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MASS TRANSFER IN ULTRASONIC ASSISTED ATMOSPHERIC FREEZE DRYING

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Abstract: Mass transfer rates and specific moisture evaporation rates (SMER) for different products were compared for an atmospheric freeze drying process with and without airborne ultrasonic assistance. Mass transfer was increased by as much as 23.2% and with an efficient airborne ultrasonic transducer, additional SMER of 0.1 to 0.4 kg_{water} kWh^{-1} are possible. The improved heat and mass transfer in ultrasonic assisted atmospheric freeze drying is most likely caused by effects at the interface between the product and drying agent. Therefore, airborne ultrasound has not only potential to accelerate freeze drying, but also other processes such as low temperature drying or freezing of food.

Keywords: acoustic drying, low temperature drying, freeze drying, airborne ultrasound

INTRODUCTION

Drying products below their freezing point is a dewatering method for high quality and/or temperature sensitive products. The low temperature protects the product from undesirable enzymatic activity, structural changes during thawing, and also results in a porous product structure with a good rehydration factor. Freeze drying is mainly conducted under vacuum (VFD), below the triple point of water/ice, which is an expensive dewatering method due to its high operational investment. Alternatively, freeze drying can be carried out at ambient pressure (AFD) which results in a product quality that is similar to VFD but with a lower operational investment when energy is recovered in a heat pump system.

Typically, AFD is carried out at temperatures between -5°C and -10°C where the vapour pressure of water is between 400Pa and 250Pa respectively. Depending on the product type, it is also possible to dry near the initial freezing point of the product, where the high vapour pressure of ice results in the highest possible sublimation rate. Drying rates for AFD can be faster than for VFD, as shown by Bantle et al. (2009) and Di Matteo et al. (2003). Drying systems can be combined with heat pumps to recover the evaporation energy (see Fig. 1). The moist air is dehumidified and cooled at the cold surfaces of the evaporator of the heat pump. Energy is thereby transferred to the refrigerant. In the condenser this energy is returned to the drying system by reheating the dehumidified drying air. Strømmen et al. (2002) show that 60-80% of the energy can be saved in heat pump assisted drying (HPD) at the same drying temperature. Colak and Hepbasli (2009a) have reviewed heat pump drying and confirm that in HPD a SMER up to 4 kg_{water} kWh^{-1} can be reached, whereas the SMER for a normal drying system is between 0.12-1.28 kg_{water} kWh^{-1}, compared to 0.72-1.2 kg_{water} kWh^{-1} respectively for VFD. Colak and Hepbasli (2009b) als present HPD applications.

Energy can also be recovered in AFD with a heat pump system, but the low sublimation rates result in long drying times. Accelerated drying can be realized in AFD by crushing the product into fine particles with a high specific surface area and drying it in fluidized or stationary beds (Bantle, et al., 2009). This is also advantageous for products that cannot exist as particles in an unfrozen condition as a result of their physical properties. However the sublimation rate in AFD is the limiting factor and therefore additional sublimation and accelerated mass transfer would be beneficial. In a feasibility study, Alves-Fihlo and Eikevik (2008) outline the potential of ultrasound as an additional energy input into a drying system.

Ultrasound (US) is a cyclic sound pressure wave with a frequency between 20kHz and 1Mhz, which means that it is higher than the frequency of the human sense of hearing. Animals use ultrasound, such as the natural sonar of whales or echolocation of bats. Industrial applications are ultrasonic welding, cleaning, disintegration, humidifiers, particle characterization and non-destructive testing, while in medicine it is used in diagnostics and therapeutics. Ultrasound requires a resonance media, which can be the water in an ultrasonic cleaning bath. Recent developments have increased the efficiency of ultrasonic transducers for air to 80% and to intensity levels of 175dB (Gallego-Juárez et al., 2007). Therefore new applications for so-called airborne ultrasound can be imagined, such as the precipitation of smoke and powder and destruction of foams (Riera et al., 2006), improved product quality under freezing (Xu et al., 2009), and an accelerated drying or dehydration rate when ultrasonic is used (García-Pérez et al., 2006, Khmelev et al., 2006).

Using airborne ultrasound in a drying system was first investigated in the 1950s when researchers such as Boucher (1958) concluded that ultrasound can accelerate drying at lower temperatures. Muralidhara et al. (1985) reviewed ultrasonic drying and identified 10 different effects that can lead to accelerated drying rates with ultrasound. With the recent developments and efficiencies for airborne ultrasonic transducers industrial applications in drying will be profitable. Ultrasound applied prior to drying has also been shown to give accelerated drying rates (Fernandes and Rodrigues, 2008 and Duan et al., 2008). Accelerated drying rates in ultrasonic assisted drying have been reported for lower velocities (García-Pérez et al., 2007) and for lower temperatures (Mulet et al., 2003). Mulet et al. (2003) reported the same diffusivity for ultrasonic assisted drying of carrots at 22°C as for drying without ultrasound at 60ºC. Therefore, lower drying temperatures can be used, which will improve the product quality. The use of ultrasound in a freeze drying process has not been investigated to date, but appears to be a promising technology to accelerate drying rates while preserving product quality.

In this feasibility investigation, different products were dried at atmospheric pressure in a frozen condition with and without ultrasonic assistance in a fluidized or stationary bed. Mass transfer during the first hours of drying with ultrasound is compared with the same drying process without ultrasound.

MATERIALS AND METHODS

The HPD system used in this investigation has previously been described by Bantle et al. (2009). The drying chamber was modified with an ultrasonic transducer (Sonotronic, DN 20/2000, 20kHz, diameter 100 mm, titanium) which was placed 10mm underneath the bottom plate of the fluidized bed (Fig. 1). The bottom plate is a 1mm hole perforated plate. The transducer was modified so it could be used as airborne ultrasonic source. Depending on the product, the power consumption of the transducer is between 80 – 120W. In earlier tests in cooperation with the supplier the efficiency of the airborne system was determined to be around 20%, which is acceptable since the transducer was originally designed for welding purposes.

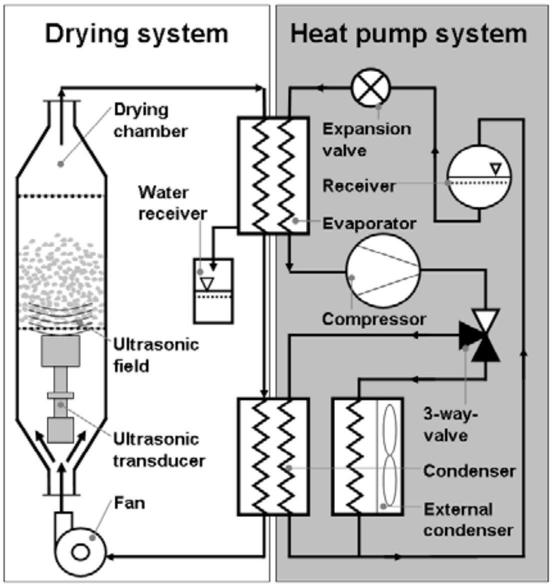

Fig. 1. Drying system with heat pump energy recovery and ultrasonic assistance

The drying chamber had a diameter of 200mm and a height of 300mm. It was sealed with a 300µm sieve and can contain approximately 1 kg of product. The drying tests were performed three times for each product with the same drying time and input weight each time. The weight reduction was determined manually by interrupting the drying process since the ultrasound made it impossible to conduct online weight measurement. The power consumption of the ultrasonic generator, temperatures and the relative humidity of the drying air were recorded below and above the drying chamber. Depending on the fluidization characteristics of the product, drying was performed in a stationary bed with manual mixing during the weight measurement, or in a fluidized bed with a velocity 10-20% higher than the minimum fluidization velocity. The drying temperature was stable at -3°C and -6°C, respectively and therefore below the initial freezing point of the products and in the normal range of AFD. The average mass transfer rates were evaluated from the measured weight reduction over drying time.

The products that were dried were apple, peas and cod, all of which were purchased at a local supermarket. All products were processed (crushed or cut) into particles and frozen in liquid nitrogen and stored at -30°C until drying. The zooplankton species *Calanus finmarchicus* (CF) was included in this investigation. *Calanus finmarchicus* is a small crustacean from the lower levels of the food chain with a size from 0.1mm to 3mm, depending on its stage of development. It represents an enormous amount of biomass in the ocean and is a natural resource for aquatic lipids and proteins. The use of this marine life form is currently being investigated since it can be harvested in industrial batches with newly developed trawls (Bailey et al., 2008). Batches of CF were harvested between the islands of Hitra and Frøya (GPS: 63º30N, 9º55E) outside of Trondheim Fjord, Norway at the end of April 2009, frozen in plates and stored at -80°C. The catch consisted mostly of CF at stages four and five, which is in accordance with Tokle (2006).

In order to determine the sublimation rate of a pure ice surface under AFD, water was frozen, crushed and classified with sieves in a frozen condition. Prior to the AFD the products (ice, CF, cod, apple and peas) were tempered for 24 hours at their drying temperature in a climate chamber. An overview of the products, the particle sizes and drying conditions can be found in Table 1. Three drying experiments were performed for each product with and without ultrasonic assistance. For the drying test without ultrasonic assistance, the transducer was not removed in order to retain the same flow profile in the drying chamber. The average mass reductions from three drying experiments were compared for the different products with and without ultrasonic assistance. SMER values were calculated for the average additional sublimated ice under ultrasound based on the power consumption of the transducer.

Table 1. Product characteristics and drying conditions for the atmospheric freeze drying experiments performed with and without ultrasonic assistance.

Product	Size [mm]	Drying time [h]	Drying temp. [°C]	Rel. humidity	Bed condition	Velocity [m/sec]	Input [g] ±2%	Moisture $\%_{\text{wet base}}$
Ice	$Ø_{50}$= 2 ±1	1	-5 ±0.3	20 ±3 %	fluid	1.20 ±0.12	1000	100
CF	$Ø_{50}$= 3 ±1	3	-6 ±0.4	22 ±5 %	fluid	1.45 ±0.18	800	81.4
Cod	l_{cube}= 14 ±2	3	-6 ±0.3	22 ±5 %	stat./fluid	1.79 ±0.36	1000	82.2
Apple	l_{cube}= 8 ±2	3	-6 ±0.3	24 ±7 %	stat.	2.47 ±0.15	1000	85.8
Peas	$Ø_{50}$= 8 ±1	3	-6 ±0.4	20 ±6 %	fluid	1.49 ±0.20	1000	76.7
Peas	$Ø_{50}$= 8 ±1	3	-3 ±0.3	20 ±6 %	fluid	1.66 ±0.28	1000	76.7

RESULTS

Three drying experiments were performed for each produt with and without ultrasonic assistance. Figure 2 shows the averaged drying curves (reduction of moisture with time) for ice particles and cod cubes with and without ultrasonic assistance. The drying time in all tests was 3 hours, respectively 1 hour for the drying of ice. The average sublimation or mass transfer rate was calculated from the weight reduction before and after drying (Table 2). Ice showed the highest sublimation rate and a linear weight reduction over time (Fig. 2), since there is no boundary between the drying agent and ice surface. The other products (CF, cod, apple and peas) had a slower and decreasing sublimation rate over time, because the outer shell of the drying product becomes an ever-thicker barrier to the mass transfer as soon as sublimation begins. This is normal for freeze drying where no first drying stage occurs. During all tests, no significant particle breakdown was observed although there was some minor shrinkage in the partly dried product (see also Bantle et al., 2009). Temperatures and relative humidities for all tests were recorded and showed no discrepancy between the tests with and without US (Table 1). The generally higher sublimation rate in US-AFD did result in a slightly higher relative

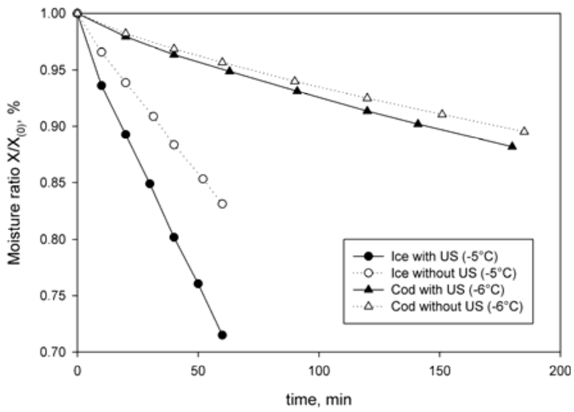

Fig. 2. Drying curve for atmospheric freeze drying of ice particles and cod cubes with and without ultrasonic assistance.

humidity (≈+2-5%) compared to AFD, whereas the temperature showed no significant or measurable difference over time. Also the higher sublimation rate in US-AFD did not influence the performance of the generously dimensioned heat pump system.

Table 3 shows the additional SMER values for the tests that were undertaken with an airborne ultrasonic efficiency of 20% for the transducer. The values are based on the average additional sublimation of ice under ultrasound and average power consumption of the ultrasonic generator. Using an airborne ultrasonic transducer with an efficiency of 80% (Gallego-Juárez et al., 2007) would increase this values by the factor of 4. US-AFD of ice results in the highest additional SMER value, followed by the marine products (Cod and CF), peas and apple. The drying temperatures of -6°C and -3°C did not influence SMER values for peas, although a difference in the sublimation rate occurred.

Table 2. Average sublimation rates for different products during atmospheric freeze drying with and without ultrasonic assistance.

Product	Sublimation $10^{-1} kg_{ice} kg_{d.m.}^{-1} h^{-1}$		Difference
	without US	with US	
CF	8.54	9.15	7.2%
Cod	6.08	7.15	17.5%
Apple	8.86	9.84	9.9%
$Peas_{-6°C}$	3.37	3.77	11.9%
$Peas_{-3°C}$	4.47	4.74	6.0%
	$10^{-1} kg_{ice} kg_{ice}^{-1} h^{-1}$		
Ice	1.77	2.18	23.2%

Table 3. Additional SMER values for atmospheric freeze drying assisted by airborne ultrasound.

Product	Additional SMER for airborne ultrasound $kg_{H_2 0} kW^{-1} h^{-1}$	
	US efficiency 20%	US efficiency 80%
Ice	0.225	0.899
CF	0.110	0.439
Cod	0.105	0.419
Apple	0.028	0.110
$Peas_{-6°C}$	0.091	0.365
$Peas_{-3°C}$	0.093	0.373

DISCUSSION

US-AFD can increase the sublimation rate up to 23.2% compared to AFD (Table 2) and will also improve the SMER of a system (Table 3). Muralidara et al. (2008) identified several possible effects of the use of US in a drying system:

1. Effects caused by cavitation:
    a. Cavitation breaks down particles, which will increase the specific surface area.
    b. Cavitation degases the liquid, which will improve the liquid flow.
    c. Cavitation disperses particulates or agglomerations, which would improve the particle contact with the drying agent.
    d. Caviation bubbles caused by US will coagulate fibrous and clinging particles (e.g. wood paper pulp). This effect can improve product quality but not necessary drying rate.
    e. Cavitation produces free chemical ions.
2. US changes the viscosity or the structural properties of the drying agent.
3. US cleans or clears the surfaces of the product, which will increase surface area in contact with the drying agent.
4. US can increase the mass transfer at a gas/liquid interface, by affecting the local pressure gradient between the drying agent and product (vapour pressure and saturation vapour pressure).
5. “Sponge” effect: the product is compressed and decompressed in the US field. The water is thereby squeezed out of the product, similar to when a wet sponge is squeezed. The “sponge” effect is often used to explain accelerated drying rates (e.g. Gallego-Juárez et al., 2007).
6. US can create high turbulence around the product, which will improve the mass transfer rates between the product and drying agent.
7. Other effects, such as Oseen forces (a rectified force attributable to the nonlinearity of high intensity waves in air) or Bernoulli forces (forces of attraction due to reduced pressure in a narrowed passageway, such as the movement of gas between stationary objects in close proximity).

Products that are dried in a frozen state have a different physical structure than products dried above the freezing point (hot air drying). In AFD, the water to be removed exists as ice, which will give a solid/solid structure and a mass transfer towards the drying agent. In hot air drying, the liquid water is enclosed in dry matter, which is a solid/liquid system. Therefore a first drying stage with free surface water results in a stable evaporation rate at the beginning of hot air drying. This first drying stage will not occur in AFD, because the ice cannot flow out of the dry matter. Based on this difference, some of the effects that have been observed in hot air drying (Muralidara et al., 2008) are not an issue for US-AFD.

Effects caused by cavitation will most likely only occur when water is present in a liquid form. Cavitation on solid surfaces (like ice) will normally cause damage to the surface. In some cases, this would influence the structure of the surface, but since AFD has no first drying stage it will most likely only change the product quality but not the sublimation rate. For the same reason cleaner surfaces (effect 3) will not increase sublimation rate in US-AFD. However, structural changes on the surface caused by cavitation could be one reason why the sublimation rate of pure ice showed the highest increase in mass transfer under US-AFD.

We did not observe changes in the properties of the drying agent during our tests (effect 2). The temperature and relative humidity of the drying agent showed some variation (Table 1) due to the characteristics of the HP-AFD, but there was no difference between US-AFD and AFD.

US could change the pressure gradient on the surface of the product (effect 4) which could increase sublimation rate. This is similar to effect 6, where a higher turbulence around the product will also increase mass and heat transfer.

The “sponge” effect will most likely not occur in AFD, because the product has a solid/solid matrix and the ice cannot flow out of the product. Also the effects listed under effect 7 (Oseen and Bernoulli forces) will not necessary improve the sublimation rate and are unlikely to be the cause of the faster US-AFD.

Based on these theoretical assumptions the higher sublimation rate in US-AFD is (most likely) caused by an improved heat and mass transfer between the products surface and the drying agent. First, a more turbulent flow profile might minimize the laminar boundary layer, while (second) the pressure gradient between the drying agent and the surface could be improved locally. It can be expected that improved heat transfer would also occur when US is applied in processes which are based on heat transfer by low-temperature air, such as freezing systems for e.g. food.

Future investigations should include the approach velocity as a variable, since flow profiles and laminar boundary layers affect heat and mass transfer.

The SMER values in Table 3 are based on the performance of the ultrasonic equipment and the additional sublimation rate. In our experiment, the additional sublimation did not affect the performance of the (oversized) heat pump system. Therefore the SMER values can be added to the drying system performance. In an industrial HPD, additional evaporation or sublimation will most likely also increase the conditions of the drying agent in a way that will influence the performance of the heat pump. The heat pump system must therefore be designed based on the additional sublimation and the overall performance will give the correct SMER. Based on this investigation, a higher SMER of up to 0.4 kg_{water} kWh^{-1} seems possible, when the HPD is correctly designed.

CONCLUSION

Improved heat and mass transfer caused by a more turbulent flow profile and/or locally changing pressure gradients are identified as the cause for the accelerated sublimation rate in US-AFD. With the US-system used, the SMER values were only slightly improved, but using an airborne US system with an efficiency of 80% would increase SMER values up to 0.4 for the products we tested and up to 0.8 for ice particles. Airborne ultrasound has therefore high potential for improving existing AFD systems, low-temperature driers and also food freezers.

NOMENCLATURE

AFD	Atmospheric Freeze Drying	-
CF	*Calanus finmarchicus*	-
HPD	Heat Pump Drying	-
SMER	Specific Moisture Evaporation Rate	kg_{water} kWh^{-1}
US	Ultrasound or Ultrasonic	-
VFD	Vacuum Freeze Drying	-
d.m.	dry matter	-
l	length	m

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