

## Characteristics of Vacuum Freeze Drying with Utilization of Internal Cooling and Condenser Waste Heat for Sublimation

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### Abstract

Vacuum freeze drying is an excellent drying method, but it is very energy-intensive because a relatively long drying time is required. This research investigates the utilization of condenser waste heat for sublimation as a way of accelerating the drying rate. In addition, it also investigates the effect of internal cooling combined with vacuum cooling in the pressure reduction process. Jelly fish tentacles were used as the specimen, with different configurations for condenser heat waste and internal cooling valve opening. The results show that heating with condenser heat waste can accelerate the drying rate up to 0.0035 kg/m<sup>2</sup>.s. In addition, pre-freezing by internal cooling prevents evaporation until the mass of the specimen is 0.47 g and promotes transition of the specimen into the solid phase.

### Abstrak

**Karakteristik Pengering Beku Vakum dengan Menggunakan Pendinginan Internal dan Panas Buang Kondenser untuk Sublimasi.** Pengering beku vakum adalah metode pengeringan yang terbaik, tetapi konsumsi energinya sangat tinggi karena lamanya waktu pengeringan yang diperlukan. Penelitian ini mengkaji penggunaan panas buang kondenser untuk proses sublimasi sebagai salah satu cara untuk mempercepat laju pengeringan. Disamping itu, juga mengkaji pengaruh pendinginan internal yang dikombinasikan dengan pendinginan vakum pada waktu dilakukan proses penurunan tekanan. Tentakel ubur-ubur (*Jelly fish tentacles*) digunakan sebagai bahan percobaan dengan berbagai konfigurasi panas buang kondensator dan pembukaan katup pendinginan internal. Hasilnya menunjukkan bahwa pemanasan dengan panas buang kondensator dapat mempercepat laju pengeringan sampai dengan 0.0035 kg/m<sup>2</sup>.s Selain itu, pembekuan awal dengan melakukan pendinginan internal dapat mencegah terjadinya penguapan sampai dengan 0.47g dan mempercepat transformasinya menjadi padatan.

*Keywords: condenser waste heat, drying rate, internal cooling, sublimation, vacuum freeze drying*

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### 1. Introduction

Cancer is a disease characterized by rampant proliferation of cells [1]. Normal cells follow a natural pattern of growth, reproduction, and death. The process of cell death is called apoptosis; if apoptosis does not occur, cancer may develop. Apoptosis is not necessarily a result of DNA damage or mutations [2]. Unlike normal cells, cancer cells do not undergo apoptosis, and they continue to grow and divide irrepressibly. There are more than 100 types of cancers, and each is

classified by the type of cells involved, for example, breast cancer, mouth cancer, and brain cancer.

Research has shown that cancer is the seventh primary cause of death in the world after stroke, tuberculosis, hypertension, prenatal trauma, and diabetes mellitus [3]. In Indonesia, cancer cases have been predicted to increase by up to seven times the current figure by 2030 [4]. Unfortunately, detection of this disease is difficult, so it is often too late to make use of the available advanced forms of treatment. Even X-ray imaging does

not always indicate this disease because of limitations in terms of bone penetration.

Fortunately, the rapid growth of technology has helped scientists conduct research and find a method to identify infected cells. This method uses the green fluorescent protein (GFP) from jellyfish (*Scyphomedusae*) to detect self-repairing cells. This technology was developed from previous research on the isolation of jellyfish GFP [5]. Scientists have used genetic engineering to infuse an infected cell with GFP. Although it could not initially be identified due to its small size, as it grew and spread, the protein became visible, and the glow was detected with the aid of a special camera.

To support this technology, scientists need prime-quality jellyfish protein. Jellyfish tentacles have a lot of nematocysts and are rich in protein as well as DNA [6]. Until now, the extraction process for basic medicinal material required evaporation. This method is not effective because water extracted from the specimen, as well as the entire specimen itself, may evaporate.

The most efficient and effective drying method is vacuum freeze drying, a process in which a specimen is exposed to vacuum to reduce the pressure to below the triple point of water (pressure, 6.1 mbar; temperature, 0 °C) so that ice crystals are created [7-8]. But this method has a weakness, i.e., the product nutrients could evaporate due to decrease in the boiling point [9]. Then, to accelerate the drying rate and reduce the drying time, additional heat is added to sublimate the ice directly into the gas phase [10-11]. The heat source could be an electrical heater, microwave or waste heat condenser [10-13]. This method preserves the nutrition content, color, taste, and biological structure of a specimen. The disadvantage of electrical and microwave heating is the intensive amount of energy required to fuel the three processes: cooling, sublimation, and water vapor trapping.

This research presents innovations to the vacuum freeze drying apparatus: the combination of a vacuum pump and refrigeration system (internal cooling process) to reduce the temperature and pressure in the specimen chamber. As part of this research, condenser waste heat was used for heating and accelerating drying time.

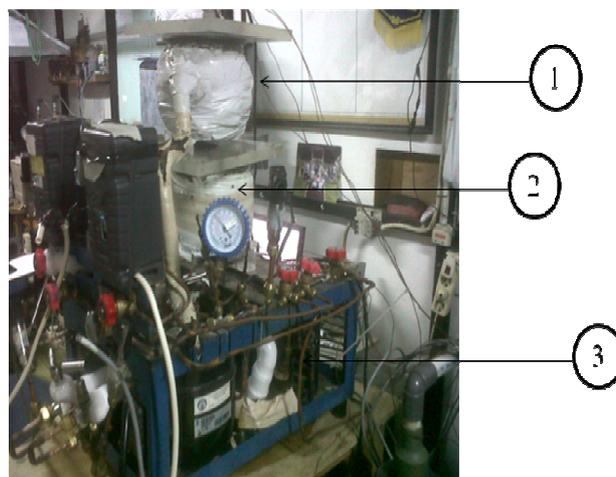
## 2. Methods

The innovation in the compact vacuum freeze drying machine is the utilization of internal cooling (using the refrigeration system) to fix the pre-freezing evaporation problem. In addition, condenser waste heat is used to address the energy problems in the sublimation process.

Research was conducted using a compact vacuum freeze drying machine (Figure 1) that is located in the Refrigeration and Air Conditioning Laboratory of the

Mechanical Engineering Department at Universitas Indonesia. This machine consists of two main systems: a vacuum system and a refrigeration system. The evaporator consists of two chambers, a drying chamber and a coldtrap. The vacuum system was used to reduce pressure in the drying chamber until it fell to below the triple point of water; this process is called vacuum cooling [9]. Simultaneously, the refrigeration system was used to reduce the temperature in the evaporator. The coldtrap chamber was used to capture the moisture from the dried specimen so that it did not flow into the vacuum pump. The instrument used to measure the vacuum pressure in the drying chamber is the Druck PTX 1400 pressure transmitter, which has a working range of 0–1600 mbar. The temperatures were measured with “k” type thermocouples which have a reading accuracy of  $\pm 0.14\%$ . The analytical balance product from AND Weighing type EK 6100i was used to weigh the initial and final specimens; the weighing capacity is 6000 g with increments of 0.1 g. During the experimental process, all data were recorded using National Instruments Data Acquisition type 9211 and 9203, at one-minute intervals. Thermocouple uncertainty ranged from 0.2% to 0.7%; the pressure transmitter uncertainty ranged from 0.1% to 0.3%; and the analytical balance uncertainty was 0.04%.

The schematic for this device is shown in Figure 2. The refrigeration scheme used in this machine was the cascade system, with a plate heat exchanger (PHE) as the “connection” between two refrigeration circuits. The higher temperature circuit (HTC) uses 450 g of R22 as the refrigerant, while the lower temperature circuit (LTC) uses 250 g of propane as the refrigerant. For internal cooling, the valve that connects with the drying chamber must be opened; to utilize condenser waste heat, the bypass valve from the compressor to the drying chamber must be opened.



**Figure 1. Compact Vacuum Freeze Drying Machine: (1) Drying Chamber, (2) Coldtrap, (3) Refrigeration System**

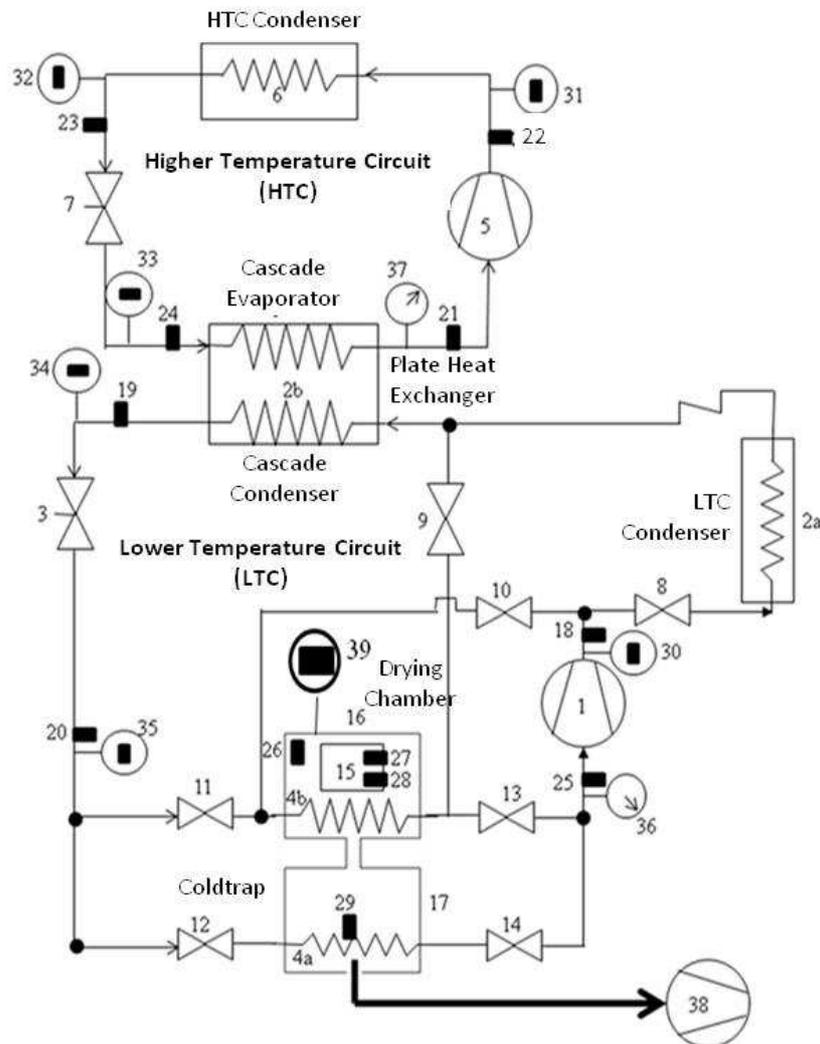


Figure 2. Experimental Schematic Diagram

**Note:**

**Main Equipment:** 1. Compressor LTC; 2a. Condenser LTC; 2b. Plate heat exchanger (PHE); 3. Expansion valve LTC; 4a. Evaporator at coldtrap; 4b. Evaporator at drying chamber; 5. Compressor HTC; 6. Condenser HTC; 7. Expansion valve HTC; 15. Tray; 16. Drying chamber; 17. Coldtrap.

**Valves:** 8. Inlet condenser LTC; 9. Inlet cascade condenser; 10. Condenser heat waste; 11. Inlet drying chamber; 12. Inlet coldtrap; 13. Outlet drying chamber; 14. Outlet coldtrap.

**Thermocouples:** 18. Outlet compressor LTC; 19. Outlet cascade condenser; 20. Inlet evaporator LTC; 21. Inlet compressor HTC; 22. Inlet condenser HTC; 23. Outlet condenser HTC; 24. Outlet expansion HTC; 25. Inlet compressor LTC; 26. Drying chamber; 27. Top position; 28. Bottom position; 29. Coldtrap.

**Pressure Transmitters:** 30. Discharge LTC; 31. Discharge HTC; 32. Outlet condenser HTC; 33. Inlet evaporator cascade; 34. Outlet cascade condenser; 35. Inlet evaporator LTC.

The specimen used in this research was tentacles of the jellyfish; and they were blended into samples weighing as much as 50 g with 95.6% moisture content (the moisture content test was conducted by the Research Center for Biological and Biotechnology Resources Bogor Agricultural Institute). The specimen was placed in small plastic containers and subsequently refrigerated. Fresh tentacles of the jelly fish weighing 50 g (moisture content, 95.6%) were stirred and placed in a teflon tray. The cascade refrigeration system was

then turned “on” at HTC, until the PHE temperature was below  $-30\text{ }^{\circ}\text{C}$  and the coldtrap temperature was approximately  $-40\text{ }^{\circ}\text{C}$ . After that, the evaporator valve at the drying chamber was opened until the specimen freezing temperature was below  $0\text{ }^{\circ}\text{C}$ . Valve opening can be varied with the experimental parameters. The next step was starting the vacuum pump until the end of the drying process. When specimen temperature was steady and reached the sublimation temperature (approximately  $-5\text{ }^{\circ}\text{C}$ ), the refrigerant condenser valve

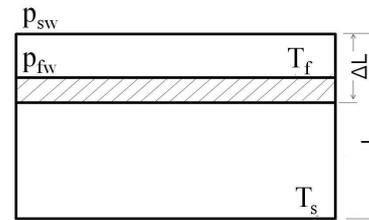
was opened in order to utilize the condenser’s heat for sublimation, and then the specimen was weighed. The two variables in this research were the temperature during internal cooling and the heating temperature while using the condenser waste heat. Internal cooling was used during the vacuum cooling process, and the heater was used in the sublimation process. To vary the temperature, we adjusted the valve opening. The testing variations are shown in Table 1.

Liquid material (fresh tentacle) of length L was placed in a tray which was kept inside the drying chamber for freezing by internal cooling; it underwent a phase change and became solid (ice). Due to the heating process afterward, the pressure inside the drying chamber was lower than the moisture pressure saturation of the material, and again the material changed from solid (ice) to gas. Due to this phenomenon, the thickness of the material decreased, starting from the upper side, until the thickness was ΔL (thin layer). During the drying process, ΔL was increased while L was decreased, and when the experiment was completed, the only material remaining in the tray was dry material.

In vacuum freeze drying, two heat transfer phenomena occur: conduction and radiation. The mathematical equation for heat transfer via conduction is shown in Eq. (1), and the schematic is shown in Figure 3.

**Table 1. Valve Opening Variation during Experiments**

No	Variation Function	Valve					Additional Treatment
		11	13	10	8	9	
1	Internal cooling	Off	Close	Close	Close	Open	Close
	Heating	Off	Close	Close	Close	Open	Close
2	Internal cooling	On	Open	Open	Close	Open	Close
	Heating	On	Close	Close	Open	Close	Open
3	Internal cooling	On	Half	Half	Close	Open	Close
	Heating	On	Close	Close	Half	Close	Half
4	Internal cooling	Off	Close	Close	Close	Open	Close
	Heating	On	Close	Close	Open	Close	Open
5	Internal cooling	On	Open	Open	Close	Open	Close
	Heating	Off	Open	Open	Close	Open	Close



**Figure 3. Mathematical Notation for Heat Transfer**

The heat flux  $q$  ( $W/m^2$ ) corresponds to ice thermal conductivity ( $\lambda$ ,  $W/m.K$ ), temperature of the surroundings ( $T_s$ ,  $K$ ), freezing front ( $T_f$ ,  $K$ ) and liquid thickness ( $L$ ,  $\Delta L$ ,  $m$ )

$$q = \frac{\lambda}{L - \Delta L} (T_s - T_f) \tag{1}$$

$$\Delta L = (1 - x)L \tag{2}$$

The drying rate due to sublimation ( $m$ ,  $kg/m^2.s$ ) is formulated as shown in Eq.(3), as the ratio of heat flux and latent heat of sublimation ( $h$ ,  $J/kg$ ).

$$\dot{m} = \frac{q}{h} \tag{3}$$

$$X_t = \frac{(m_d + m_w) - m_t}{m_d + m_w} \tag{4}$$

The moisture content of the final product is shown in Eq.(4).  $m_d$ ,  $m_w$  and  $m_t$  represent the mass of dry material (g), mass of water (g) and mass of material after time  $t$  (g), respectively.

Some assumptions are used. They are as follows [10]: 1) Heat transfer and mass transfer occur in one dimension only, which is normal to the sublimation interface and surface; 2) Sublimation occurs at an interface parallel to and at distance  $X$  from the surface; 3) The thickness of the sublimation border is infinitesimal; 4) The frozen region remains at a temperature equal to the interface temperature; 5) Only water vapor flows through the dried layer; 6) In porous regions, the solid matrix and the trapped water vapor are in thermal equilibrium; 7) The sides and bottom of the specimen container are assumed to be perfectly isolated.

## 4. Results and Discussion

**Specimen Phase Change.** The change in the specimen phase can be seen in Figures 4–8. Testing performed using certain variations revealed that the specimen evaporated before entering the solid state because the reduced pressure caused the water to reach its boiling point [15]. Unfortunately, some specimens either evaporated or were sucked into the vacuum pump.

The material in the liquid state was inserted into the chamber. At variation 1, the valve configurations were as shown in Table 1; when the vacuum pump was started, the drying chamber pressure was decreased. This phenomenon causes a decrease in the moisture boiling point, and the (latent) heat transferred causes a decrease in the temperature of the material until the solid state is reached (Figure 4). With variation 2, shown in Figure 5, the material in the liquid phase was placed in the drying chamber, and the temperature inside the chamber was reduced until the solid state was reached. Figure 5 shows the path at almost the same pressure. Then, the vacuum pump was turned on until the pressure inside the drying chamber reached below the triple point condition (less than 6.1 mbar).

For variation 3 (Figure 6) and variation 4 (Figure 7), internal cooling and the vacuum pump were turned on at the same time. The pressure and temperature of the drying chamber were decreased. The evaporation effect still occurs in variations 1, 3 and 4. For variation 2, the effect of evaporation was not evident because the pressure was decreased due to turning on of the vacuum pump after the material turned solid. For variation 5, shown in Figure 8, the temperature inside the drying chamber (internal cooling) was decreased until the end of the process, and the drying chamber pressure was

also reduced. The drying process did not take place on account of the saturation pressure of the material being lower than the drying chamber pressure (Figure 8).

**Specimen saturation pressure.** For mass transfer to occur, a difference must exist between the saturation pressure of the water in the specimen layer and the vacuum pressure [16-18]. Figures 9–13 illustrate that after a certain amount of time, the difference between the saturation pressure and the vacuum pressure begin to emerge, indicating that mass transfer occurred in all specimens, except in the case of the 5th testing variation. There is also the possibility that mass transfer occurs from the drying chamber to the specimen, but this condition can be ignored due to the vacuum pressure

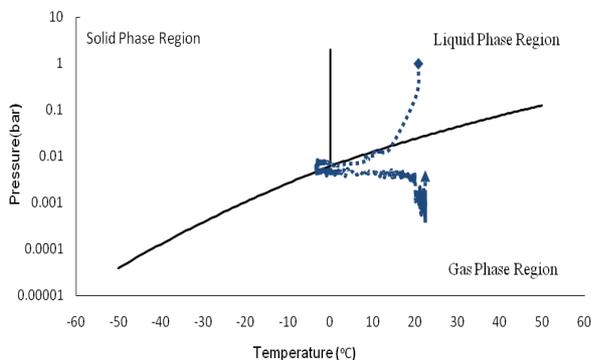


Figure 4. Pressure and Temperature diagram Variation 1

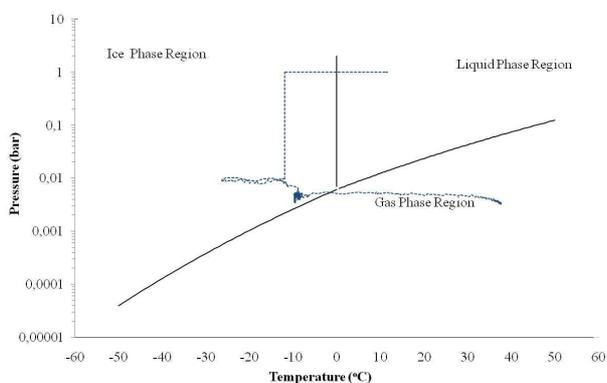


Figure 5. Pressure and Temperature diagram Variation 2

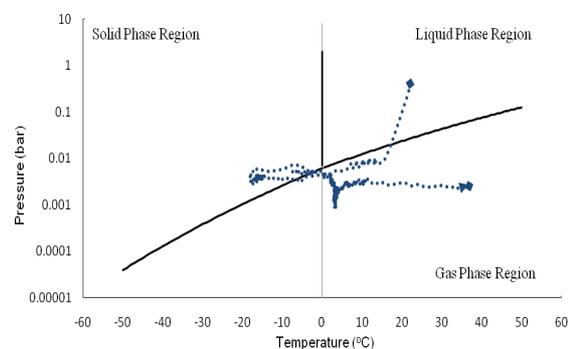


Figure 6. Pressure and Temperature diagram Variation 3

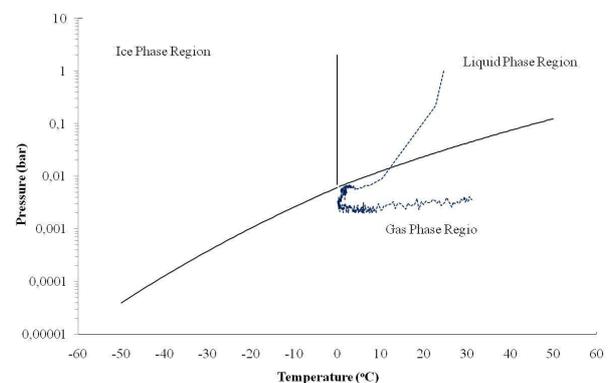


Figure 7. Pressure and Temperature diagram Variation 4

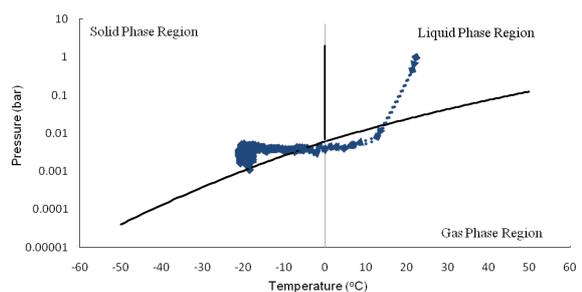


Figure 8. Pressure and Temperature diagram Variation 5

inside the drying chamber. Sublimation occurs when the drying process condition is below the triple point condition (temperatur below 0° and pressure below 6.1 mbar). This is the advantage of vacuum freeze drying compared to other drying process, that the content of the material is unaffected [15].

**Specimen Temperature History.** The temperature history shown in Figure 14 is similar to the temperature history for the freeze-drying process: the three zones are freezing, primary drying, and secondary drying. In this figure, it can be seen that every variation has a resulting drying time. In Figure 15, the 2<sup>nd</sup> trial variation is used for comparison. The 2<sup>nd</sup> and 3<sup>rd</sup> trial variations did not have distinguishable borders between zones because internal cooling was not used at the beginning of the process; therefore, the temperature of the specimen appears to be the same as the temperature during primary drying [11,19].

**Drying rate.** The drying rate due to conduction is calculated in Eq. (3); q is obtained from Eq. (1). The thickness of the material (L), thermal conductivity ( $\lambda$ ),

and latent heat of sublimation of ice ( $\Delta h$ ) are 0.008 m, 2.24 W/mK, and  $2.83 \times 10^6$  W/mK, respectively. Figure 16 shows the drying rate for each testing variation.

The graph showing the drying rate due to conduction (Figure 16) appears to be similar to the graph for the common drying rate of the freeze-drying process. The

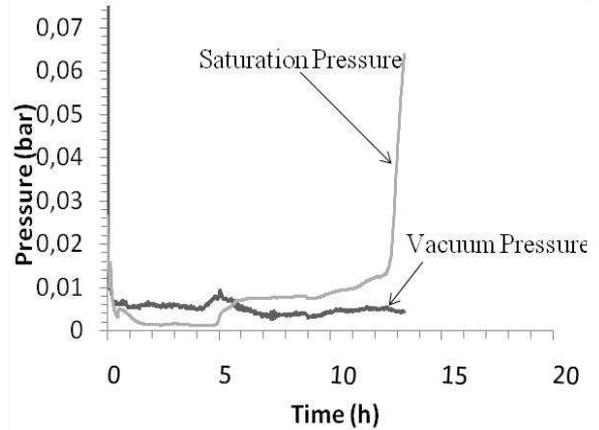


Figure 11. Saturation and Vacuum Pressure at Variation 3

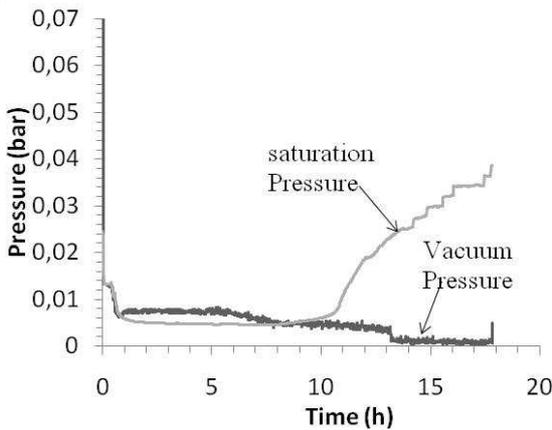


Figure 9. Saturation and Vacuum Pressure at Variation 1

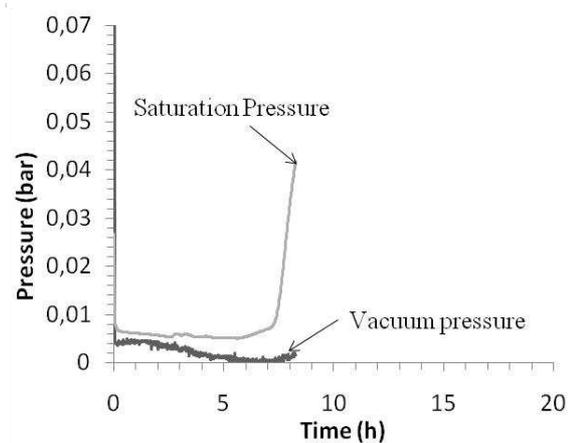


Figure 12. Saturation and Vacuum Pressure at Variation 4

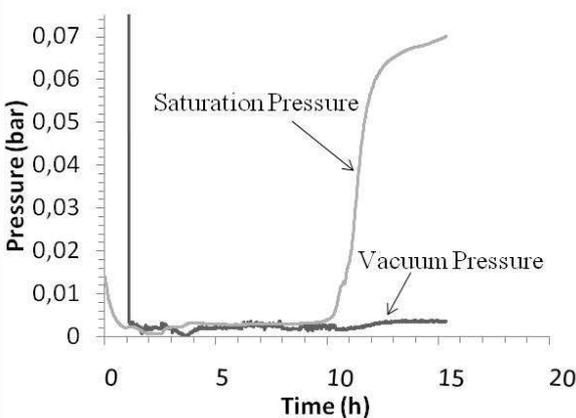


Figure 10. Saturation and Vacuum Pressure at Variation 2

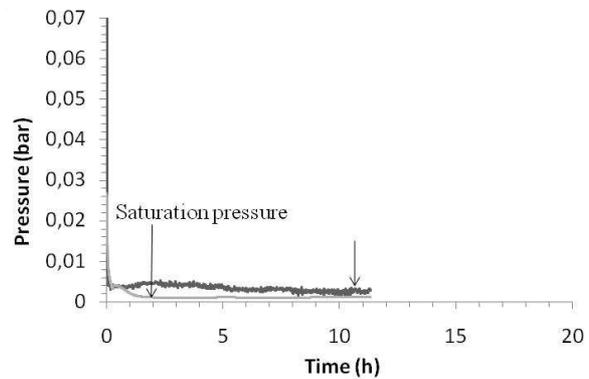


Figure 13. Saturation and Vacuum Pressure at Variation 5

graph consists of two main zones: the constant drying rate and the falling rate [20].

**Moisture content and drying time.** The final product mass can be seen in Table 2: the initial weight of all the specimens was 50 g for all variations. The final product has a smaller mass than expected for the final product (2.2 g) due to excessive losses during the drying process. Several reasons could explain the loss. First, there is the possibility that some parts of the specimen were sucked into the vacuum pump due to evaporation at the beginning of the process. Second, the dried product could have stuck to the gauze used to cover the pan container to prevent the specimen from being

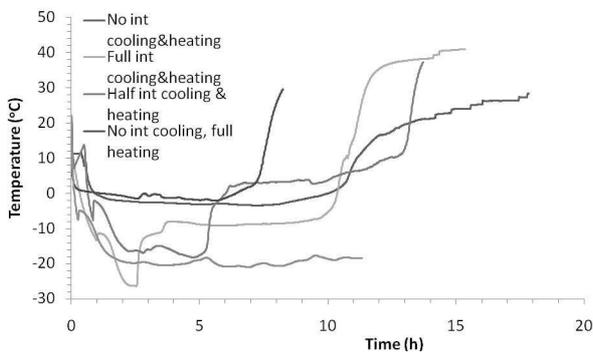


Figure 14. Profile of Material Temperature During the Drying Process

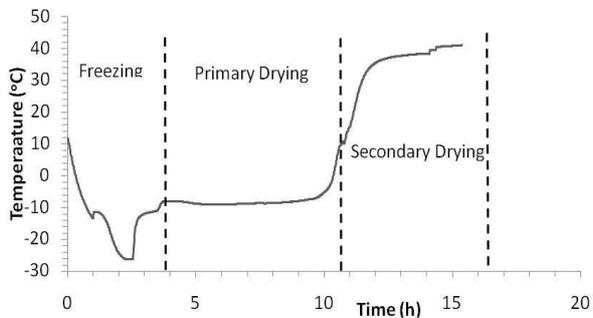


Figure 15. Temperature Region during the Drying Process

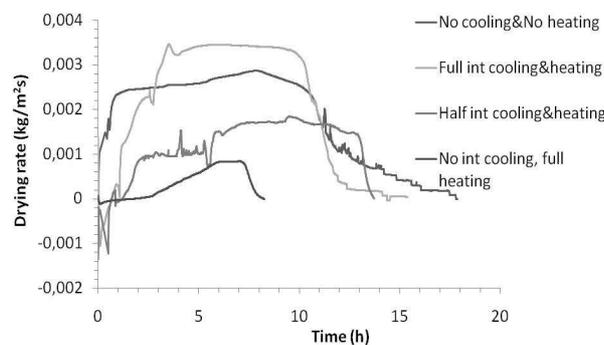


Figure 16. Drying Rate based on the Conduction Equation

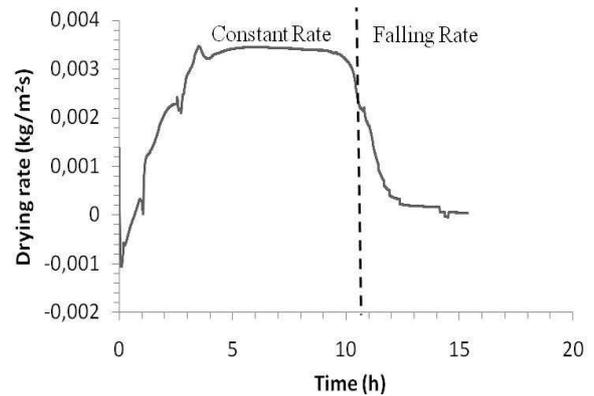


Figure 17. Drying rate based on the Constant and Falling Rate Periods

Table 2. Specimen Mass Loss

Variation Number	Final mass (g)	Moisture Evaporated	
		During drying (g)	During evaporation (g)
1	0.78	49.22	1.42
2	1.73	48.27	0.47
3	1.66	48.34	0.54
4	0.55	49.45	1.65
5	48.5	-	-



Figure 18. Final Product: Dried Jelly Fish

sucked into the vacuum pump. Third, the collection method used to obtain the final product may have had some drawbacks. From Table 2, it can be seen that the 2<sup>nd</sup> trial variation has a greater final product mass than other trial variations because the mass of the product did not evaporate due to pre-freezing. The dried specimens have the same structure as seen visually (Figure 18).

#### 4. Conclusions

After conducting the research and analyzing the acquired data, there are several conclusions that can be drawn. The vacuum freeze-drying rate could be modeled with the conduction equation. Internal cooling utilized during the pre-freezing process has an advantage with regard to avoiding early specimen evaporation in the drying process. Valve arrangements could be used to extract condenser waste heat and to accelerate the drying rate. The saturation pressure in the specimen layer must be greater than the vacuum pressure for sublimation to take place. The temperature history curve shows three zones in the drying process: freezing, primary drying, and secondary drying. The drying rate curve has two main zones: the constant rate and falling rate. The fastest experimental variation is the 4<sup>th</sup> variation (no internal cooling, full heater).

#### Acknowledgement

The authors acknowledged financial support from DRPM UI (Universitas Indonesia Research Centre) Contract: DRPM /RSNUI/2011/1/10721 and Directorate of Higher Education (DIKTI) contract no : 3398/H2.R12/HKP.05.00/2012.

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