held stroboscope.

While the computer measured bubbles, the operator measured the air flow and the time required to produce 50 bubbles. Unfortunately, it was not possible to directly measure the time to produce 50 bubbles in the case of a moving tube. Thus, the rate of bubble production was evaluated by taking the ratio of the measured linear velocity of the end of the capillary tube to the mean horizontal distance between two consecutive bubbles.

A total of 2450 bubbles were measured by the vision system. These measurements were performed on bubbles ranging in diameter from 0.34 to 4.0 mm. The results





obtained from the camera and the flowmeter were used to calibrate the image analysis system.

# 4.3 Results and Discussion

The results obtained by the ruler measurements allowed the determination of the pixel size at both magnification levels. The pixel size results for the maximum magnification level were plotted as a function of their position on the screen (Fig. 4.5). Student's t-tests were performed and confirmed that the pixel sizes are independent of their position on the screen: for X axes ( $t_{1,24} = -0.410$ , P = 0.68) and Y axis ( $t_{1,25} = 0.337$ , P = 0.74). The average pixel sizes were 0.02571 mm along the X axis and 0.02616 along the Y axis.

#### **4.3.1** Bubble Measurement

The volume and equivalent diameter of the bubbles were calculated using the flowmeter data and were compared with the results from the image analysis system (Fig. 4.6). A linear regression was calculated and gave a slope of 0.9972 and an intercept of +0.0104. A Student's t-test was performed on the data and confirmed that the slope  $(t_{1,49} = 0.757, P = 0.459)$  and intercept  $(t_{1,49} = 1.53, P - 0.133)$  were not significantly different from 1.00 and 0.00 respectively. This implied that the two methods of bubble diameter measurement were not significantly different. Furthermore, the difference between the two methods of measurement were smaller than 0.05 mm or 2 pixels in size, and were not correlated to bubble size (Fig. 4.7).

#### 4.3.2 Vertical Velocity

The vertical velocities obtained from the image analysis system were compared to semi-empirical values presented by Clift *et al.* (1978). The comparison is presented in Fig. 4.8. The agreement was considered good given the usual amount of scatter of such data. The velocities measured with the image analysis system were generally slightly higher than those found in the literature. The high scatter observed for the large bubbles (D > 1.3 mm) is likely due to the oscillation of the bubbles.

Regressions were computed for the terminal velocity based on collected data. These are:



Figure 4.5 Pixel size with respect to vertical and horizontal location on the screen.



# Figure 4.6 Comparison between bubble diameters obtained from a flowmeter, and from the camera analysis.



Figure 4.7 Plot of differences between flowmeter and camera diameters.



# Figure 4.8

Comparison between semi-empirical correlations for terminal velocity presented by Clift *et al.* (1978) and the results obtained using the vision system.

$$\ln(V_t) = 1.21 + 8.48D - 5.52D^2 + 1.51D^3 - 0.15D^4$$
 (4.3)

for D < 1.3 mm

$$V_{r} = 348.71 - 14.73D \tag{4.4}$$

for 1.3 mm  $\leq$  D  $\leq$  4.0 mm

where,  $V_t$  is the terminal velocity and D is the equivalent bubble diameter.

# 4.4 Conclusion

A method based on digital image analysis was developed to automate the measurement of bubble parameters in a non-intrusive manner. The bubbles were identified and measured using thresholded digitized images.

Bubble diameters in the range of 0.3 to 4.0 mm as measured by the image analysis system agreed very well with those obtained by the flowmeter technique. The vertical velocities measured using the image analysis system compared well with published data. Regression equations for the terminal velocity of bubbles in the range of 0 to 4 mm are presented.

#### 4.5 <u>References</u>

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# CONNECTING TEXT

The image analysis system presented in the previous chapter permitted determination of the volume, the equivalent spherical diameter and the vertical velocity of a single bubble formed at the tip of a circular orifice immersed in water. However, when a cloud of bubbles is generated in a 3-D space, a discriminatory parameter must be used in order to define the depth of the field where measurements can be taken within known limits of accuracy.

The following chapter presents the technique followed to develop the required discriminatory parameter.

# V IMAGE ANALYSIS OF 3-D CLOUDS OF BUBBLES

### 5.1 Introduction

There is a wide range of operations and processes in industry and pollution control in which gas, in a dispersed phase, plays an important role. For many industrially used bubble generators, the bubble size can be variable within a certain range. The determination of bubble size and motion is helpful in determining the efficiency of the bubble generator with respect to its intended application.

The mechanism of bubble formation has been a subject of interest for several decades (Hadamard, 1911). The main research tool used to determine bubble velocity was the analysis of a series of pictures, measuring the difference between the bubble position on two successive pictures. Bubble size has usually been determined by measuring the liquid displacement generated by a given number of bubbles (Haberman and Morton, 1953).

Novel techniques have been recently experimented for the measurement of bubble parameters. Image analysis has been classed as one of the best (Tsukada and Horio, 1990). The development of real time digital image analysis has permitted the measurement of key bubble parameters in a two-dimensional bed (Lim *et al.*, 1990). To avoid the field depth effect, Lim *et al.* (1990) built their fluidized bed from a 9 mm deep aquarium and positioned its centre in the field of view of a camera. The drawbacks of this technique were the wall effects and the restricted dispersion of bubbles to only two dimensions.

Vigneault *et al.* (1990) developed an image analysis system to determine the volume, equivalent spherical diameter and vertical velocity of gas bubbles formed at the tip of a circular orifice immersed in water. The image taken by the camera was focused on the vertical axis of the orifice and gave accurate measurements of the volume and vertical speed of the bubbles. However, when a cloud of bubbles is generated in a 3-D space, a discriminatory parameter must be established in order to define a depth of field where measurements can be performed while preserving accuracy within known limits.

The first objective of this research was to develop a discriminatory parameter required for the accurate sampling and characterization of a cloud of bubbles in a 3-D

system and evaluate the application of this parameter to bubble sizes varying from 0.5 to 3.5 mm in diameter. The second objective was to evaluate the performance of the image analysis system, applied to a 3-D system, by measuring the size of bubbles generated as well as the flow rate produced by two different sintered stainless steel filters, so called diffusers, having pore sizes of 0.5 and 2  $\mu$ m diameter.

### 5.2 Materials and Method

The image analysis system has been described in detail by Vigneault *et al.* (1990). The images were captured by a camera which produced a video signal and transferred it to a digitizing board. The digitized images were then displayed in real time on a monitor. Up to four images could be stored in memory for further analysis.

## 5.2.1 Discriminatory Parameter

When using a fixed capillary tube to generate bubbles, the distance between the camera and the tip of the tube can be adjusted to obtain well focused images of bubbles. The plot of grey levels versus pixel position on a horizontal line across the centre of a bubble shows a slope with a sharp drop at the edge of the bubble (Fig. 5.1). The magnitude of the slope on the curve of grey levels versus pixel position at the bubble boundary decreases as the field of view of the camera is moved away from the capillary tube. The slope can be used as a parameter to discriminate between focused and out of focus bubbles provided that one can define a range of slope magnitudes over which the sizing accuracy is acceptable. To evaluate the relation between the magnitude of the slope and the accuracy of bubble size, measurements of both the slope at the bubble boundary and the bubble equivalent spherical diameter (D) were obtained for bubbles of known sizes. Measurements were realized with bubbles randomly placed within 3 cm on either side of the plane of best focus. Bubbles of known sizes were generated using the technique described in Vigneault et al. (1991) which also explains bubble sizing with the vision system. The slope magnitude was quantified by taking the difference in grey levels over a length of two pixels. The slope was evaluated around  $X_{min}$  and  $X_{max}$  over a horizontal length of 11 pixels (Fig. 5.2) resulting in 22 estimates of the slope from which the maximum magnitude was referred to as the jump. The  $X_{min}$  and  $X_{max}$  values were defined



Figure 5.1: Grey level along a horizontal line centred on a bubble.





as the minimum and the maximum values of the X coordinate of the contour pixels of a bubble. Plotting  $D_s$  as a function of the jump showed how the measurement accuracy degrades as the jump decreases. The measurements were performed using four bubble diameters: 3.35 mm, 2.29 mm, 1.40 mm and 0.74 mm. For each bubble size, 200 bubbles were analyzed.

### 5.2.2 Depth of Field

The depth of field (DOF) is the sum of the offset distance fore- and -aft from the object plane over which a sufficient sizing accuracy is retained. Knowing the minimum jump for acceptable accuracy, the DOF was measured by moving a bubble attached to a capillary tube fore and aft of the object plane until the minimum jump was reached. The bubble was moved using 1 mm increments. Bubbles ranging from 0.75 to 3.25 mm in diameter were used and 50 replicates were taken for each bubble size. Plots of the jump as a function of the offset from the object plane were used to define the DOF.

## 5.2.3 Gas Flow Rate and Bubble Size Distribution

Once the DOF is defined, the gas flow rate and the bubble size distribution of any bubble generator can, in principle, be measured. This was achieved by looking at a slice of the bubble cloud perpendicular to the gravity vector and of dimensions covering the entire cross section of the cloud. The slice was divided into cubes with a side length equal to the DOF (Fig. 5.3). The width and height of the cubes could be made larger or smaller if desired by adjusting the extent of the portion of the field of view of the camera used. The camera position was such that the object plane was in the middle of each cube. 20 images were analyzed at each measurement point. The analysis proceeded in three steps. First, the objects were identified by thresholding the image. Their contours were traced and the jump calculated for each object. The objects were then classified. Objects with a jump smaller than the minimum jump defining the DOF were classified as object out of focus (OOFO). Other objects were presented to the operator who had to decide how many bubble shadows created an object. When only one bubble (OOB) was identified, its volume and  $D_s$  were computed. If more than one bubble was included in an object, the number of grouped bubbles was stored for further use. Finally, a drop size distribution and a gas flow



Figure 5.3: Bubble cloud sampling.

rate were computed. The drop size distribution was built from OOB data and then corrected to account for the grouped bubbles and for the possibility that large objects were hiding smaller ones directly in front or behind them. The correction for grouped bubbles was obtained using Eq. 5.1:

$$N_i' = \frac{N_i}{\sum_{i=0}^n OOB} N_T$$
(5.1)

Where  $N_i' =$  number of bubble in the class i corrected to account for the grouped bubbles;

= number of bubbles in the class i determined from the OOB;

 $\sum_{i=0}^{n} OOB =$  sum of bubbles in all classes determined from the OOB;

 $N_T$  = sum of the bubbles in all classes including the OOB and the grouped bubbles;

n = number of classes.

N.

 $N_i$ 

V,

=

=

The correction to account for the possibility that large objects were hiding smaller ones was performed using Eq. 5.2:

$$N_{i}^{\prime\prime} = \frac{N_{i}^{\prime} V_{t}}{V_{t} - V_{Hi}}$$
(5.2)

Where

number of bubbles in class i corrected to account for the grouped bubbles and for the possibility of hidden bubbles; total volume of the sample, mm<sup>3</sup>;

 $V_{Hi}$  = volume of bubble of class *i* which could be hidden, mm<sup>3</sup>. The V<sub>Hi</sub> was calculated from Eq. 5.3.

$$V_{Hi} = \frac{\pi}{4} \sum_{j=i}^{n} (DOF - D_{sj}) (D_{sj} - D_{si})^2 N_j'$$
 (5.3)

Where

DOF = depth of the sample, mm;

 $D_{si}$  and  $D_{si}$  = mean diameter of classes *i* and *j* respectively, mm;

 $N'_j$  = number of bubbles in the class *j* corrected to account for grouped bubbles.

The results obtained from the cubic samples were assembled into a single number distribution. Then, for each class, the volume fraction was computed using Eqs. 5.4 and 5.5.

$$R_i = \frac{N_i'' V_i v_i}{20 Q H}$$
(5.4)

Where:

and

$$Q = \frac{1}{20 \ H} \sum_{i=1}^{n} N_{i}^{\prime\prime} V_{i} v_{i}$$
 (5.5)

R <sub>i</sub>	=	volume fraction in class <i>i</i> ;
$N_i$ "	=	corrected number of bubbles in class i;
V <sub>i</sub>	=	mean volume of the bubbles in class $i$ , mm <sup>3</sup> ;
v <sub>i</sub>	=	vertical speed of the bubble in class $i$ , mm•s <sup>-1</sup> ;
H	=	height of the cubic samples taken, mm;
Q	=	gas flow rate, mm <sup>3</sup> •s <sup>-1</sup> ;
n	=	number of classes

The vertical speed of the bubbles was determined using the empirical Eqs. 5.6 and 5.7 reported in the fourth Chapter (Vigneault *et al.*, 1992), for air bubbles in tap water.

$$\ln(v_{c}) = 1.21 + 8.48D - 5.52D^{2} + 1.51D^{3} - 0.15D^{4} \qquad \text{for } D < 1.3\,\text{mm} \quad (5.6)$$

 $v_{t} = 348.71 - 14.73D$  for  $1.3 mm \le D \le 4.0 mm$  (5.7)

Where  $v_t =$  equilibrium rising velocity with respect to ambient water,

 $mm \bullet s^{-1};$ 

D = equivalent spherical bubble diameter, mm.

In the case of high air flow rates, the water is entrained by the bubble cloud. The velocity of the bubble with respect to the camera frame of reference is therefore the sum of  $v_t$  and the liquid velocity. Measurement of water velocity was performed at each measurement point. It was achieved by measuring the velocity of 5 bubbles by the camera system using the same method as that described by Vigneault *et al.* (1991). The mean vertical liquid speed was then taken to be the average of the differences between the measured bubble speeds and  $v_t$ .

An independent measure of gas flow rate was used for comparison purposes. A volume displacement method was used where a water filled graduated cylinder was positioned to capture the whole bubble flux. The time required to capture 10 cc of air was measured with a stopwatch. Given a total reaction time of 0.5 s, the accuracy of this method was better than 1%.

## 5.3 <u>Results and Discussion</u>

### **5.3.1 Discriminatory Parameter**

Fig. 5.4 shows the variation of the measured  $D_s$  with the jump at nominal bubble diameters of 3.35 mm, 2.29 mm, 1.40 mm and 0.74 mm. The precision in the measurement of the bubble diameter was related to the jump. The greatest fluctuations in all cases was seen for jumps less than about 30. Furthermore, all the curves showed a significant slope change at a jump value of about 30, indicating a rapid increase of the error of measurement of the bubble diameter with decreasing jump. Table 5.1 shows the  $R^2$  and the equations of the curves fitted to the data in Fig. 5.4. The constant term on the right-hand side of equations in Table 5.1 is equal to the nominal bubble diameter. Thus, the other term presents the systematic error associated with a given jump. Fig. 5.5 is a plot of this error term. An error associated with a jump larger than 30 is considered acceptable since it is equivalent to the size of two pixels or an error of about 50  $\mu$ m on the equivalent spherical diameter. On Fig. 5.5, it can be seen that the magnitude of the error term is approximately constant over the range of bubble size considered for a jump larger than 30. Therefore, a single equation can be written for the error term that can be used to correct



Figure 5.4: Diameter variations for different bubble sizes as a function of the jump.

Bubble diameter (mm)	Equivalent spherical diameter $(D_s)$ vs jump $(x)$	R <sup>2</sup>	
3.35	$D_s = 3.349 - 0.831 e^{-0.097x}$	0.644	
2.29	$D_s = 2.286 - 0.588 e^{-0.091x}$	0.666	
1.40	$D_s = 1.395 - 1.080 e^{-0.120x}$	0.821	
0.74	$D_s = 0.743 - 1.750 e^{-0.123x}$	0.715	

Table 5.1: Regression equations and  $R^2s$  of the curves presented in Fig. 5.4.



Figure 5.5: Systematic error in bubble diameter as a function of the jump.

the measured bubble  $D_s$ . Eq. 5.8 was obtained by a least squares fit of the data of the jump in grey level (x) in Fig. 5.5 for  $x \ge 30$ .

$$\epsilon = 1.062 \exp(-0.108 x)$$
 for  $x \ge 30$ 

(5.8)

Where

= error on  $D_s$ , mm;

x = jump of grey level.

### 5.3.2 Depth of Field

ε

Fig. 5.6 shows the jump as a function of the offset from the object plane. At a jump of 30, the average depth of field was 6.67 mm for all the bubble diameters tested. A linear regression was performed on the depth of field measured at a jump of 30 for all bubble diameters presented at Fig. 5.7. The regression gave a slope of 0.076 and an intercept of 6.519. A Students' *t*-test was performed and confirmed that the slope was not significantly different from zero (t = 0.505, P = 0.632). This indicated that the depth of the field as defined here is independent of bubble diameter. Therefore, the depth of the field was considered to be equal to 6.67 mm. Fig. 5.6 shows that the degradation in the jump is more gradual for negative offsets (closer to camera) with an acceptable offset of approximately 3.7 mm compared to approximately 3 mm on the opposite side.

### **5.3.3** Gas Flow Rate and Bubble Size Distribution Measurements

To obtain the real dimensions of the bubbles analyzed by the imaging system, the diameters were readjusted using Eq. 5.9 for the error term.

$$D_{sc} = D_s + \epsilon \tag{5.9}$$

where:

Dsc

=

corrected diameter, (mm);

 $D_s = measured$  equivalent spherical diameter, (mm);

 $\epsilon$  = error on D<sub>s</sub>, mm;



Figure 5.6: Variation of the jump with the offset from the object plane for bubble diameters ranging from 0.75 to 3.25 mm.



Figure 5.7: Depth of field as a function of bubble diameters.

The distributions of the two distinct populations obtained with the two diffusers are presented in Figs. 5.8 and 5.9. No published data or alternate method of measurements were available to check the accuracy of these distributions. It is interesting to note that both distributions are smoother in the small diameter range suggesting that these sintered metal diffusers tend to form small bubbles more regularly than larger ones. Also despite a large difference in pore sizes (0.5 versus  $2.0\mu$ m) and small differences in flow rates (350.4 mm<sup>3</sup>•s<sup>-1</sup> for 0.5  $\mu$ m and 414.8 mm<sup>3</sup>•s<sup>-1</sup> for the 2  $\mu$ m), both distributions have a peak at bubble diameters between 1.1 and 1.2 mm. Moreover, the distribution for the 0.5  $\mu$ m diffuser is shifted towards larger diameters compared to the one obtained with the 2.0  $\mu$ m diffuser. This result suggests that coalescence of bubbles originating from adjacent pores dominates the process of bubble formation with these diffusers; when the pore size becomes smaller, it leads to a higher pore density.

Trials were conducted using higher air flow rates generating dense clouds of bubbles. The system did not successfully distinguish between the background and the bubble boundaries due to the great number of overlapping bubbles. The bubble populations generated were not characterized since only very few individual bubbles were analyzed for size distribution and were therefore not considered representative of the real bubble size distribution. A maximum bubble surface projection density of approximately 30 % was found to be the limit for a successful population characterization but this is highly dependent on bubble size.

Flow rates measured by the vision system and the water displacement method are compared in Table 5.2. For sintered metal diffuser having calibrated orifices of 0.5  $\mu$ m and 2  $\mu$ m, the average flow rates were respectively 350.4 mm<sup>3</sup>•s<sup>-1</sup> and 414.8 mm<sup>3</sup>•s<sup>-1</sup>. One should note that the vision system has a tendency to underestimate the flow rate by up to 24%. No explanation could be put forward to justify this discrepancy. However, the agreement between the two methods of measurement was good, given the uncertainty of bubble velocity as the diameter increases (Vigneault *et al.*, 1991).



Figure 5.8: Volume distribution for a 0.5  $\mu$ m diffuser at 350 mm<sup>3</sup>•s<sup>-1</sup>.

75



Figure 5.9: Volume distribution for a 2  $\mu$ m diffuser at 414 mm<sup>3</sup>•s<sup>-1</sup>.

Table 5.2:Flow rate comparisons.

Diffuser		Flow (1	<u>nm<sup>3</sup>•s<sup>-1</sup>)</u>	Difference	Mean of
pore		Camera	Flowmeter	<u>100 (Q<sub>F</sub> - Q<sub>c</sub>)</u>	differences
size		system	system	Q <sub>F</sub>	
(µm)	Rep	Q <sub>c</sub>	Q <sub>F</sub>	%	%
0.5	1	377.39	349.79	-7.89	
	2	300.96	350.80	14.21	
	3	346.47	350.81	1.24	
	4	290.32	352.19	17.57	
	5	323.36	348.43	7.21	6.46
2.0	1	348.24	413.03	15.69	
	2	311.22	408.26	23.77	
	3	364.90	423.29	13.80	17.75

## 5.4 <u>Conclusion</u>

A discriminatory parameter required for the characterization of clouds of bubbles has been developed. It was demonstrated that the discriminatory parameter, a minimum jump of 30, was independent of bubble size for the system under analysis. It permitted the determination of the depth of field and allowed the elimination of bubbles out of focus. With this discriminatory parameter, clouds of bubbles generated by two different diffusers were studied and the population distributions were determined. However, this system is limited to relatively low density bubble populations because it does not analyze the characteristics of overlapping bubbles in the images and it cannot "see" inside a dense cloud.

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## **CONNECTING TEXT**

A preliminary trial of bubble measurement demonstrated that the density of bubble clouds produced by the gas diffuser at the air flow rate required by the HPWS was too high for the image analysis system to be effective. A mechanical sampling technique was therefore developed to isolate representative samples from the high-density cloud which could then be analyzed using the above-mentioned system.

# VI DIFFUSER CHARACTERIZATION USING A MECHANICAL SAMPLER FOR HIGH DENSITY CLOUDS OF BUBBLES

## 6.1 Introduction

The experimental evaluation of a high pressure water scrubber (HPWS) presented by Vigneault and Raghavan (1991) requires the determination of the Sauter mean diameter  $(d_{3,2})$  of the cloud of bubbles generated at the bottom of a water column, since it has a potentially large influence on the solubilization process involved in the HPWS. This characteristic  $(d_{3,2})$  is not usually supplied with other specifications of commercially available bubble generators. It appears to be a function of the pressure differential through the bubble generator.

To be efficient, the HPWS has to function with a high air flow rate generating a fairly dense cloud of bubbles. Pal *et al.* (1988) conducted trials in order to characterize microbubble activity by optical and photographic surveys. Their experimental set up consisted of a water tunnel in which a porous section produced microbubbles. Photographs were taken of the clouds of bubbles generated under different sets of conditions. It has been difficult to obtain quantitative information on individual bubble characteristics whenever the measurements were taken in a very dense population of bubbles. Nevertheless general qualitative characteristics were derived. For example, an increase in bubble size created an increase in air flow rate. However the measurement system did not permit the determination of the rate of this increase. Photographs taken at low air flow rates permitted the capture of a lower population density from which quantitative analyses could be made.

A real time digitizing image analysis system, similar to the system developed by Tsukada and Horio (1990) was considered for the characterization of the bubble population generated in the HPWS. This system could be automated but had the drawback of restricting the dispersion of the bubbles to two dimensions (Lim *et al.*, 1990). When dealing with dense bubble populations, the existing imaging systems did not perform well and were therefore limited to fairly low density bubble populations (Vigneault *et al.*, 1992).

The first objective of this research was to develop and test a mechanical sampling technique which isolates a sample of the bubble population for image analysis. Such a sample should have a size distribution which is representative of the population. The second objective was to characterize the three air diffusers selected and to determine the most suitable diffuser for use in an experimental HPWS.

### 6.2 Materials and Methods

#### **6.2.1** Bubble generator

Many techniques for microbubble production have been investigated and reported. Among these, multistage bubble fractionation (Leonard and Blacyki, 1978), cavitation induced by asymmetric distorted pulses of ultrasound (Aymé and Carstensen, 1989), electrostatic atomization of a gas in a liquid medium (Sato, 1980) and utilization of a column packed with uniform microbead (Bowley and Hammond, 1978) were studied but none of them was retained. The application of these techniques to industrial uses was still limited and required equipment too sophisticated for our application. The most appropriate technique described in the literature was the injection of gas into the liquid phase through porous material casted in tube or plate shape. This system required only a compressed air supply and the initial investment was reasonable.

Porous materials can be used to obtain smaller bubbles than would be feasible using a nozzle tip. The porous materials are made by bonding or sintering together fine particles of carbon, ceramic, polymer, or metal. They can be used for gas dispersion or as foam generators. The diameter of the generated bubbles can be of the order of 100  $\mu$ m or more. In practice, the size of bubbles produced depends on both the size of the material's pores and the imposed pressure drop. The effectiveness of dispersion of the gas is a function of the pore size, the structure and the surface roughness of the material, the liquid medium as well as the depth, the turbulence, and coalescence of the gas bubbles (Fair *et al*, 1984).

Three samples of commercial porous material were selected to determine their suitability for the HPWS application. Selection criteria were: the availability of the product, the mechanical strength and the expected bubble size. The three samples were 125 mm long tubes made from sintered stainless steel, having nominal orifices of 0.5  $\mu$ m.

One tube was supplied by the manufacturer Warco<sup>1</sup> and was 12.7 mm nominal outside diameter. The two other tubes were supplied by the manufacturer Newmet and were 12.7 mm and 20 mm nominal outside diameter (O.D.).

# 6.2.2 Bubble Size Measurement Method

The image analysis system has been described in detail by Vigneault *et al.* (1991). It consisted of a CCD camera mounted on a two axis positioner allowing movement in a horizontal plane capturing images of rising bubbles generated at the bottom of a glassmade aquarium. The signal from the camera was digitized using a video board mounted in a microcomputer. For each camera position, the system took a prescribed number of images. Each image represented a cubic sample from the bubble population. The camera was positioned to capture, one by one, the cubes covering the entire surface through which the bubbles circulated without cubic samples overlap (Fig. 6.1). The cubic samples had an edge of 6.67 mm corresponding to the depth of the field of the camera (Vigneault *et al.*, 1992). The bubble size results obtained from all the cubic samples were grouped in eighty classes numbered from 0 to 79 with corresponding diameters varying from 0.275 mm to 4.225 mm with 0.050 mm increments.

### **6.2.3** Bubble Mechanical Sampler

A bubble mechanical sampler was designed to channel a small portion of the bubble population (Fig. 6.2). It had two main components: a frame and a pair of sliding gates. The two sliding gates were used to define a rectangular channel through which a sample of the bubble cloud flew. The gates were supported by a v-shaped frame which also directed the portion of the bubble cloud which was not sampled outside the field of view of the camera. By adjusting the position of the gates, the depth of the slice of sampled bubbles could be adjusted so it could be easily analyzed by the image analysis system. The length of the sampler's frame was such that it was longer than the dimension of the

<sup>&</sup>lt;sup>1</sup> Mention of specific trade names does not imply endorsement of those products by Agriculture Canada or McGill University. Similarly, omission of product names is not intended as discrimination against those not mentioned.







Figure 6.2: Mechanical bubble sampler shown in three principal views.

undisturbed bubble cloud along the main axis of the sampler. Its width was chosen to guarantee that deflected bubbles could not enter the field of view of the camera. The sampler was made of stainless steel. The wall of the gates exposed to the bubble flow were covered by a material that would prevent bubble attachment. As will be discussed later, various materials have been tried. Visual observations were made to determine on which material the bubbles did not stick and agglomerate. Thin sheets of aluminium, copper, stainless steel, plastic, glass and paper were tried. If the bubbles stuck to the material, they were likely to coalesce and disturb the real diameter distribution of the bubble population. The bubble generators were installed 5 mm below the lowest part of the bubble sampler and were located in the centre of the aquarium. The camera axis was perpendicular to the gates to capture images of the bubbles that had circulated between them.

# 6.2.4 Mechanical Bubble Sampling Effect on the Bubble Size Distribution

A 5 mm diameter sintered stainless steel diffuser, having nominal orifices of 2  $\mu$ m was used to evaluate the effect of the mechanical sampling of a cloud of rising bubbles. The size distribution of the bubble population was determined using the image analysis system. To determine the effect of the sampling technique, the bubble population distribution was measured for a constant air flow rate with 5 replicates with and without using the sampler. The samples were taken by positioning the camera to cover the entire horizontal surface through which the generated bubble flowed without overlapping the measured space. Ten images were analyzed at each camera position.

From the raw bubble diameter data, the Sauter mean diameter  $(d_{3,2})$  and the bubble diameter distribution were calculated. The  $d_{3,2}$  is a measure of the volume to surface ratio of the population of bubbles generated and is expressed by Eq. 6.1.

n

$$d_{3,2} = \frac{\sum_{i=1}^{n} d_i^3}{\sum_{i=1}^{n} d_i^2}$$
(6.1)

Where:

 $d_{3,2}$  = Sauter Mean Diameter, mm;

 $d_i$  = diameter of the bubble i, mm;

n = number of the bubble in the sample.

### 6.2.5 Diffuser Characterization

The minimum pressure differential through the diffuser required to generate bubbles offers a simple means to control the maximum pore size applicable to the diffusers (Anon., 1990). Each diffuser was immersed in a wetting liquid, e.g. alcohol, and pressure was increased so that the liquid could gradually be pressed out of the pores. The differential pressure was measured as soon as the first bubbles were observed. This differential pressure is a measure of the surface tension which is in equilibrium with this pressure, and is related to the pore diameter as follows.

$$d^* = \frac{4 \gamma}{\Delta P} \tag{6.2}$$

where:

 $d^*$  = apparent diameter of the largest pores;

 $\gamma$  = surface tension of the liquid;

 $\Delta P$  = differential pressure on the diffuser at which bubbling first occurs. The tests were repeated six times for each diffuser and the mean of these results was used to calculate the  $d^*$  of the diffuser.

The air flow rate was measured using a volumetric displacement measuring device. A manometer was connected to the gas inlet line to monitor the pressure at every gas flow rate measured to determine the effect of the pressure on the air flow rate.

The image analysis system was used to measure the diameter of the bubble generated by the three porous diffusers. The tests were carried from the lowest pressure required to generate bubbles, to the maximum flow rate still permitting measures when using the mechanical sampler. At low gas flow rates, the bubble sampler was not used since the bubble cloud density was low. The bubble sampler was essential to the analysis at higher gas flow rates which produced denser bubble populations. The camera captured images at four different positions over the length of the diffusers to obtain a bubble size distribution representative of the population produced. Ten images were analyzed at each camera position. The  $d_{3,2}$  was calculated using Eq. 6.1 for the results obtained over the 4 positions of the camera and a relationship between the  $d_{3,2}$  and the air flow rate per unit of length of the diffuser was established.

# 6.3 <u>Results and Discussion</u>

## 6.3.1 Mechanical sampler

The visual trials on the effect of different materials used for covering the gate walls showed that bubbles stuck to all materials except paper, permitting air bubble agglomeration and producing much larger bubbles. With paper no bubble appeared to stick on the gate of the sampler and the flow of the bubbles through the gates of the sampler was virtually undisturbed. The paper was cut to exceed the gates by approximately 1 mm. The paper rectangles were able to withstand the wet conditions for one day before visibly deteriorating.

## 6.3.2 Bubble Sampling-Technique Effect on the Bubble Size Distribution

A  $\chi^2$  goodness-of-fit test described by Daniel (1978) was conducted on the assumption that the frequency distribution of the population measured using the mechanical bubble sampler would be the same as that of the population measured without using the mechanical sampler. Fig. 6.3 shows the histograms of the volumetric distribution of the bubble clouds for the 2  $\mu$ m diffuser measured with and without the sampler. The  $\chi^2$  test conducted on the two distributions showed no significant differences in the two populations sampled ( $\chi^2_{21} = 16.055$ , P = 0.762).

Table 6.1 presents the  $d_{3,2}$  for the characterization of the 2  $\mu$ m diffuser at a nominal air flow of 350 mm<sup>3</sup> s<sup>-1</sup>. The averaged  $d_{3,2}$  of 1.047 mm and 1.059 mm were obtained for the population measured using and not using the mechanical sampler respectively. An F-test conducted on these  $d_{3,2}$  showed no significant difference between the two sets of measurements (F<sub>1,4</sub> = 0.23699, P = 0.6377).

Trials were conducted using higher flow rate but it was impossible for the image analysis system to determine the bubble population distribution without using mechanical bubble sampler. No other trustable method was found to evaluate the effect of the mechanical bubble sampler. Based on these results, it was considered that no bubble



Figure 6.3: Mean bubble size distribution measured with and without using the mechanical sampler for a 2  $\mu$ m diffuser.

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diffuser measured with and without using a bubble sampler.	

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Using the sampler	Replicate #	Air flow mm <sup>3</sup> s <sup>-1</sup> mm <sup>-1</sup>	<i>d</i> <sub>3,2</sub> (mm)	Average of $d_{3,2}$ (mm)
Yes	1	349.79	1.115	
Yes	2	350.80	1.045	
Yes	3	350.81	1.000	
Yes	4	352.19	1.035	
Yes	5	348.43	1.040	1.047
No	1	349.79	1.090	
No	2	350.80	1.062	
No	3	350.81	0.994	
No	. 4	352.19	1.138	
No	5	348.43	1.013	1.059

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agglomeration occurred on the paper surface and the sampler used was adequate for mechanical sampling of a cloud of bubbles without affecting its size distribution.

## 6.3.3 Diffuser Characterization

The minimum differential pressures measured were 11.56 kPa, 13.28 kPa and 19.10 kPa which corresponded to  $d^*$  of 7.78  $\mu$ m, 6.78  $\mu$ m and 4.71  $\mu$ m respectively for the Warco 12 mm, Newmet 12 mm and the Newmet 20 mm. The values of  $d^*$  obtained this way, however, are not equal to the actual pore size because the correlation is only valid for circular capillaries. By correlating the results of a glass bead test and the bubble point test, a correction factor can be determined. The shape of the pore influences this correction factor. It is approximately 0.2 for very irregular pores and 0.4 for pores which are formed by spherical particles. These figures are only to be considered as approximate and valid for a range of wall thickness ranging from 1 to 4 mm (Anon. 1991). Even if it was not possible to have the exact value of the correction factor applicable to the tested diffusers, it was clear that the largest pores of the Warco and the Newmet 12 mm O.D. were larger than the ones of the Newmet 20 mm O.D diffuser.

The effect of pressure on the gas flow rate can be seen in Fig. 6.4 for the three diffusers tested. Eqs. 6.3, 6.4 and 6.5 correspond to the curves presented in this figure for the Warco 12 mm, Newmet 12 mm and Newmet 20 mm respectively.

$$Q = -1.886 + 0.418 P$$
  $5 \le P \le 40$   $R^2 = 0.992$  (0.3)

$$Q = \left(\frac{P - 16.45}{5.09}\right)^{2.5} \qquad 18 \le P \le 42 \qquad R^2 = 0.995 \qquad (6.4)$$

$$Q = \left(\frac{P - 12.73}{14.69}\right)^{2.6} \qquad 10 \le P \le 82 \qquad R^2 = 0.997 \qquad (6.5)$$

Where:

Q = flow rate per unit of length of diffuser, mm<sup>3</sup> s<sup>-1</sup> mm<sup>-1</sup>;

P = pressure, kPa;

R =correlation coefficient.



Figure 6.4: Gas flow rate per unit of length expressed as a function of pressure for the three 0.5  $\mu$ m diffusers tested.

Fig. 6.5, shows the  $d_{3,2}$  as a function of the gas flow rates for the three diffusers tested. The upper limit was imposed by the excessive size of the bubbles produced. In fact, bubbles having a diameter larger than 4.0 mm were not measurable using the imaging system. The gate of the sampler had to be opened too wide to permit those bubbles to circulate and the cloud of bubbles seen by the camera was then too dense. The measurements were stopped when bubbles of this size were produced.

At low air flow rates, the results showed a decrease in the  $d_{3,2}$  with an increase in the air flow rate for the 12 mm O.D. diffusers. After reaching a minimum, the  $d_{3,2}$ increased with an increase in the air flow rate. This decrease at the low air flow rate might be explained by a non-uniform pore size which correlates with the larger  $d^*$  calculated from the results using the two 12 mm O.D. diffusers. Pressure and flow rate being directly related, at low pressures, only the larger pores produce bubbles. As the pressure increases, the smaller pores begin to produce smaller bubbles which decrease the  $d_{3,2}$ . With further increase in pressure and flow rate, the bubbles produced increase in diameter. Datta *et al.* (1950) suggested that a part of the kinetic energy of the gas stream emerging into an expanding bubble may be converted into pressure energy which would help to inflate the bubble but they could not demonstrate it. This increase in diameter with the increase of the air flow rate was also noticed by Pal *et al.* (1988). The decrease in the  $d_{3,2}$  at low pressure and flow rate was not present for the Newmet 20 mm O.D. which would be due to more uniform pore size.

Eqs. 6.6, 6.7 and 6.8 correspond to the curves presented in Fig. 6.5 for the Warco 12 mm, Newmet 12 mm and Newmet 20 mm respectively.

$$d_{3,2} = 0.875 \ Q^{0.6} - 0.1 \qquad 3 \le Q \le 11 \qquad R^2 = 0.944$$
 (6.6)

$$d_{3,2} = 0.751 + (0.143Q - 0.245)^{0.6}$$
  $2 \le Q \le 14$   $R^2 = 0.994$  (6.7)

$$d_{3,2} = 0.4 \ Q^{0.37} + 0.4 \qquad 0 \le Q \le 58 \qquad R^2 = 0.989$$
 (6.8)

Where

 $d_{3,2}$  = Sauter mean diameter, mm;





: Sauter mean diameter expressed as a function of gas flow rate per unit of length for the three 0.5  $\mu$ m diffusers tested.
Q = flow rate per unit of length of diffuser, mm<sup>3</sup> s<sup>-1</sup> mm<sup>-1</sup>.

For the development of a HPWS, the Newmet 20 mm O.D. was considered as the most appropriate for the two following reasons. First, the largest pores of this diffuser were fairly small in  $d^*$  and its  $d_{3,2}$  did not decrease as the air flow increased producing a smaller  $d_{3,2}$  than the two other diffusers when using low air flow rate. Secondly, the rate of change of the  $d_{3,2}$  with the flow rate was smaller for this diffuser compared to the two others. This helped to produce a relatively low  $d_{3,2}$  at high air flow per unit of length reducing the length of the diffuser required to produce a given total air flow while keeping the  $d_{3,2}$  low.

# 6.4 <u>Conclusion</u>

A mechanical means of sampling a small bubble population from a dense cloud of bubbles has been developed. No bubble agglomeration was visually detected on the surface of the sampler. Bubble size distributions and Sauter mean diameter  $(d_{3,2})$  measured using the bubble sampler were not significantly different from the ones obtained without the help of the sampler.

The sampler could therefore be used to characterize three diffusers. It permitted to determine the most suitable diffuser for producing bubbles in the HPWS presented in Chapter 3 (Vigneault and Raghavan, 1991). The  $d_{3,2}$ , the air flow per unit of length of the diffuser and the air flow ratio to the differential pressure through the wall of the diffuser were all established.

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# CONNECTING TEXT

The efficiency of a HPWS in transporting a gas is mainly due to two parameters: the quantity of gas it can contain at saturation (gas solubility) and the degree of saturation the liquid reached before exhausting the system. The degree of saturation is a function of various parameters of the system used, such as the height of the water column, the size of the bubble and the water flow rate. The solubility is related to the gas, pressure, temperature and liquid properties.

The solubility of the three gases involved in the HPWS can be found in the literature for pure distilled water. Since the water used in the system was continuously recirculated, its purity differed from that of distilled water. The gas solubility in this contaminated water varied significantly and had to be measured. The following chapter describes the solubilimeter developed and used in this research.

# VII A METHOD FOR MEASURING GAS SOLUBILITY

# 7.1 Introduction

The development of a high pressure water scrubber (HPWS) used to modify and control the gas composition in a fruit or vegetable storage room (Vigneault and Raghavan, 1991) required the measurement of the solubility of  $CO_2$ ,  $O_2$  and  $N_2$  in water. The objectives of this work were: 1) to develop a simple and inexpensive method of measurement of pure gas solubility in various water qualities; and, 2) to estimate the precision of this method by comparing the results obtained for the pure gas solubility in distilled water to published data.

# 7.2 <u>Review of Literature</u>

Accurate knowledge of equilibrium air-liquid partitioning coefficients, or gas solubility, is essential to a variety of applications such as the design of air stripping towers or water aeration processes involved in environment control processes. Current methods for measuring gas solubility usually involve batch air stripping in a bubble column as proposed by Mackay *et al.* (1979). This technique is popular because it only requires the measurement of relative concentrations so that the preparation of standard curves for determining exact concentrations is unnecessary. The technique is, however, limited; equilibrium is assumed to be reached between the gas concentration in the liquid and in the bubbles, and the time available for reaching equilibrium is set by the height of the column.

The EPICS technique (Equilibrium Partitioning in Closed Systems) was developed by Lincoff and Gossett (1984) for gas mixtures. This technique requires only the measurement of relative concentrations of the gas in one phase (gas or liquid). Since this technique uses closed systems which may be equilibrated indefinitely, it is free of mass transfer limitations.

The above two techniques require gas chromatography or other gas analysis methods to determine the relative gas concentrations in either or both the gas and the liquid phases. The standard method described in ASTM (1990) has the advantage of avoiding the utilisation of gas analysis method but involves hazardous operations, equipment and materials such as mercury. This method is also fairly tedious and requires expensive equipments.

## 7.2.1 Gas Solubility

In 1803, William Henry observed that the amount of gas that dissolves in liquid at a given temperature is directly proportional to its partial pressure (Gerrard, 1980). Henry's Law is expressed by Eq. 7.1.

$$C_l = \frac{P_{eq}}{H} \tag{7.1}$$

Where  $C_i =$  liquid phase solute concentration at equilibrium, mole of gas•L<sup>-1</sup> of liquid;

 $P_{eq}$  = partial pressure of the gas at equilibrium, kPa.

There are theoretical reasons for employing mole fraction, but there are no universally accepted units for expressing the solubility of a gas and this unit is not convenient for general use. Since the solubility of a gas is usually determined by measuring the dissolved volume, rather than the dissolved mass, units of volume of gas per volume of liquid are more frequently used (Glasstone, 1946). Eq. 7.2 can be used to transform the  $C_l$  to solubility expressed in terms of volume of gas per unit of volume of liquid.

$$S = \frac{C_l}{\rho} = \frac{P_{eq}}{\rho H}$$
(7.2)

Where S = gas solubility,  $L_s \bullet L^{-1}$  of liquid;  $\rho$  = specific gravity of the gas, mole  $\bullet L_s^{-1}$ .

The unit  $L_s$  is the standardized litre of gas, that is, the volume occupied by the a mass of gas at 0°C (273.15°K) and 101.3 kPa absolute pressure. The conversion from L to  $L_s$  is performed using the perfect gas law.

The solubility of gas in liquid is inversely proportional to the temperature. Published data of gas solubility in distilled water (Weast and Astle, 1979) for the three gases involved in the separation process of the HPWS are presented as a function of temperature in Fig. 7.1. The van't Hoff equation describes the effect of the temperature on the solubility and shows that the natural logarithm of S varies linearly with the reciprocal of the absolute temperature (Glasstone, 1954). Eqs. 7.3, 7.4 and 7.5 can be used to compute solubilities at various temperatures between 0°C and 30°C. These were obtained by applying linear regression to the data in Fig. 7.1.

$$S_{o_2} = \exp\left[-9.3633 + \frac{1728.7}{T}\right]$$
 (7.3)

$$S_{CO_2} = \exp\left(-9.0254 + \frac{2608.8}{T}\right)$$
 (7.4)

$$S_{N_2} = \exp\left(-9.4479 + \frac{1550.7}{T}\right)$$
 (7.5)

Where  $S_{O2}$ ,  $S_{CO2}$ ,  $S_{N2}$  = solubilities of  $O_2$ ,  $CO_2$  and  $N_2$  respectively,  $L_s \cdot L^{-1}$  of liquid; T = temperature of the liquid, °K.

## 7.2.2 Heat of Solution

When a gas dissolves in a liquid, heat may be released or absorbed in the process (Glasstone, 1946). The magnitude of this heat exchange per unit mass of gas dissolved varies with the gas concentration in the liquid; it is larger at low gas concentrations. For distilled water at zero gas concentration, 8.94 kJ per gram of gas dissolved is released for  $CO_2$  while solubilization of  $O_2$  and  $N_2$  occurs without any heat exchange (Weast and Astle, 1979). For  $CO_2$ , the significant amount of heat released during solubilization may affect the experimental conditions by varying the system temperature.

## 7.3 Material and Methods

#### 7.3.1 Experimental Apparatus

Theoretically, the solubility of a pure gas may be indirectly obtained by measuring the pressure drop caused by gas solubilization in a pressurized closed container. A gas solubility measuring method is proposed based on this principle. Initially, the container is filled with a known quantity of degasified liquid and pure gas. When temperature is kept



Figure 7.1 Gas solubilities of  $O_2$ ,  $CO_2$  and  $N_2$  in water under standard pressure (101.3 kPa) as a function of the temperatures (Weast and Astle, 1979).

constant, the system evolves to an equilibrium state where the system pressure is recorded. The difference between the initial and equilibrium pressures is used to calculate the mass of gas dissolved using the perfect gas law.

The apparatus developed consisted of an airtight cylindrical container made from a PVC tube, 700 mm long, 100 mm inside diameter and having a net volumetric capacity of 5600 mL (Fig. 7.2). A thermocouple of  $0.5^{\circ}$ C precision was installed to sense the temperature of the gas approximately 25 mm below the top of the cylinder. A 0 to  $1000 \pm 3.5$  kPa pressure gauge and a self shut off coupling valve were installed at the same end of the cylinder to allow gas flushing and pressure measurement. Two valves (A and B in Fig. 7.3) isolated the pressure gauge and the coupling valve when necessary. At the opposite end of the cylinder, two valves (C and D) were installed to fill and empty the cylinder with water. An S-shaped water trap was installed at the bottom of the valve D to prevent air infiltration while emptying the cylinder. A baffle plate was installed in the centre of the cylinder. The baffle plate was maintained in a 45° position by two supports. This baffle slowed down the air and water flow and forced them to mix together when reversing the cylinder top and bottom. The 180° rotation of the cylinder was possible by mounting the apparatus on a horizontal shaft positioned at the centre of gravity of the apparatus.

## 7.3.2 Structural Resistance Test Procedure

A pressure test was conducted to determine the volumetric deformation of the apparatus due to the internal pressure. This test consisted of lowering the apparatus into a water container 150 mm in diameter and 1250 mm high. The apparatus was firmly held in place leaving only the top part of the apparatus exposed to air, i.e. the pressure gauge and the two valves A and B. The water level was precisely marked using a needle tip touching the water surface. The apparatus was pressurized at five different levels varying from 240 kPa to 800 kPa. Some water was removed to readjust the water level to the needle tip level. The volume of water removed was measured using a 10  $\pm$ 0.1 ml syringe. This volume of water corresponded to the increase in the volume of the apparatus when submitted to pressure. The series of test were replicated three times. At the end of each series of tests the pressure was released and the decrease in the volume of the apparatus









B)



was measured to detect any permanent volumetric deformation.

#### 7.3.3 Solubility Test Procedure

The procedure for measuring gas solubility in distilled water consisted of two stages. First, a water reservoir was positioned at a higher level than the apparatus (Fig. 7.3). This reservoir was connected to the valve C to fill the cylinder by gravity exhausting the air through the valve B. Water was added until the control vessel connected to the valve B and positioned at a higher level than the apparatus was half filled (Fig. 7.3A). After filling the pressurized cylinder, the water was degasified using a vacuum pump operated for 4 minutes at an absolute pressure of 11 kPa. Then as shown in Fig 7.3B, the pressurized pure gas cylinder (7 MPa) was connected to the valve B and the apparatus was partially emptied to make room for the introduction of the pure gas. The water removed was weighed using a 0 to 60 kg scale  $\pm 1.0$  g precision. A pressure regulator installed on the pure gas cylinder was used to maintain the gas in the apparatus at a gauge pressure inferior to 10 kPa. A heat exchanger consisting of a 2.5 m long copper tube of 6.4 mm O.D. was installed between the pressure regulator and the valve B to allow the gas to reach ambient temperature before entering the apparatus.

The volume of water replaced by pure gas was approximately 2500, 600 and 600 mL while testing respectively the  $CO_2$ ,  $O_2$  and  $N_2$ .

After the water was replaced by gas, the pressure regulator on the pressurized pure gas cylinder was rapidly adjusted at approximately 775 kPa to force more gas to enter into the cylinder. The valve B was then closed and the time, the temperature and the pressure inside the cylinder were measured to establish the initial condition of the solubilization process. The rapidity of gas filling was very important in minimizing gas solubilization and gas temperature variations before reading the initial pressure and temperature.

The second stage of the test consisted in forcing the solubilization process by rotating the apparatus about once a minute, forcing the gas and water to mix together. The pressure was monitored every two minutes. When the pressure had stabilized for 6 minutes, the gas and water were considered at equilibrium and the test was terminated. The cylinder was emptied and the water it contained was weighed. The temperature of the water was measured using a thermometer of  $\pm 0.5^{\circ}$ C precision before filling the apparatus

and immediately after the test. Since there was very little variation in water temperature through the duration of the test, the mean of these two temperatures was used as the temperature of the test.

The relative solubility of the gas was defined as the ratio between the measured solubility and the published solubility of the gas for the temperature of the test. It was calculated using the Eqs. 7.6 and 7.7.

$$S_r = \frac{V_{gs} P_s}{P_{ca} V_l S}$$
(7.6)

$$V_{gs} = V_i - V_e \tag{7.7}$$

Where	S <sub>r</sub>	=	relative solubility of the gas at the temperature of the liquid,
			dimensionless;

S = published solubility of the gas at the temperature of the test,  $L_{s} \bullet L^{-1}$  of liquid (Eqs. 7.3, 7.4 and 7.5);

 $P_{eq}$  = pressure at equilibrium, kPa;

 $P_s = standard pressure, kPa;$ 

 $V_{I}$ 

volume of liquid contained in the apparatus during the test,
 L;

$$V_{gs}$$
 = volume of gas solubilised in the liquid,  $L_s$ ;

 $V_i = initial volume of gas, L_s;$ 

 $V_e$  = volume of the gas remaining in the apparatus when equilibrium had been reached,  $L_s$ .

The solubility tests were conducted with distilled and contaminated water. The contaminated water was taken from the HPWS after having passed through the system several times and is referred to as "system water". The samples were taken at different times. The quality of the system water was quite variable since some tap water had to be added to compensate for water losses during the test of the HPWS. The tests using the system water were conducted at ambient temperature which varied from 22°C to 24°C. The tests using distilled water were conducted at ambient temperature tests were conducted by the system water water water water water water were conducted by the system water wat

moving the whole system to a controlled temperature room maintained at about 4°C. Each test was replicated four times.

#### 7.3.4 Temperature Test Procedure

Since compressed gases are used in the gas solubility measuring method, the impact of system cooling due to gas expansion must be considered. When decompressing a gas, a cooling effect is produced by heat absorbtion ( $\Delta$ H) due to a gas expanding to equilibrate with its new conditions. This  $\Delta$ H may produce a significant decrease in temperature. Using the mean conditions for the test, which are initial pressure = 6.65 MPa, release pressure = 0.86 MPa and constant temperature = 295 °K; the theoretical  $\Delta$ H of CO<sub>2</sub>, O<sub>2</sub> and N<sub>2</sub> were evaluated to be 220 J•g<sup>-1</sup>, 15 J•g<sup>-1</sup>, 16 J•g<sup>-1</sup> respectively (ASHRAE, 1981).

A temperature test was conducted to evaluate the efficiency of the heat exchanger (Fig. 7.3B) in supplying the heat to compensate for the cooling due to gas expansion and to evaluate the importance of the temperature increase due to gas solubilization at the beginning of the test. The test consisted in filling the apparatus in the same way as the solubility test using approximately 600 mL, 2600 mL and 5600 mL (5600 mL is the total volume of the apparatus) of CO<sub>2</sub> at 775 kPa and monitoring the temperature of the gas inside the apparatus at 5 s intervals. Tests using O<sub>2</sub> or N<sub>2</sub> were not conducted since trial showed no detectable temperature variation. Furthermore, theoretically the temperature decrease should theoretically be approximately 14 times lower in the case of these two gases compared to CO<sub>2</sub> and there is not any temperature increase due to O<sub>2</sub> or N<sub>2</sub> gas solubilization.

#### 7.3.5 Error Analysis

This error analysis is presented to assess the relative importance of the accuracy in the measurements of pressure, volume and temperature on the resulting solubility. Further, the results of the error analysis will be used to specify a ratio of the volume of liquid to the volume of the apparatus where the error is minimized.

From the basic measurements, the solubility is computed as follows:

Where  $V_a =$  volume of the apparatus (corrected for expansion);

$$S = \frac{V_a - V_l}{V_l} \frac{T_s}{T} \frac{(P_o - P_{eq})}{P_s}$$
(7.8)

 $V_1$  = volume of liquid in the apparatus, L;

= standard temperature, 273, °K;

= test temperature,  $^{\circ}K$ ;

T,

Т

 $P_o = initial pressure, kPa;$ 

 $P_{eq}$  = equilibrium pressure at the end of the test, kPa;

$$P_s = standard pressure, 101.3 kPa.$$

At the first order, the maximum absolute error  $|\Delta S|$  on S is obtained using:

$$|\Delta S| = |\Delta S_{\nu}| + |\Delta S_{p}| + |\Delta S_{T}|$$
(7.9)

$$|\Delta S_{v}| = \left|\frac{\partial S}{\partial V_{a}}\right|_{S_{0}} |\Delta V_{a}| + \left|\frac{\partial S}{\partial V_{l}}\right|_{S_{0}} |\Delta V_{l}|$$
(7.9-a)

$$|\Delta S_{p}| = \left|\frac{\partial S}{\partial P_{o}}\right|_{S_{0}} |\Delta P_{o}| + \left|\frac{\partial S}{\partial P_{eq}}\right|_{S_{0}} |\Delta P_{eq}|^{-1}$$
(7.9-b)

$$|\Delta S_T| = \left| \frac{\partial S}{\partial T} \right|_{S_0} |\Delta T|$$
 (7.9-c)

Where

Expanding (7.9-a) gives:

$$\frac{|\Delta S_V|}{S} = \frac{1}{1-\alpha} \left[ \frac{|\Delta V_a|}{V_a} + \frac{|\Delta V_l|}{V_a} \frac{1}{\alpha} \right]$$
(7.10)

Where  $\alpha = \frac{V_l}{V_a}$ .

Noting that  $V_a$  and  $V_l$  are measured by the same method, it follows that

 $|\Delta V_a| = |\Delta V_l|$ , and

$$\frac{|\Delta S_V|}{S} = \frac{1}{1-\alpha} \left[1 + \frac{1}{\alpha}\right] \frac{|\Delta V|}{V_a} = \frac{|\Delta V|}{V_a} f_v(\alpha)$$
(7.11)

Taking the derivative in (7.9-b) yields:  $\frac{|\Delta S_p|}{S} = \frac{2 |\Delta P|}{(P_o - P_{eq})}$ 

$$= 2 \frac{T_r}{T} \frac{|\Delta P|}{P_s} \frac{1}{S} \left[ \frac{1-\alpha}{\alpha} \right]$$
  
$$= K \frac{|\Delta P|}{P_s} f_p(\alpha)$$
(7.12)

Where  $K = 2 \frac{T_r}{TS}$  and  $|\Delta P_o| = |\Delta P_{eq}|$  has been assumed by the same argument as for the equality of volume errors. Finally, the error on S due to temperature is:

$$\frac{|\Delta S_i|}{S} = \frac{|\Delta T|}{T}$$
(7.13)

Combining Eqs. 7.11, 7.12 and 7.13 gives:

$$\frac{|\Delta S|}{S} = \frac{|\Delta V|}{V_a} f_{\nu}(\alpha) + K \frac{|\Delta P|}{P_s} f_p(\alpha) + \frac{|\Delta T|}{T}$$
(7.14)

In Eq. 7.14,  $\Delta V$ ,  $V_a$ ,  $\Delta P$  and  $P_s$  are fixed by design and convention; K and T vary from test to test while  $\alpha$  can be chosen to suit any particular test. For the test conditions used here, the fixed terms in Eq. 7.14 are:  $\frac{|\Delta V|}{V_a} = 0.00071$  and  $\frac{|\Delta P|}{P_s} = 0.035$ . For T = 20 °C:  $K = \frac{1.86}{S}$  and  $\frac{|\Delta T|}{T} = 0.0017$ . Inserting these values in Eq. 7.14 and taking S = 6.9 L<sub>s</sub> • L<sup>-1</sup>, typical of CO<sub>2</sub> solubility at 770 kPa yields:

$$\frac{|\Delta S|}{S} = 0.00071 f_{\nu}(\alpha) + 0.0094 f_{p}(\alpha) + 0.0017$$
(7.15)

Eq. 7.15 is plotted in Fig. 7.4 which shows the relative contribution of the various terms to the total error. The contribution of the error on temperature is small and constant.



Figure 7.4 Relative contribution of the various terms to the total error.

At low  $\alpha$ , the error on the pressure dominates. It decreases with increasing  $\alpha$ , and is smaller than the error on volume at high values of  $\alpha$ . The total error has a minimum at  $\alpha = 0.73$  which means that the test should be performed using 1680 mL of CO<sub>2</sub> to maximize the precision of the results.

Taking the derivative of Eq. 7.14, the minimum in the curve is found. It is given by:

$$\alpha_{\min} = -\frac{a_1 S + a_2}{a_1 S - a_2} + \frac{\sqrt{2}}{a_1 S - a_2} \sqrt{a_1^2 S^2 + a_1 a_2 S}$$
(7.16)

Where

$$a_1 = \frac{|\Delta V|}{V_a}$$
 and  $a_2 = 2\frac{T_r}{T} \frac{|\Delta P|}{P_c}$ 

Eq. 7.16 is plotted in Fig. 7.5 for the same conditions as Fig. 7.4. Also shown in Fig. 7.5 is the error associated with  $\alpha_{min}$  for solubilities varying from 0 to 10. Fig. 7.5 can be used to optimize  $\alpha$ . It shows that increasing the pressure at which the test is performed, thus increasing the measured solubility, reduces the error.

# 7.4 Results and Discussion

#### 7.4.1 Structural resistance

The volumetric deformation caused by internal pressure is presented in Fig. 7.6. The volumetric deformation followed a straight line when plotted against the applied pressure. This means that the deformation occurred in the elastic range of the material used. Furthermore, the water added to adjust the water level when the pressure was released at the end of each series of test was equal to the sum of the volume of water removed at each increase of the pressure. This means that the apparatus returned to its initial volume when the pressure was removed.

Eq. 7.17 represents the volumetric deformation of the apparatus as a function of the pressure applied.

$$\Delta V = 0.0368 P \qquad R^2 = 0.996 \qquad (7.17)$$

Where P = pressure applied, kPa;



Figure 7.5 Minimum and maximum error associated with  $\alpha_{min}$  for solubilities



Figure 7.6 Increase in the volume of the apparatus with increasing pressure.

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 $\Delta V =$  volume increase, cm<sup>3</sup>; R = correlation coefficient.

Since the water is an incompressible fluid and was introduced while no pressure was applied, the volume increase caused by the pressure was added to the volume of gas contained in the apparatus in the calculation of the solubility of the gas.

## 7.4.2 Temperature History

The variation of the temperature with time is shown in Fig. 7.7 for each of the three volumes of gas tested. As it can be seen, the temperature decreases very quickly when the gas is introduced to pressurize the apparatus. As expected, the cooling effect increased as the volume of gas increased. The temperature decrease was fairly small  $(0.3^{\circ}C)$  when using 600 mL of gas and would result in an error of 0.1% in the calculation of the initial volume of gas if neglected. The temperature decrease of 1.4°C when using 2600 mL of gas was much more important but still represents an error of 0.5% in the calculation of the initial volume of gas if neglected.

The gas temperature increase due to gas solubilization was  $1.7^{\circ}C$  and  $2.9^{\circ}C$  when using 600 mL and 2600 mL of gas respectively. To avoid any error in the calculation of the initial volume of gas, it is clear that the initial condition should be measured within about 5 seconds after pressurization of the apparatus. When using 600 mL of gas, if one uses the water temperature as the temperature of the test, neglecting to monitor the temperature of the gas, the error on the calculation of the solubility of the gas would be fairly low (0.1%); the important point being to read the pressure very quickly (within 5 s) after the apparatus is pressurized.

Rapid temperature increase was not observed in the case of 5600 mL of gas. This is explained by the absence of water, and therefore, of solubilization in the apparatus. The slow temperature increase was due to heat exchange through the apparatus wall.

#### 7.4.3 Solubility

The pressure variation in time is shown in Fig. 7.8. The test duration did not exceed 30 minutes in all cases. The precision in reading the initial pressure had a large impact on the solubility measurement accuracy. Experience demonstrated that the difficulty



Figure 7.7 General variation of temperature inside the apparatus over time.



Figure 7.8 General variation of pressure inside the apparatus over time.

of getting good readings was due to the rapid drop in pressure immediately after closing the valve B following gas pressurization as can be seen in Fig. 7.8. After a few tests, operators had no problem obtaining good readings within the 5 s that followed the closure of valve B and precision was thereby improved.

The relative solubility presented in Table 7.1 varied between 0.977 and 1.030. The mean and the standard deviation were  $1.004 \pm 0.004$  for CO<sub>2</sub>,  $1.006 \pm .012$  for O<sub>2</sub> and 0.997  $\pm 0.016$  for N<sub>2</sub>. The lower value obtained for N<sub>2</sub> solubility was expected since the tests were begun by degasifying the water at an absolute pressure of 11 kPa. At this pressure, some gases are still soluble. At 23 °C and the mean pressure at which the tests were conducted, the vapour pressure is 2.8 kPa (ASHRAE, 1981). The gas solubilized in distilled water should equilibrate with the remaining pressure, 8.2 kPa, which should result in a systematic underestimation of the CO<sub>2</sub>, O<sub>2</sub> and N<sub>2</sub> compared to that of CO<sub>2</sub>. This is explained by the error analysis. The pressure drop for the test done on O<sub>2</sub> and N<sub>2</sub> was much smaller even though the volume of gas was about four times lower. The precision of the reading was about 2.3% of the pressure drop for the O<sub>2</sub> and N<sub>2</sub> while it was about 0.8% in the case of the CO<sub>2</sub>. However the precision obtained was sufficient to evaluate the solubilities for the HPWS assessment.

Table 7.2 shows the relative solubility for the system water at ambient temperature. As can be seen, the relative solubilities are smaller than 1. This reduction in solubilities was expected since the solubility of gases decreases with a decrease in water purity. The larger variability of these results is likely due to the variability of the quality of the system water. In fact, it was suggested that the quality of the system water continuously changed with the time. It was, indeed, the reason why monitoring of the gas solubility gas was recommended for the development of the HPWS.

# 7.5 <u>Conclusion</u>

A simple method to measure the solubility of pure gases in water was developed. The importance of rapidly reading the pressure at the initial conditions was demonstrated. The temperature monitoring during the test may be neglected since the variations were not important and did not significantly affect the precision in calculating the initial volume of

Table 7.1	Comparison between the measured values of the solubility of distilled water
	at both ambient and cold temperatures with the solubility published in the
	literature.

Temperature		Relative Gas solubilities (measured/published)		
	Run #	CO <sub>2</sub>	<b>O</b> <sub>2</sub>	N <sub>2</sub>
Ambient	1	1.006	1.005	1.002
	2	0.996	1.030	1.017
	3	1.004	0.998	0.979
	4	1.006	1.007	0.999
Cold	1	1.006	0.999	1.002
	2	1.010	0.987	0.977
	3	1.003	1.011	1.012
	4	1.004	1.017	0.973
Average $\pm \sigma$		$1.004 \pm 0.004$	$1.006 \pm 0.012$	0.997±0.016

Table 7.2	Comparison between the measured values of the solubility of used water at						
	ambient temperature and the values published in the literature for the						
	solubility of distilled water.						

D	Relative Gases solubilities (measured/published)				
Kun #	CO <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>		
1	0.881	0.829	0.929		
2	0.930	0.840	0.921		
3	0.870	0.893	0.842		
4	0.878	0.888	0.921		
Average $\pm \sigma$	$0.890 \pm 0.024$	$0.863 \pm 0.028$	$0.903 \pm 0.036$		

gas. The results obtained using this method of measuring the solubility of  $CO_2$ ,  $O_2$  and  $N_2$  in distilled water were compared to published data. Measured and published data agreed within 3% in all cases and the mean deviation for each gas did not exceed 0.6%. An error analysis enable the optimization of the volume of gas to increase the precision in the measurement of gas solubility. A more precise pressure gauge may also be used when greater precision in the gas solubility measurement is required. This simple method permitted the measure of gas solubility in water of various qualities encountered in the development of a high pressure water scrubber for controlled atmosphere storage of fruits and vegetables.

# 7.6 <u>References</u>

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# **CONNECTING TEXT**

During the two first steps of the present work, a methodology for characterizing populations of bubbles produced by bubble generators as a function of the gas flow rate was developed. A gas solubilimeter permitting measurement of pure gas solubility in pure and contaminated water was also developed.

To complete this work, the last step was to design, build and test an HPWS. The components and the instrumentation used in this research were chosen mainly based on their cost and availability. Some important pieces of equipment, such as the solubilization column and the pump, were already available when the HPWS was designed. The choice of the other components was based on the size of these pieces of equipment. The precision of some sensors was quite low but more precise equipment was too expensive. The number of tests was increased to compensate for the imprecision of these sensors. The following chapter presents the testing and evaluation of the HPWS.

# VIII EVALUATION OF A HIGH PRESSURE WATER SCRUBBER FOR CONTROLLED ATMOSPHERE STORAGE

## 8.1 Introduction

Controlling the level of gases in a sealed cold storage room at a given cold temperature and relative humidity has been called controlled atmosphere storage (CA). This process slows down the physiological activities of fruits and vegetables and prolongs their life in stores, warehouses, houses or other relatively warm places (Bartsch and Blanpied, 1984). The CA storage gas concentrations practically range from 0 to 10% of  $O_2$ , 0 to 30% of  $CO_2$ . N<sub>2</sub> remains the most important of the other gas components of CA storage (Kader, 1985; Salveit, 1985 and Meheriuk, 1985). The air we breathe contains 21% of  $O_2$ , 0.004% of  $CO_2$  and 78% of N<sub>2</sub>. Part of the O<sub>2</sub> is naturally absorbed in the respiration process of the stored commodities. The O<sub>2</sub> may also be eliminated using artificial processes such as burning or N<sub>2</sub> gas flushing (Raghavan and Gariépy, 1985).

There is a natural  $O_2$  uptake and a  $CO_2$  release in the respiration of the CA stored living plants, fruits or vegetables. An excessive accumulation of  $CO_2$  must be prevented as well as an excessive intake of  $O_2$ -rich air into the rooms. Natural ventilation cannot be used to flush out the  $CO_2$  since surrounding air contains too much  $O_2$ . A proper gas control system implies an airtight storage room, a system to partially replace the  $O_2$  and a system to eliminate the  $CO_2$ . Pflug (1960) demonstrated that a  $CO_2$  control system which did not introduce  $O_2$  into the storage room required approximately 25% less  $O_2$  pull-down time at the beginning of the storage season.

Since a perfectly airtight storage room is difficult to obtain on a commercial scale, the introduction of  $O_2$  by the  $CO_2$  control system could make the  $O_2$  pull-down period unacceptable, especially since the respiration of commodities in storage decreases with time (Vigneault *et al.*, 1991). In general,  $O_2$  pull-down by respiration alone occurs within 480 h of closure of the storage room. However, it has been proved, for some commodities, that an  $O_2$  pull-down period of five days results in a better post storage quality (Lau *et al.*, 1983 and Bartsch, 1986). The commercial use of  $O_2$  pull-down techniques did not follow the results of post harvest physiology research mainly because of the lack of a trustable, economical and efficient  $O_2$  pull-down system.

Vigneault and Raghavan (1991) presented a theoretical study demonstrating the feasibility of reducing the net  $O_2$  transport into the CA room using a high pressure water scrubber (HPWS). An HPWS can create a difference in  $O_2$  partial pressures that could result in a net transport of  $O_2$  out of the CA room.

The objective of this research was to build and test an HPWS and determine its ability to reduce the  $O_2$  entering the storage room while scrubbing the  $CO_2$  contained in the circulating gas. The investigation also aimed at examining the effects of  $CO_2$  scrubbing on the  $O_2$  pull-down period in CA storage. It was also intended to demonstrate that, at high pressure, there is a net transport of  $O_2$  out of the CA storage.

## 8.2 Material and Methods

# **8.2.1** Description of the Apparatus

The HPWS presented in Chapter 3 (Vigneault and Raghavan, 1991) was a design allowing pressurized air to be in contact with water (Fig. 8.1). It consisted of a compressor which pressurizes the air coming from the storage room and introduces it to a water stream. Air bubbles generated by the forced air rise to the top of a water column, during which time the  $O_2$ ,  $CO_2$  and  $N_2$  contained in the bubbles are absorbed by the water. Air from the top of the water column is returned to the storage room. Concurrently, water is pumped from a reservoir to the top of the water column. The water flows down to the bottom of the column absorbing gases so that equilibrium with their partial pressures is established, and then flows to the water reservoir. Since the water is under atmospheric pressure, it releases or absorbs the gases in a state of equilibrium.

An HPWS was built to test this concept. It consisted of both an air and a water circulation system (Fig. 8.2). The air circulation system consisted primarily of an air filter and a 3.75 kW compressor. The ambient air was pressurized, cooled to about 18°C using an air-water heat exchanger and stored in a 120 L compressed air reservoir. Compressed pure gas cylinders were used to supply  $N_2$  and  $CO_2$  to the air coming from the compressed air reservoir to produce the desired gas mixtures. The manual valves A and B were used







Figure 8.2: Diagram of the HPWS equipped with the instrumentation needed to assess its performance.

to regulate the compressed air and gas flows respectively. The gas mixture was then introduced through a diffuser located at 350 mm from the bottom of the solubilization column. After reaching the top of the water column, the remaining gas exhausted to the atmosphere through the manual valve C. This valve was adjusted to maintain the pressure inside the column at the desired level.

The solubilization column consisted of an airtight cylinder 600 mm in diameter and 1547 mm in height, having a semi spherical end and a capacity of 570 L. Two types of diffuser were used. The diffuser type A (Fig. 8.3a) consisted of a 230 mm long bar of sintered silica with a 35 mm square cross section. The diffuser type B (Fig. 8.3b) consisted of thirteen parallel cylindrical segments of sintered stainless steel, 20.3 mm O.D and having a total length of 4606 mm. The segments were positioned at 45 mm centre to centre to cover the entire cross section of the column. The specification of the bubble population generated by these two diffusers was measured using the methods described by Vigneault *et al.* (1992a). The Sauter mean diameters ( $d_{3,2}$ ) of the bubble population generated by the diffuser A and B are presented as a function of the gas flow rate by Eqs. 8.1 and 8.2.

$$d_{3,2} = 3.4 + \left(\frac{Q_i - 46}{46}\right)^{0.37} R^2 = 0.996$$
 (8.1)

(8.2)

 $d_{3,2}$  = Sauter mean diameter, mm;  $Q_i$  = gas flow rate through the diffuser, L•s<sup>-1</sup>•m<sup>-1</sup>; R = correlation coefficient.

Where

The description of the water circulation system starts with the lower water reservoir (Fig. 8.2). The water was circulated from this reservoir and through the system using a 10 kW pump having a nominal capacity of 2.5  $L \cdot s^{-1}$ . The water flow was controlled using the manual valve D. The entrance of the water was positioned at 3 mm from the top of the solubilization column. The water exited from the solubilization column by the exhaust pipe



Figure 8.3: Diagram of the position of the diffuser A and B in the solubilization column.

aperture centred at 5 mm from the bottom of the solubilization column. The water flowed to the upper water reservoir through five nozzles. Four nozzles were controlled by manual valves adjusted for coarse control to maintain the water level in the solubilization column at the desired level. The other nozzle was controlled by a solenoid valve for fine control. A water level control device opened and closed the solenoid value to automatically maintain the water level within  $\pm 5$  mm. The water level control device consisted of a magnet mounted on a floater which activated two sensors included in an electronic device controlling a solenoid valve. These sensors were mounted 10 mm vertical distance apart on a support. The support assembly was held on a vertical rod and was adjusted in an upright position to maintain the water at one of the four following levels: 300, 600, 900 and 1200 mm above the diffuser position. These water levels were chosen based on the theoretical exposure time required to solubilise the CO<sub>2</sub> contained in gas bubbles of different sizes (Bird et al. 1960). The heights were calculated using the terminal velocity of a bubble containing 5% CO<sub>2</sub>, submitted to a 0.5 MPa pressure and rising in pure water. As shown in Fig. 8.4, the height required to solubilise the  $CO_2$  increased with the size of the bubble, ranging from 80 mm to 1000 mm when the size of the bubble increased from 1 mm to 5 mm.

The water contained in the upper water reservoir overflowed in the lower water reservoir completing the water circuit. This two-step water reservoir was designed to prevent water from returning to the pump without achieving equilibrium with the atmospheric conditions. Cool tap water was continuously added to the lower reservoir to compensate water losses and maintain the water temperature around 18°C.

# **8.2.2** Monitoring System and Calibration

To evaluate the performance of the HPWS, the following parameters were measured through the water and gas systems. These were: temperature, gas and water flow rates, pressure, height of water column, gas concentrations and gas solubility.

The temperature was measured at five points using thermocouples. These points were: ambient air near the compressor (T1), in the gas mixture before entering the solubilization column (T2), at the gas exhausted from the system (T3), in the water before



Figure 8.4: Height required to solubilize the  $CO_2$  of gas bubble containing initially 5%  $CO_2$ , submitted at a 0.5 MPa pressure and rising through pure and quiescent water.

entering the solubilization column (T4) and at the water exit of the solubilization column (T5). The thermocouples were calibrated before their installation using a 0.2°C precision thermometer.

The gas flow rates were measured using rotometers at the three following points (Fig. 8.2): on the compressed air line (A), on the compressed gas line (B) and on the compressed air exhaust line (C). The water flow rate was measured before the water entered into the water solubilization column using a water flowmeter (electronic padded wheel type).

The pressure was measured using 0 to 1050 kPa  $\pm$  3.5 kPa precision pressure gauges at four different points in the system (Fig.2). These are: before both gas flowmeters (A and B), in the solubilization column (C) and before the water entry to the solubilization column. The pressure gauges were calibrated before installation by comparing their readings while mounted side by side on pressurized lines.

The water level in the solubilization column was measured with a measuring tape to an accuracy of  $\pm 1.5$  mm.

The gas was alternately sampled before entering the solubilization column and at the gas exhaust at a rate of 10 mL·s<sup>-1</sup>. The gas concentrations were measured using a  $O_2$ and  $CO_2$  gas analyzer. The N<sub>2</sub> gas concentration was calculated by difference. The gas analyzer was calibrated using two certified calibration gases containing respectively: 3.22%  $O_2$ , 4.85%  $CO_2$  and 91.93% N<sub>2</sub>; and 21.00%  $O_2$  and 79.00% N<sub>2</sub>. The accuracy of the gas analyzer was  $\pm 0.1\%$  for the two analyzed gases.

The solubilities of  $O_2$  and  $CO_2$  in the water used during the tests were measured twice a day according to the method described in Chapter 7 (Vigneault *et al.*, 1992b).

The water pump performance was determined by measuring the water flow rate and the pressure at the pressure gauge D. During this test, the valve D was fully opened and the system was operated at pressures ranging from 290 to 690 kPa.

The compressor performance was considered to be its capacity to maintain the air flow rates for different pressures inside the compressed air reservoir. The measurements were taken while the compressor was operated continuously and the pressure inside the compressed air reservoir was constant.
## 8.2.3 Experimental Design

The performance of the HPWS was evaluated by varying the following parameters: the pressure and the height of the water in the solubilization column, the water flow rate, the composition and flow rate of the gas entering into the system and the type of diffuser. The tests were carried out using the conditions presented in Table 8.1. At least three values covering the range of the system capacity were tested for each parameter. Each set of conditions tested was replicated twice. For each test, the water flow rate and the pressure inside the solubilization column were first adjusted. The water output flow rate was then adjusted to maintain the height of water at the desired level. The input gas flow rate and the concentration of gas were adjusted. Finally, the output gas flow rate was adjusted so as to maintain all the desired conditions stable. The parameters were measured when the flow rate and the concentration of the output gas were stable for at least five minutes. All tests using diffuser A were conducted first. This diffuser was then replaced by diffuser B.

# 8.2.4 Effect of CO<sub>2</sub> Scrubbing on O<sub>2</sub> Pull-Down Period

The effect of using a HPWS to control the  $CO_2$  in a CA storage room was evaluated as shown by Vigneault and Raghavan (1991). It simulated the conditions which exist in a 1818 m<sup>3</sup> room. The stored product selected was 410 t of McIntosh apples. The space factor was 3.6 m<sup>3</sup> of air per m<sup>3</sup> of stored product. O<sub>2</sub> and CO<sub>2</sub> levels in the room were 2.5% and 5% respectively. The room was considered to be airtight and the ventilation was operated as required to maintain the O<sub>2</sub> level at 2.5%. The air and water circulation rate through the water scrubber were based on the performance of the HPWS and the quantity of CO<sub>2</sub> to be scrubbed.

When a water scrubber system is used, there is  $O_2$  transportation through water circulation. There is also air infiltration due to the vacuum created by the gas transportation from the storage room. This air containing 21%  $O_2$ , increases the mass of  $O_2$  contained in the storage room. A so-called hybrid system based on using  $N_2$  pure gas to compensate for the gas transportation of the scrubbing system was used as suggested by Pflug (1960). The performance of the HPWS system was compared with a conventional water scrubber system and hybrid systems.

	HPWS.					
Type of Diffuser	Height of Water Column	Internal Pressure	Water Flow	Gas Composition		Total Input Gas flow
	(mm)	(kPa)	(L/s)	%O <sub>2</sub>	%CO <sub>2</sub>	(L/s)
A (260)*	300 (40)	140 (32)	0.7 (102)	21 (125)	0 (162)	0.6 (81)
	600 (78)	280 (78)	1.4 (114)	16 (98)	3 (33)	1.3 (113)
	900 (58)	420 (39)	2.1 (44)	10 (33)	6 (33)	2.0 (48)
	1200 (84)	550 (111)		5 (32)	12 (32)	2.8 (18)
B (284)	300 (39)	140 (35	0.7 (125)	21(138)	0 (182)	0.6 (122)
	600 (91)	280 (84)	1.4 (117)	16 (102)	3 (34)	1.3 (72)
	900 (63)	420 (63)	2.1 (42)	10 (23)	6 (34)	2.0 (65)
	1200 (91)	550 (102)		5 (21)	12 (34)	2.8 (25)

Table 8.1:Operation levels of the different parameters tested in the evaluation of THE<br/>HPWS.

number of tests performed for the category in indicated in parantheses.

# 8.3 Results and Discussion

#### 8.3.1 Pump Performance

The maximum water flow rate the pump was able to supply is presented in Fig. 8.5 as a function of the operating pressure. The capacity of the pump corresponded to Eq. 8.3.

(8.3)

Where  $Q_g = maximum gas flow rate, L \cdot s^{-1};$ 

P = absolute operating pressure, MPa.

The pump could not adequately supply water when the pressure was in excess of 0.69 MPa. At this pressure the pump cavitated and the pressure had to be released completely before the pump supplied water again. Based on this result, the experimental design was set not to exceed 0.56 MPa.

#### **8.3.2** Compressor Performance

 $Q_g$ 

Ρ

The maximum gas flow rate the compressor supplied is presented in Fig. 8.6 and corresponded to Eq. 8.4.

$$Q_{\rm r} = 2.98 - 0.42P$$
  $R^2 = 0.3842$  (8.4)

Where

= maximum gas flow rate,  $L \bullet s^{-1}$ ;

= absolute operating pressure, MPa.

The maximum gas flow rate presented a relatively high variability when expressed only as a function of the operating pressure. Nevertheless, the results obtained were judged sufficiently clear to establish an experimental design. The maximum gas flow rate to be tested was established at 90% of the flow rate calculated using Eq. 8.4.



Figure 8.5: Maximum water flow rate supplied by the pump as a function of the pressure.



Figure 8.6: Maximum gas flow rate supplied by the compressor as a function of the pressure.

## 8.3.3 HPWS Performance

ε

A total of 544 tests were conducted. The number of tests conducted for each given value of the parameters is presented in Table 8.1. Before analyzing the results, the gas flow rates and the solubility of the gas were corrected for temperature. In fact, the effect of temperature on the mass transfer is usually neglected (Marais, 1975). Only its effect on solubility is important.

The model used for the statistical analysis of the results was:

(8.5)

Where	$Q_{og}$	=	output gas flow rate, $L_s s^{-1}$ ;
	$Q_{ig}$	=	input gas flow rate, $L_s s^{-1}$ ;
	Q <sub>w</sub>	=	water flow rate through the system, L $s^{-1}$ ;
	Sg	=	solubility of the gas, $L_s$ of gas $L^{-1}$ of water;
	Н	<b>=</b> .	height of the water above the diffuser in the solubilization
			column, mm;
	d <sub>3,2</sub>	=	Sauter mean diameter of the bubble population, mm;
	a <sub>i</sub>	=	regression coefficient (i = 0, 1, 2, 3);

= experimental error associated with each set of data.

The unit  $L_s$  is the standardized litre of gas, that is, the volume occupied by the mass of gas at 0°C (273.15°K) and 101.3 kPa absolute pressure. The terms  $Q_{og}$ ,  $Q_{ig}$  and  $Q_wS_g$  represent the mass balance of the system assuming the water to be saturated before leaving the solubilization column. Since the saturation of water is proportional to H and inversely proportional to the  $d_{3,2}$ , the term H/ $d_{3,2}$  was added. The effect of this term should not be significant if saturation is reached.

A Stepwise regression analysis was performed on the results obtained. The procedure rejected any variable in the model which did not meet the 0.05 significance level. The values of the  $a_i$  coefficients are presented in Table 8.2 for the three gas tested. The relative importance of the effects of  $Q_{ig}$ ,  $Q_wS_g$  and  $H/d_{3,2}$  on  $Q_{og}$  is represented by the partial  $R^2$ . The differences between the output gas flow rates calculated using the regression equation and the measured ones, so-called residuals, were calculated and presented

Gas	Parameter	Coefficient	Value	Partial R <sup>2</sup>	F	Р
<b>O</b> <sub>2</sub>		a <sub>0</sub>	0.0623		613.6	0.0001
	$Q_{ig}$	$\mathbf{a}_1$	0.641	0.9461	9437.5	0.0000
	$Q_w S_g$	a <sub>2</sub>	-0.219	0.0016	17.6	0.0001
	H/d <sub>3,2</sub>	a <sub>3</sub>	5.51×10 <sup>-6</sup>	0.0006	6.4	0.0117
CO <sub>2</sub>		a <sub>0</sub>	0.0031		5.2	0.0232
	$Q_{ig}$	a <sub>l</sub>	0.314	0.6180	712.4	0.0001
	$Q_w S_g$	a <sub>2</sub>	-0.0462	0.1543	145.9	0.0001
	H/d <sub>3,2</sub>	a <sub>3</sub>	4.28×10 <sup>-6</sup>	0.0111	10.2	0.0017
N <sub>2</sub>		a <sub>o</sub>	0.122		67.6	0.0001
	$Q_{ig}$	a <sub>i</sub>	0.767	0.9084	5506.2	0.0001
	$Q_w S_g$	a <sub>2</sub>	rejected*			
	H/d <sub>3,2</sub>	a <sub>3</sub>	5.78×10 <sup>-5</sup>	0.0026	15.7	0.0001

Table 8.2:Value and significance level of the parameter for each gas tested using the<br/>HPWS.

\* This variable did not meet the 0.05 significance level.

graphically in Fig 8.7 for the case of  $CO_2$ . Similar results were obtained using the results of the two other gases. These results did not indicate any apparent error in using the proposed model to correlate  $Q_{og}$  to the other parameters.

In general, the input gas flow rate was the most important parameter. The water flow rate combined with the solubility was relatively important only in the case of  $CO_2$  and did not have any significant effect on the N<sub>2</sub> output gas flow rate. This is likely justified by the relatively high solubility of  $CO_2$  which is, about 35 and 80 times those of  $O_2$  and N<sub>2</sub>, respectively.

Although the  $H/d_{3,2}$  was also significant, it exhibited very little effect on the output gas flow rate for each gas. Based on the Fig. 8.4, the combined effect of the height with the water in the solubilization column and size of the bubble should have been much more important. This unexpected result was investigated and it was highly suspected that the gas continued solubilising in water after reaching the water surface. This solubilization happened in the water forming a falling film on the wall of the solubilization column. This film of water was in continuous contact with the gas and its surface contact was inversely proportional to the height of the water in the column. It was likely that the solubilization process occurring in the falling film compensated for the lack of solubilization occurring while using a low level of water. Thus, the measure of the combined effects of H and  $d_{3,2}$  was in reality a measure of the difference of the efficiency of the rising bubble in the solubilization process. To really measure the effect of H and  $d_{3,2}$ , the design of the HPWS should have consisted of a system in which the falling film is not present.

The effect of the operating pressure on the CO<sub>2</sub> concentration of the exhaust gas of the HPWS under from a 1.5 L s<sup>-1</sup> gas flow rate of 5% O<sub>2</sub>, 5% CO<sub>2</sub> and 90% N<sub>2</sub> conditions is presented for three water flow rates in Fig. 8.8. By increasing either the water flow rate or the pressure, it is feasible to completely eliminate the CO<sub>2</sub> from the gas output. The CO<sub>2</sub> concentration reduced from 5% to 0.1% while using a water flow of 2.1 L s<sup>-1</sup> and an operating pressure of 0.55 MPa. These results were much lower than what was predicted by Vigneault and Raghavan (1991) while using the theoretical value of distilled water. The difference between the gas solubility in the water used during the test and the solubility of



Figure 8.7: Residual of the  $CO_2$  flow rate measured as a function of the estimated  $CO_2$  flow rate.



Figure 8.8:  $CO_2$  concentration in the gas outlet as a function of the pressure inside the solubilization column for three water flow rates while using a gas inlet flow rate of 1.5 L•s<sup>-1</sup>, an O<sub>2</sub> and a CO<sub>2</sub> concentrations of 5% each.

the gas in distilled water cannot explain more than 5% of the difference between the results obtained and the predicted one. Indeed, the solubility of the three gases measured during the test period was not lower than 95% of the one in distilled water in all cases.

The  $CO_2$  saturation of the water was about 10 to 25% depending on the experimental conditions while some  $CO_2$  was still present in the exhausting gas. For the same conditions, the  $O_2$  saturation reached 75 to 90%. To completely eliminate the  $CO_2$  from the exhausting gas, the water flow rate was required to be maintained at too high a level, solubilizing a large amount of  $O_2$  and  $N_2$ .

At 21%  $O_2$ , the  $O_2$  output flow rate was lower than the input flow rate. The output/input O<sub>2</sub> gas flow rate varied between 83% to 93% depending on the pressure and the total input gas flow rate (Fig. 8.9). This indicated a net transport of the  $O_2$  solubilised in the water. The difference in O<sub>2</sub> concentration increased with pressure and gas flow rate (Fig. 8.10). However, the decrease in  $O_2$  concentration did not exceed 1.9% in any case. These reductions in  $O_2$  concentration were much lower than the predicted value presented by Vigneault and Raghavan (1991). This was likely due to the solubilization of N<sub>2</sub> gas reducing the total gas flow rate leaving the HPWS and increasing the O<sub>2</sub> concentration of the exhausting gas. While calculating the  $N_2$  saturation in water, results showed a fairly high solubilization. In fact, the amount of N<sub>2</sub> exhausting from the solubilization column showed generally a 100% of N<sub>2</sub> saturation of water and even more in some cases. This over saturation of water in the case of N2 only cannot be clearly explained. Some gas bubbles might have been swept along by the water current but this was unlikely. The vertical velocity of the water corresponding to the maximum flow rate, 2.1 L s<sup>-1</sup>, was 1.8 mm s<sup>-1</sup>. At this velocity, only bubbles smaller than 0.06 mm would be swept out by the water current (Clift et al., 1978). This diameter corresponded to a bubble size 10 times smaller than the smallest  $d_{3,2}$  used during the test. Even if some bubbles might have been this size, their total volume could not have accounted for a large portion of the total gas flow rate. The experimental error was the most logical explanation due to the relatively low precision of the three air flowmeters used; but this has not been proved.



Figure 8.9: Output/input gas flow rate ratio (%) as a function of the pressure inside the solubilization column for three gas inlet flow rate (Q<sub>i</sub>) having an  $O_2$  concentration of 21% and using a water flow rate of 1.4 L•s<sup>-1</sup>.



Figure 8.10:  $O_2$  concentration in the gas outlet as a function of the pressure inside the solubilization column for three gas inlet flow rate (Q<sub>i</sub>) having an  $O_2$  concentration of 21% and using a water flow rate of 1.4 L•s<sup>-1</sup>.

When the  $O_2$  concentration of the input gas decreased, the concentration of the  $O_2$  stopped decreasing and even increased as shown in Fig. 8.11. The difference in the efficiency of the HPWS in decreasing the  $O_2$  concentration was lower when using a higher gas flow rate (Fig. 8.9). The low efficiency of the HPWS in decreasing the  $O_2$  concentration of the treated gas, even when this gas contained 21%  $O_2$ , made this system uncompetitive with other  $O_2$  pull-down methods. It might be interesting to use the system to eliminate the  $CO_2$  from a CA storage room since it would allow fairly little  $O_2$  to enter the room. However, this advantage alone does not justify the investment cost of the system.

# 8.3.4 Comparison of an HPWS and a Conventional Water Scrubber

In all the simulated cases, the systems were operated when the  $CO_2$  concentration inside the storage room exceeded the desired level (5%). The room was ventilated using surrounding air when  $O_2$  was lower than 2.5%. The results obtained for the four systems are presented in Fig. 8.12.

While using the conventional system, the simulation showed that a 417 h period from the sealing time of the storage room was required to reach 2.5%  $O_2$  (line A, Fig. 8.12). This corresponded to the  $O_2$  pull-down period encountered in commercial apple storage facilities using commodity respiration to decrease the  $O_2$  concentration inside the room.

The hybrid conventional water scrubber system using  $N_2$  to compensate the gas losses required a 312 h O<sub>2</sub> pull-down period to reach 2.5% O<sub>2</sub> (line B, Fig. 8.12). This was approximately 25% shorter than the O<sub>2</sub> pull-down period obtained while using a conventional water scrubber system as predicted by Pflug (1960).

The HPWS using outside air as a compensating gas for gas losses (line C, Fig. 8.12) did not reach the 2.5% level of  $O_2$  within the maximum  $O_2$  pull-down period required by regulations (Vigneault and Raghavan, 1991). This was due to the fact that too much gas, including  $O_2$ ,  $CO_2$  and  $N_2$  was absorbed by the water circulating through the HPWS. This large amount of gas being replaced by air containing 21%  $O_2$  resulted in slowly decreasing  $O_2$  in the storage room.



Figure 8.11:  $O_2$  output gas concentration as a function of the  $O_2$  input gas concentration for two pressures(P) inside the solubilization column using a gas flow rate of 1.5 L•s<sup>-1</sup> and the water flow rate of 1.4 L•s<sup>-1</sup>.



Figure 8.12: O<sub>2</sub> concentration as a function of the time resulting from the use of four CO<sub>2</sub> control system: A) water scrubber; B) hybrid water scrubber; C) HPWS and D) hybrid HPWS.

The hybrid HPWS system (line D, Fig. 8.12) reached the 2.5%  $O_2$  level within 208 h. This period was still too long to be considered "Rapid CA". This long period was due to the poor efficiency of the HPWS at eliminating the  $O_2$  contained in the air. From this simulation, it can be deduced that the use of a HPWS to scrub the  $CO_2$  from a CA storage room is not worthy. A conventional system combined with pure  $N_2$  system compensating the gas losses due to scrubbing would be much more appropriate for normal  $O_2$  pull-down. Other techniques should be investigated for "Rapid CA" storage.

# 8.4 Conclusion

An HPWS was built, instrumented and tested under conditions usually encountered during and after the  $O_2$  pull-down period of the CA storage of fruits and vegetables. The capacity of the pump to circulate water through the HPWS was too low to permit the testing of the HPWS at a pressure superior to 0.55 MPa. From this investigation, it can be concluded that:

1) The HPWS is effective in decreasing the  $CO_2$  concentration of the treated gas.

- 2) By increasing the water flow rate or the pressure, it is possible to completely eliminate the  $CO_2$  from the gases circulating through the HPWS but a fairly large amount of  $O_2$  and  $N_2$  will be solubilized under these conditions.
- 3) The reduction of  $O_2$  is fairly low. It does not exceed 1.9% in any case.
- 4) It is feasible to reduce the net  $O_2$  transport into a CA room.
- 5) Using a hybrid HPWS could lead to a reduction in the  $O_2$  pull-down period.
- 6) The net transport of  $O_2$  out of the CA room is not sufficient to justify the investment of building an HPWS. Alternatives for "Rapid CA" storage should be investigated.

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## IX GENERAL DISCUSSION AND CONCLUSION

The length of the  $O_2$  pull-down period is inversely related to the post-storage quality of produce stored in CA conditions. Rapid establishment of CA, results in a better post storage quality for some commodities. In general, the best results are obtained when the storage room was filled, cooled, and flushed to 5%  $O_2$  in no more than five days.  $O_2$  pulldown techniques have yet to be applied commercially due to the lack of a trustable, economical and efficient  $O_2$  pull-down system.

Since a perfectly airtight storage room is difficult to obtain on a commercial scale, the introduction of  $O_2$  by the  $CO_2$  control system could make the  $O_2$  pull-down period unacceptable. It was shown that some  $O_2$  is introduced to the storage room by the water scrubber when it is used to control the  $CO_2$  concentration. A theoretical study demonstrated the feasibility of reducing the  $O_2$  pull-down period in CA storage with an HPWS that minimizes the amount of  $O_2$  entering the storage while scrubbing the  $CO_2$ . This study also highlighted the importance of the effect of the following parameters on the performance of a HPWS: the bubble diameter, the water level in the solubilization column, the pressure, the air and water flow rates and the quality of the water used in the HPWS which affects the solubility of the gas. Affordable apparatuses or techniques exist for measuring most of these parameters except bubble size and gas solubility.

A method based on digital image analysis was developed to automate the measurement of bubble size and vertical velocity in a non-intrusive manner. Bubbles of diameters varying from 0.3 to 4.0 mm were detected and measured using a thresholded digitized image. The image system was calibrated by comparing diameters measured by a camera system to the ones deduced from gas flow rate and bubble production rate measurements for both fixed and moving capillary tube bubble generators. The differences between the results obtained using these two methods of measurement were less than 0.05 mm for each diameter measured and were not correlated with bubble size. Furthermore, the results from the two methods of measurement were not significantly different. The measured vertical velocity data were compared to published data.

However, there were two drawbacks in this technique. These were; the error generated by the effect of the offset distance between the bubble position and object plane of the camera; and the density of the cloud of bubbles generated by gas diffuser. The two problems associated with the measuring technique used were solved as follows:

First, a discriminatory parameter was conceived for the characterization of clouds of bubbles. This parameter was the magnitude of "jump" in the slope on the curve of grey levels versus pixel position at the bubble boundary. This "jump" decreased as the field of view of the camera was moved away from the bubble position. It was demonstrated that the discriminatory parameter was independent of bubble size for the system under analysis. It also showed that a depth of field of 6.67 mm corresponded to the depth of the sample space. The use of this parameter therefore allowed discrimination of bubbles in and out of the sample space. Within the sample space, the error in bubble diameter was related to the magnitude of the discriminatory parameter. A single equation was written to relate the apparent diameter of the measured bubble to its real size. The accuracy of measurement of bubbles contained in the sample space was within  $\pm 50\mu$ m.

Second, a mechanical bubble sampler which permits isolation of a representative sample of the population was devised, built and tested. Test results showed that the samples had a size distribution and mean ratio of volume to surface which were not significantly different from those of the population. The sampling technique combined with air flow and pressure measurements facilitated the characterization of the bubble generator used in the high pressure water scrubber.

A simple pressure method for determining the solubility of pure gases in liquid was developed. The results obtained using this method for  $CO_2$ ,  $O_2$  and  $N_2$  solubility in distilled water were compared to published data to assess its accuracy. The differences between the measured and published data did not exceed 3% and the mean difference for each gas did not exceed 0.6%. An error analysis led to optimization of the testing conditions and, consequently, to better precision. This simple method was used to measure  $CO_2$ ,  $O_2$  and  $N_2$  solubilities in water of varying quality used in the development of the HPWS.

An HPWS was built and tested under typical controlled atmosphere (CA) conditions. The results obtained demonstrated the efficiency of the scrubber to eliminate the  $CO_2$ . The  $O_2$  output flow rate was generally lower than the  $O_2$  input flow rate.

However, at the gas concentrations and pressures tested, the  $O_2$  concentration of the entering gas minus that of the exhausting gas was greater than 1.9 %. This low efficiency at reducing the  $O_2$  led to the conclusion that this system could not be used for "Rapid CA". Moreover, operation of the HPWS using outside air as the compensating gas did not lead to 2.5%  $O_2$  within the 480 h  $O_2$  pull-down period allowed by regulations.

Other techniques should be investigated for "Rapid CA" storage.

# **X CONTRIBUTION TO KNOWLEDGE**

The following are original contributions to knowledge:

- A method based on digital analysis to automate the measurement of bubble size in a non-intrusive manner.
- 2) A discriminatory parameter to facilitate the measurement of bubble size in a 3-D environment.
- 3) A mechanical bubble sampling technique which enables the isolation of a representative sample of the population.
- 4) A simple method for measuring the solubility of pure gases in liquid.
- 5) Performance evaluation of a high pressure water scrubber used to eliminate  $CO_2$ and  $O_2$ .

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