

## A Novel Freeze Drying Process by Using Self-Heat Recuperation Technology

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Freeze drying technology produces high quality products, because a drying condition at low temperature and pressure makes less physical and chemical changes to the dried products than any other drying technology. Therefore, this technology has been gathering attention for applying not only to foodstuffs but also to pharmaceutical products. However, it is well known that freeze drying technology consumes a lot of energy. In the process using freeze drying technology, heat is added by the heater and then removed by the condenser to prevent for vapour to enter a vacuum pump, results in wasting heat into environment. Therefore, the currently available applications are not so many. Thus, to propagate this technology to the other drying processes, it is necessary to reduce the energy required for the process and to reduce the energy costs for the production.

In this paper, we proposed three new freeze drying processes based on the concept of self-heat recuperation technology for energy saving and compared the energy required for them in a commercial process simulator. From the simulation results, it is expected that maximum 94 % of energy can be saved by using proposed processes.

### 1. Introduction

Freeze drying is one of drying technologies. The drying makes progress at low pressure and low temperature. The low drying temperature is not conducive to most degradative processes such as nonenzymatic browning, protein deterioration and enzymatic reactions (Okos et al., 2006). As a result, food products with freeze drying technology have the advantage of keeping their own taste and aroma. Simultaneously, as freezing substances can enhance the rigidity of the product, prevents collapse of the solid matrix remaining after drying. The result is a porous, nonshrunken structure in the dried product that facilitates rapid and almost complete rehydration (Liapis and Bruttini, 2006). As a result, freeze drying technology produces high quality products. Recently, the market of products by freeze drying technology grows larger and larger, because it has been utilized for not only food products but also pharmaceutical products. This technology can be used for substances, which are weak in high temperature such as low foodstuffs, biological materials, and pharmaceuticals.

Freeze drying technology has mainly three stages for drying: (1) freezing the water of product, (2) producing vacuum below triple point in a drying chamber, and (3) heating for removing the frozen water as a vapour by sublimation. Standard drying technologies use evaporation to remove water inside of drying targets, but freeze drying technology uses sublimation to remove the water. Sublimation occurs under the triple point. But if water contain some solutions, the triple point would change lower. Thus, most freeze drying is done at -10 °C or lower at absolute pressures of about 2 mmHg or less (Liapis and Bruttini, 2006).

The energy required for the above mentioned three stages are mainly accounting for the followings (1) latent heat for solidification (330 J/g-ice), cooling, (2) work for vacuum pump (ideally 820 J/L-air), (3) latent heat for sublimation (2.8 kJ/g-ice).

A conceptual flow diagram of a conventional process in the 3<sup>rd</sup> stage is shown by Figure 1. Equipment of a conventional process contains a heater, a vacuum pump, and a condenser. A heater, such as an electric heater or a boiler, is used to add heat to drying target for sublimation. A vacuum pump is used to promote

vapour to leave from a drying target. A condenser is used to remove heat for anti-sublimation (condensation) to prevent vapour from going into a vacuum pump, and condenser consists of a refrigeration cycle. Then, heat is wasted through the environment by cooling water of a refrigeration cycle. Thus, an energy consumption of freeze drying technology is so large. As a result, freeze drying technology has been industrially applied to only producing high added value products.

Huang et al. (2009) reported that a reason for large energy consumption is slow drying rate because of slow heat and mass transfer. Actually, the drying time of freeze drying is three times more than that of tunnel drying (Reyes et al., 2015). There are some researches to decrease drying rate. Chen et al. (2016) reported that by using ultrasound in freezing to obtain bigger crystals of ice. The drying time with ultrasound in freezing was reduced by 21.8 %. Also, Parniakov et al. (2016) reported that the drying rate could be reduced by using pulsed electric field in freezing a target, 1st stage. Huang et al. (2009) reported that by using microwave and heat is transferred by radiation, the rate of heat transfer was increased and drying time was decreased. As a result, more than 50 % of its energy consumption could be reduced. However, if the drying time is reduced, energy consumption should be same under ideal condition. To decrease energy consumption with decreasing the drying time corresponds to reduce mechanical loss. It is necessary to reduce and recuperate the waste of heat.

In this research, we focused on reducing waste of heat and proposed three new freeze drying processes with the concept of reducing a waste of heat by introducing the concept of SHR (Self-Heat Recuperation) technology to freeze drying process, leading to the minimization of the exergy loss for heat transfer.

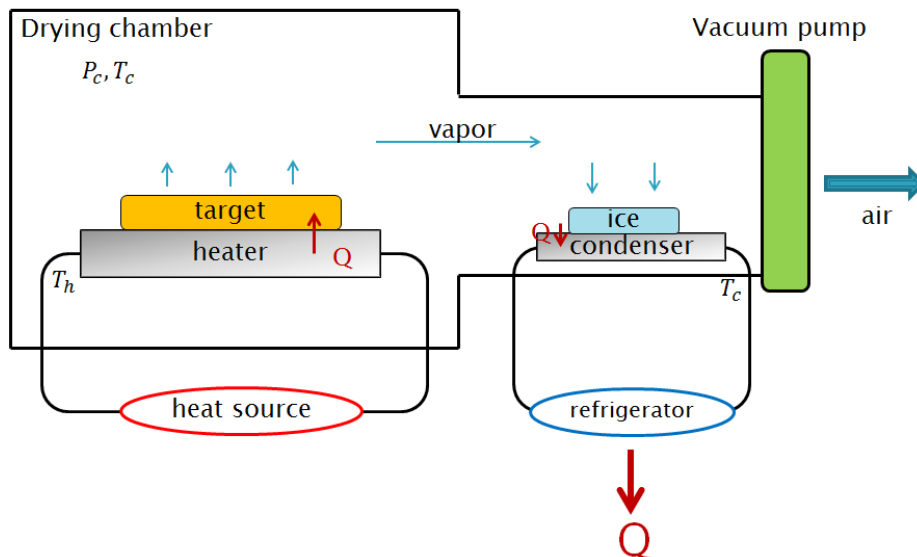


Figure 1: Energy flow diagram of a conventional process

## 2. Concepts of proposed processes

### 2.1 New process (A)

Figure 2 shows a proposed process (A). The concept of new process (A) is that waste heat is reduced and recuperated using heat pump from a condenser to a heater.

In this process, heat for sublimation added by a heater is the same as heat for anti-sublimation (condensation) removed by condenser. Thus, it is possible to introduce perfect internal heat circulation that heat for sublimation is provided through heat pump. The heat source was latent heat for sublimation by condenser. If waste heat from condenser can be recycled for heat of a heater through a heat pump, waste of heat would be decreased. As a result, it is expected that the energy consumption of freeze drying technology could be saved. However, a larger temperature difference between sublimation and condensation requires, more work of heat pump requires.

The work required for this process is work for a compressor of a heat pump. To balance energy flow, cooler is used. Then, waste heat of this process is amount of work for a compressor.

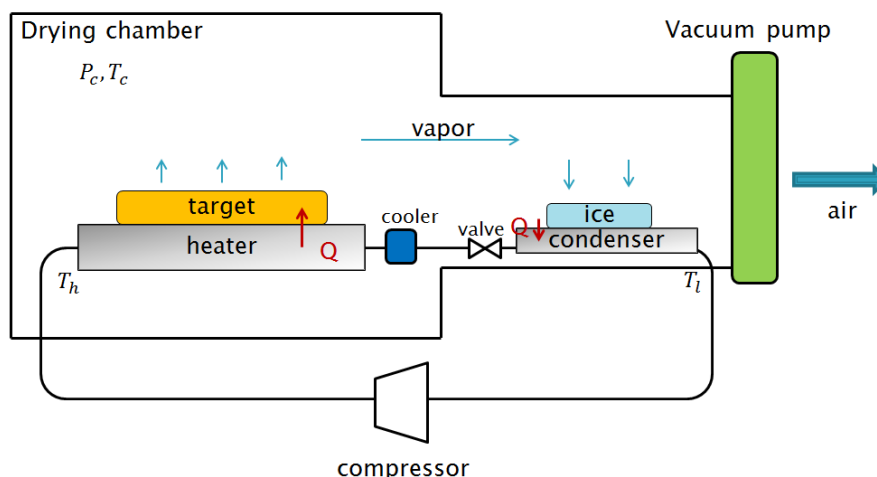


Figure 2: Energy flow diagram of new process (A): heat for sublimation is provided through heat pump

### 2.2 New process (B)

Figure 3 shows new process (B). In this process, a drying chamber is separated into two chambers. These two chambers are connected through a compressor. A compressor makes a difference in pressure between two chambers. Then, pressure of one part of chambers is lower and pressure of the other part is higher. Both pressures in the chambers are adjusted for the change of sublimation and condensation occurring at the same temperature by compressor. Therefore, heat is carried from a condenser to a heater through heat medium.

To balance the amount of heat, cooling water is put next to compressor to waste heat corresponding to the work for compressor. Assuming that phase changes would occur inside a heater and a condenser, temperatures of a heater and a condenser are the same. Then, heat medium keeps the constant temperature through the heat medium circulation. In this case, work for a fan is almost zero.

In this new process (B), the energy consumption is only work for a compressor making a difference in pressure of two chambers.

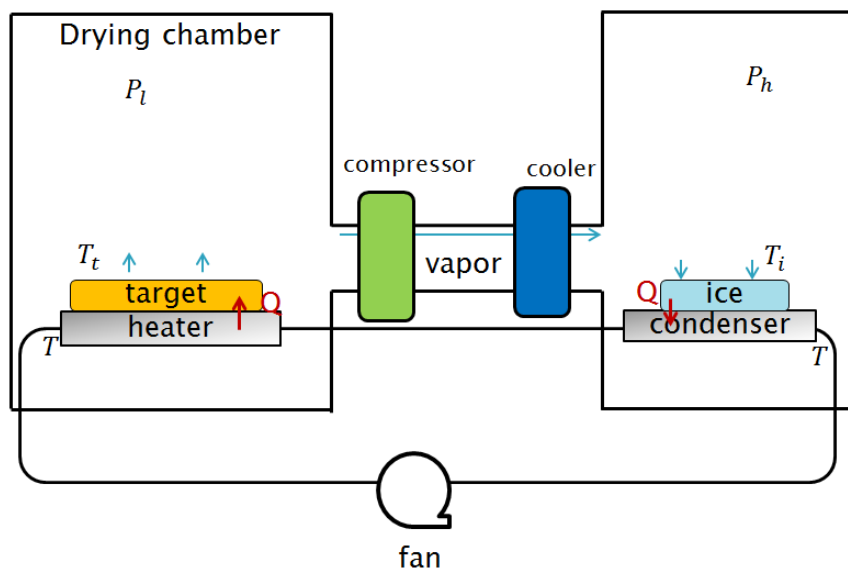


Figure 3: Energy flow diagram of new process (B): difference is pressure changes the point of sublimation

### 2.3 New process (C)

Figure 4 shows last new process. Its concept is the same as new process (B). By making a difference in pressure, sublimation point would change. Sublimation and anti-sublimation (condensation) occur at the same temperature. Then, the temperature of a target in chamber with low pressure is lower than the temperature of

ice in chamber with high pressure. New process (C) does not have heat medium through heater and condenser, and two plates for heater and condenser are connected each other. Then, heat is directly transferred from a condenser to a heater by conductive. In this way, heat is circulated through the vapour from drying target. Without heat medium, it would be expected to have a less resistance of heat transfer than that of new process (B).

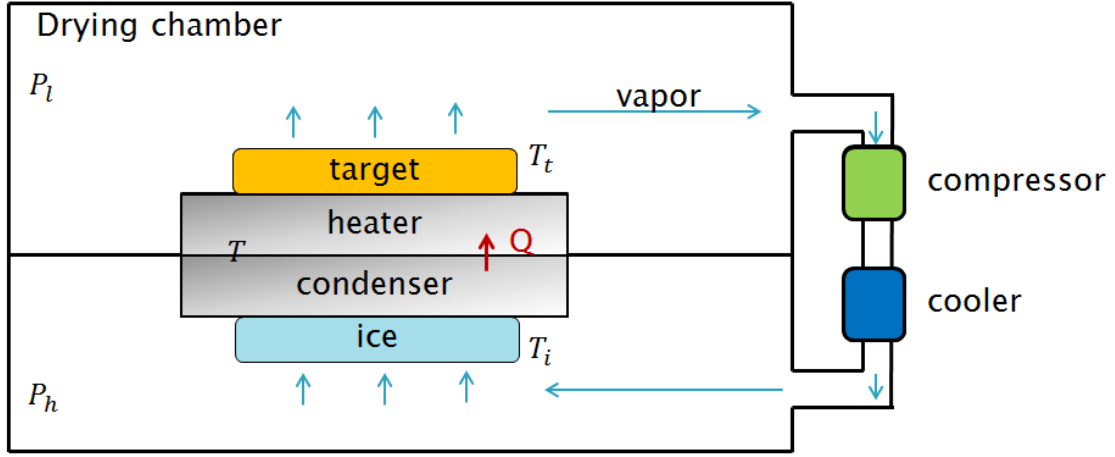


Figure 4: Energy flow diagram of new process (C): vapour takes charge of a heat carrier

### 3. Simulations

In these simulations, the following assumptions were defined;

In a conventional process and new process (A), pressure of drying chamber ( $P_c$ ) was 20 Pa, temperature of sublimation point was 243 K equal to temperature of drying chamber ( $T_c$ ). Temperature of heater ( $T_h$ ) was 313 K, and temperature of condenser ( $T_i$ ) was 228 K. In new process (B) and new process (C), pressure of drying chamber ( $P_l$ ) was 20 Pa. Pressure for condensation ( $P_h$ ) was 100 Pa. At 20 Pa, the point of sublimation was 243 K, corresponding to the temperature of target ( $T_t$ ). Also at 100 Pa, the point of sublimation was 263 K, corresponding to the temperature of target ( $T_t$ ).

To evaluate the energy consumption for compressors of new process (B) and (C), we conducted simulations by using a commercial process simulator (PRO/II Ver. 9.1, Invensys). The Soave-Redlich-Kwong equation of state was used. The adiabatic efficiency of the compressor was 100 %. Finally,  $\text{CO}_2$  was assumed to be the working fluid in the refrigerator and the heat pump.

And a coefficient of performance (COP) was calculated from Eq(1)

$$COP = \frac{Q}{W} \quad (1)$$

$W$  was required energy for a compressor.  $Q$  was amount of transferred heat.

In a conventional process, latent heat for sublimation was 2.8 MJ/kg-ice at 20 Pa, supplied by a heater. The condensation heat at the condenser was also 2.8 MJ/kg-ice, removed by the refrigeration cycle. The required energy for condenser, which transferred heat from 228 K to ambient temperature (298 K), was 0.6 MJ/kg-ice by the simulator. Then, COP of this refrigerator was 4.6 from Eq(1). From these evaluations, total required energy of conventional process was 3.4 MJ/kg-ice.

In new process (A), latent heat for sublimation was also 2.8 MJ/kg-ice at 20 Pa, supplied through heat pump from condensation heat. Therefore, the required work of the heat pump, which transferred heat from 228 K to 313 K, was 0.7 MJ/kg-ice by the simulator. Then, COP of this heat pump was 4.1 from Eq(1). In new process (B), temperature of a heater and a condenser and heat medium ( $T$ ) was 243 K. Under this condition, the work of the compressor to compress water vapour from 20 Pa to 100 Pa was 0.2 MJ/kg-ice. In new process (C), temperature of a condenser and a heater ( $T$ ), was 243 K. Then, work of a compressor to compress water vapour from 20 Pa to 100 Pa was 0.2 MJ/kg-ice.

The energy required for each process is summarized in Table 1. It can be seen from this table that maximum 94 % of energy is saved by new process (B) and new process (C) as compared with conventional process.

Table 1: Energy consumptions for conventional process and three new processes

Process name	Conventional process	New process (A)	New process (B)	New process (C)
Energy consumption	3.4 MJ/kg-ice	0.7 MJ/kg-ice	0.2 MJ/kg-ice	0.2 MJ/kg-ice
Ratio of conventional process	100 %	20 %	6 %	6 %

#### 4. Discussion

In these simulations, we assumed the steady-state conditions. To realize the above-mentioned process, we must consider the dynamic systems. The most important factors for considering dynamics are a heat transfer rate and a mass transfer rate.

In new process (A), temperatures and pressures in drying chamber are the same as those of a conventional process. In addition, a difference is only a part of heat medium. Therefore, it is conceivable that new process (A) has feasibility.

However, new process (B) and (C) is far different from conventional process. New process (B) and (C) use a compressor at path connecting two chambers in high vacuum condition. Depend on this connecting pipe, there is some possibility that molecular motion of vapour could not move as viscous flow but molecular flow. In pressure of molecular flow region, some molecular would have back stream that counter flow from a pump to a chamber. Then, required work for compressor could become larger because it is hard to move molecular flow.

Thus, it should be checked whether flow of vapour is molecular flow or not. And to see the effect of the mass transfer rate, we conducted the following calculations.

A mean free path is calculated by Eq(2).

$$\lambda = \frac{kT}{4\sqrt{2}p\pi r^2} \quad (2)$$

Assuming that the part of chamber with low pressure.  $k$  is Boltzmann constant ( $1.38 \times 10^{-23}$  J/K<sup>-1</sup>).  $T$  is the temperature of drying chamber (233 K).  $r$  is the radius of water molecular ( $\sim 3 \times 10^{-10}$  m).

By substituting these parameters for Eq(2), a mean free path is  $1.0 \times 10^{-4}$  m.

Next, this result is substituted for Knudsen number ( $K_n$ ). Knudsen number is following:

$$K_n = \frac{\lambda}{D} \quad (3)$$

$D$  is a diameter of a pipe. Knudsen number condition for viscous flow is  $K_n < 10^{-2}$ . From Eq(2) and Eq(3), The condition of a diameter of a pipe is  $D > 10^{-2}$  m at 20 Pa, and  $D > 2 \times 10^{-3}$  m at 100 Pa.

Also, Knudsen number condition for molecular flow is  $K_n > 1$ . From Eq(2) and Eq(3), The condition of a diameter of a pipe is  $D < 10^{-4}$  m at 20 Pa, and  $D < 2 \times 10^{-5}$  m at 100 Pa. We do not suppose molecular flow region.

Also, Eq(4) shows mass flow in viscous flow region.

$$Q = \frac{\pi a^4 p}{8\eta L} \frac{p_1 - p_2}{L} \quad (4)$$

$Q$  is mass flow.  $a$  is a radius of a pipe.  $P$  is an average of  $P_1$  and  $P_2$ .  $P_1$  and  $P_2$  are pressure of the each end of a pipe.  $\eta$  is a coefficient of viscosity.  $L$  is a length of a pipe.

From Eq(4), we could estimate a distance that vapour can move. Then, an area limit of heat transfer could be estimated. Finally, we could fix a difference in temperature of heat transfer. If a difference in temperature of heat transfer, we could estimate an accurate energy consumption of new drying process.

Furthermore, it is necessary to understand the heat transfer rate. New process (B) and (C) has problem about a rate of heat transfer. Because pressure where sublimation occurs is under the triple point, a difference in temperature would be small. If a difference in temperature is small, its heat transfer rate could be also small. Moreover, in a part of heater, heat is transferred by conduction. Thus, it is expected that a heat transfer rate is not too small. However, a part of condenser, heat is transferred by convection under low pressure. Thus, a rate of heat transfer could be too small. Then, a rate of drying would be small, and it takes too long time to drying a target.

We would conduct some experiments to grasp behaviour of vapour and a heat transfer rate under low pressure and temperature.

## 5. Conclusion

In this paper, we proposed three new processes with concept that waste of heat is reduced and recuperated. New process (A) reduce maximum 80 % of an energy consumption of conventional process, also new processes (B) and (C) reduce maximum 94 % of that.

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