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Basic Study on the Heat Transfer during Rapid Freezing of Soybean Seed by Liquid Nitrogen

Toshio KAWANO, Kohei NAKANO and Satoshi MURATA

Agricultural Process Engineering Labolatory
Department of Agricultural Engineering
Faculty of Agriculure
Kyushu University
6-10-1, Hakozaki, Higashi-ku, Fukuoka, 812 Japan

ABSTRACT

Direct freezing tests of soybean seed by liquid nitrogen were carried out at various moisture contents and the following important conclusions were drawn from the results of temperature measurements of soybean seed and photographs of bubbles generated on its surface:

1)Assuming that the temperature gradient in a soybean seed is negligible because of its small seed size and the freezing ratio is followed the Heiss's formula, and a differential equation based on the heat energy balance was introduced. The equation was easily solved by the Runge-Kutta-Gill method and the predicted values of the temperature were in good agreement with the observed data.

2)The photographs of bubble generation during freezing showed the boiling mode was nucleate, and then the most suitable formula on the nucleate boiling heat transfer was introduced from many formulae proposed up to now by fitting the calculated values bas-ed on the formula to the observed data.

The formula used for the prediction of the seed temperature was as follows:

$$\frac{\partial T_S}{\partial \theta} = - \frac{\alpha (T_S - T_L)^{3}\ ^{3}}{W\left(C_S - \frac{\delta m(CT_S + \sigma)}{T_S{}^{2}}\right)}$$

where C = difference of the specific heat between pure ice and water
m = moisture content of soybean seed
T_S = seed temperature
T_L = temperature of liquid nitrogen
W = mass of soybean seed
α = proportional constant
δ = constant depends on variety or the type of seed
θ = time
σ = latent heat of melting of pure ice

This study will give important informations in the hydro-freezing techni-

que by liquid nitrogen, available as a new technique of processing agricultural products in the near future.

INTRODUCTION

The boiling is a kind of heat transfer phenomena which is caused by liquid being overheated in higher temperature than the boiling point at the pressure of the system when temperature difference exists between solid phase and liquid phase or between liquid phases. The boiling of water is one of its familiar cases. The efficiency of the heat transfer due to boiling is higher than the one of normal heat transfer.

The freeze-grinding of soybean seeds is usually carried out at low moisture contents and the study on the change of mechanical characteristics of soybean seed during freezing is already investigated. According to the results of freezing tests of various kinds of agricultural products by low temperature nitrogen gas reported by Mitsuda(1978), Fukusato(1983) and Matsushita(1984), the freezing curves for high moisture contents of soybean seeds had a unique shape like a saddle shape.

In this study, the basic experiments and theoretical analysis about the hydro-freezing method were conducted, directly using the boiling heat transfer by liquid nitrogen, which is high heat transfer efficiency, instead of freezing by nitrogen gas. Soybean seeds were adopted as materials used in this study.

MATERIALS AND METHODS

Soybean seeds used in this study were TAMAHOMARE veriety harvested at Fukuoka Agricultural Experimental Station on 9th of November, 1991.

These seeds were hand-harvested without using bean-harvester not to make any bruise on the surface and stored in a refrigerator at about 3°C until the time of the tests.

The schematic diagram of the apparatus used in this investigation is shown in Fig 1. It is hard to take photographs inside a dewar because it usually has mirror coat. Then, a dewar without mirror coating, which is 95mm in internal diameter and 190mm in hight, was newly made and used for the test.

When thermo-couples were inserted into a soybean seed at low moisture contents, it gets loose in contact with seed, with the increased in moisture content.

While, adjusting the moisture contents by adding distilled water before insertion of a thermo couple into the soybean seed caused some external damages such as wrinkling, splitting of the seed coat.

For these reasons, adjust of the moisture contents of soybean seeds and insertion of a thermo couple were conducted as follows.

First, about 50 seeds of soybean were put into a beaker for one day to get saturated. Then, a thermo couple was inserted into the soybean seed and was fixed with instant glue. Consequently, these soybean seeds inserted with a thermo couple were dried in a refrigerator to adjust its moisture contents. Soybean seeds were taken out of the refrigerator every other day and used for the tests.

Since soybean seeds are in rich in oily ingredients, drying 10g of soybean seeds in oven at 130°C for 4 hours, which was proposed by Tsutsumi et al.(1963), was adopted as a method for measuring the moisture contents of soybean seeds.

About 1 liter of liquid nitrogen was poured into the dewar bin and the boiling due to the heat of inner wall of the dewar bin was allowed to settle down with time.

When it came to the least bubbling state, T type thermo-couple and a soybean seed inserted with T type thermo-couple were immersed in the liquid nitrogen. Then, the temperature of the soybean seed and liquid nitrogen temperature were measured with the thermo-couples and recorded in a pen-recorder and also the photographs of the bubbling on seed surface were taken with a camera at the shutter speed of 1/4000 second.

DISCUSSION

1. Seed temperature and the boiling status of its surface

Fig. 2 shows the temperature change of soybean seed(12.1%, wet basis)during freezing in liquid nitrogen. Solid line in the figure is a freezing curve caliculated by a freezing theory written below. And Figs 3 through 5 are also photographs of the soybean seed surface. The numbers in parenthesis of each photograph correspond to the ones in Fig. 2. At 1.5 and 2.9 second after immersing, the bubbles generated on the surface by the nucleate boiling were combined and formed mushroom shape as the figure shows. This boiling mode is close to the film boiling mode, but it can be basically considered as the nucleate boiling mode because the soybean seed was not completely covered with bubbles. At 6.2 second after, it's more clear to be nucleate boilingmode. Compared to soybean seed, the size of the bubbles was ranged from 0.6to 1.0mm.

In the early stages, the rise of the bubbles generated from the bottom of soybean seed affected the heat transfer of the upper part of seed. The front line of the bubbles generated, moved from the bottom of the seed to the top withtime.

2. The relationship between freezing ratio and seed temperature

Fig. 6 is a typical figure showing the relationship between the solution temperature contained single solute and its composition. For example, when the state of the solution in agricultural products before freezing has the temperature and solution composition as shown at point A, it changes from the state A to B along the vertical line with cooling and ice is first formed at point B.

When the temperature of the solution goes down further reaching to the point C, more ice is generated and the solution becomes concentrated because the solute isn't desolved in the ice. Finally, the temperature reaching to the eutectic point where all the matter including the solute get freezed.

Since the solution of all the agricultural products includes many kinds of soluble ingredients, the concentration of the solution in the products goes up proceeded with cooling and then, what is called, freezing point lowering due to dripping is caused.

The relationship between freezing ratio and food temperature is already introduced from the Raoult's law on the freezing point lowering.

$$X = 1 - \frac{T_f}{T_s} \qquad (1)$$

However, this equation isn't applied in many case because most of the agricultural products have more than one kind of ingredient. Murata(1973) succeeded in simulating the fluidized bed freezing of peas precisely by using the following equation similar to the above equation.

$$X = \gamma - \frac{\delta}{T_s} \qquad (2)$$

The value of T_s in this equation was determined as the value when the freezing ration X equals to zero.

Then, this equation was applied to soybean seed in this study as well and the values of γ and δ in this case were determined by fitting data for beans to this equation because there is no data about soybean seeds. The data for beans were cited from Riedel's data printed in Kato's book(1966).

Fig. 7 shows the result of the fitting. The value of γ and δ for soybean seeds were determined as follows:

$\gamma = 1.03[-]$, $\delta = -0.949[^\circ C]$

These data plotted in Fig. 7 are for 89%(wet basis) initial moisture content of soybean seed. Therefore, the equation on freezing ratio X' for m'% initial moisture contents of soybean seed is

$$X' = \gamma' - \frac{\delta'}{T_s} \qquad (3)$$

where γ' and δ' are

$$\gamma' = 1 - \frac{m}{m'}(1-\gamma) \qquad (4)$$

$$\delta' = \frac{m}{m'}\delta \qquad (5)$$

3. The equations on boiling heat transfer

As seen in photographs, the boiling mode was the nucleate boiling mode and then the heat transfer equation on nucleate boiling was needed to simulate the freezing process of soybean seeds. Various forms of equations were proposed by many investigators because of the complex mechanism of nucleate boiling.

In this study, the applicability of the following three forms of equations were investigated by freezing simulation mentioned later.

Kutateladze Equation(1952)

$$\frac{q_s}{T_s - T_L}\cdot\frac{l}{\lambda_L} = 7.0\times10^{-4}\left(\frac{q_s l}{\nu_L L \rho_v}\right)^{0.7}\times Pr_L^{0.35}$$

$$\times\left(\frac{p}{\{\varepsilon g(\rho_L - \rho_v)\}^{1/2}}\right)^{0.7} \qquad (6)$$

Nishikawa and Yamagata Equation(1960)

$$\frac{q_s}{T_s - T_L}\cdot\frac{l}{\lambda_L} = 0.8\ \{(\frac{q_s l}{\nu_L L \rho_v})^{1/3}(\frac{q_s}{B^2C})^{1/3}\}\ \times Pr_L^{1/3}(\frac{p}{pa})^{2/3} \qquad (7)$$

where B and C are following constant.

$B = 900[1/m],\ C = 7.119[kJ/h]$

Labountzov Equation(1960)

Putting

$$\frac{q_s l}{\nu_L L \rho_v} = D$$

then, in case of D is less than 10^{-2}

$$\frac{q_s}{T_s - T_L}\cdot\frac{l}{\lambda_L} = 0.0625\cdot D^{0.5} Pr_L^{1/3} \qquad (8)$$

In case of D is larger than 10^{-2}

$$\frac{q_s}{T_s - T_L}\cdot\frac{l}{\lambda_L} = 0.125\cdot D^{0.65} \qquad (9)$$

l in the equation (6) and (7) is defined as follows:

$$l = \left(\frac{\varepsilon}{g(\rho_L - \rho_v)}\right)^{1/2}$$

also l in the equation (8) and (9) is of length demension not including gravity g. Therefore, the relationships between heat flux q_s and degree of superheating $T_s - T_L$ are simplified as:

Kutateladze Equation

$$q_s \propto (T_s - T_L)^{1/0.3} = (T_s - T_L)^{3.3} \qquad (10)$$

Nishikawa and Yamagata Equation

$$q_s \propto (T_s - T_L)^3 \qquad (11)$$

Labountzov Equation

$$q_s \propto (T_s - T_L)^2 \qquad (D<10^{-2})$$

$$q_s \propto (T_s - T_L)^{1/0.35} \qquad (D>10^{-2}) \qquad (12)$$

4. The equation for simulating the freezing of soybean seed

As seen in the above section, the heat flux q_s is propotional to the degree of superheating to the power in the range from about 2.0 to 3.3. Then, putting the power of degree of superheating as the sign of n and also the proportional constant as α, the equation on heat balance was introduced as follows:

$$-\frac{\partial}{\partial\theta}(C_s W T_s) + \sigma W_m\frac{\partial X}{\partial\theta} = \alpha(T_s - T_L)^n \qquad (13)$$

The first part of the left hand side of this equation expresses the change of latent heat, and the second part, the change of solidification heat due to freezing of the water contained in soybean seed.

The relationship among the specific heat C_s, moisture contents m and freezing ratio X for soybean seed is expressed by:

$$C_s = (3.352 - 2.149X)m + 1.159 \tag{14}$$

where the value of specific heat of dry mattter in soybean seed was cited from the report of Murata et al.(1987)

Substituting equation(3) and (14) into equation(13) gives:

$$\frac{\partial T_s}{\partial \theta} = - \frac{\alpha (T_s - T_L)^{3.3}}{W(C_s - \frac{\delta m(CT_s + \sigma)}{T_s^2})} \tag{15}$$

5. Freezing simulation and nucleate boiling heat transfer coefficient

Calculating equation (15) by the Runge-Kutta-Gill method, the freezing simulation curves were obtained as shown in Fig. 2, 8, 9 and 10. For comparing the applicability of the three forms of equations on nucleate boiling transfer mentioned above, the simulation curves calculated by those forms of equations were also plotted in the figures. The reason why the solid lines are seen as one line is that these three solid lines overlaped each other.

As the results of simulating, the proportional constant α for Kutateladze's equation was determined and the values are shown in each figure. In order to decide the real heat transfer coefficient h from this proportional constant α, following equation was used.

$$\alpha (T_s - T_L)^n = hA(T_s - T_L)$$

Therefore,

$$h = \frac{\alpha (T_s - T_L)^{n-1}}{A} \tag{16}$$

That is to say, the heat transfer coefficient changes with the freezing proceeds. The values of the heat transfer coefficient for each condition calculated from equation(16) were

Fig. 2(at 211°C of superheating) 1.25×10^3(kJ/m^2·h·°C)
Fig. 8(at 211°C of superheating) 1.34×10^3(kJ/m^2·h·°C)
Fig. 9(at 212°C of superheating) 1.31×10^3(kJ/m^2·h·°C)
Fig. 10(at 208°C of superheating) 1.44×10^3(kJ/m^2·h·°C)

CONCLUSIONS

In this study, the basic data needed for the application of boiling heat transfer in agricultural processing were taken and analyzed. Namely, the temperature change of soybean seed during freezing were measured and taken photographs of its surface with high speed camera and high sensitive films.

The following conclusions were drawn from the analysis of the results.

1)It was found that it is possible to simulate the freezing process of soybean seed by the following equation which includes the freezing equation as a function of seed termperature.

$$\frac{\partial T_s}{\partial \theta} = -\frac{\alpha (T_s - T_L)^{3.3}}{W\left(C_s - \frac{\delta m (CT_s + \sigma)}{T_s^2}\right)}$$

2)It was found from the photographs that the boiling mode was a nucleate boiling mode. The average heat transfer coefficients in nucleate boiling were obtained by simulating as follows:

$h = 1.45 \times 10^3 (kJ/m^2 \cdot h \cdot {}^\circ C)$ (at 218°C of superheating)

SYMBOLS

A = surface area of a soybean seed, [m^2]

C_s = specific heat of soybean seed, [$kJ/kg \cdot {}^\circ C$]

C_1 = specific heat of pure ice, [$kJ/kg \cdot {}^\circ C$]

C_2 = specific heat of pure water, [$kJ/kg \cdot {}^\circ C$]

C_3 = specific heat of dry matter contained in a soybean seed, [$kJ/kg \cdot {}^\circ C$]

g = gravitational acceleration, [m/s^2]

h = heat transfer coefficient in nucleate boiling, [$kJ/m^2 \cdot h \cdot {}^\circ C$]

L = latent heat of liquid nitrogen, [kJ/kg]

m = moisture content of soybean seed, [%, wet basis]

m' = do.

p = pressure of the system, [N/m^2]

p_a = atomospheric pressure at standard conditions, [N/m^2]

Pr_L = Prantle number of liquid, [-]

q_s = heat flux, [$kJ/m^2 \cdot h$]

T_f = freezing point, [°C]

T_L = temperature of liquid nitrogen, [°C]

T_s = temperature of soybean seed, [°C]

V_L = specific volume of liquid nitrogen, [m^3/kg]

V_s = specific volume of nitrogen gas, [m^3/kg]

W = mass of a soybean seed, [kg]

X = freezing ratio, [-]

α = proportional constant, [$kJ/h \cdot {}^\circ C^n$]

γ = a constant defined by equation(2), [no dimension]

γ' = a constant defined by equation(4), [no dimension]

δ = a constant defined by equation(2), [°C]

δ' = a constant defined by equation(5), [°C]

ε = surface tension of liquid nitrogen, [mN/m]

θ = time, [h]

λ_L = heat conduction coefficient of fluid, [m^2/s]

ν_L = kinematic viscosity of fluid, [m^2/s]

ρ_v = density of saturated vapour, [kg/m^3]

ρ_L = density of liquid, [kg/m^3]

σ = latent heat of melting of pure ice, [kJ/kg]

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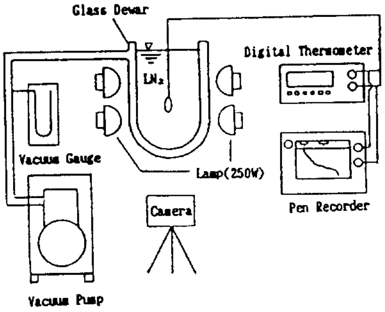


Fig.1 Schematic diagram of the experimental apparatus

Fig.3 Boiling status on the surface of soybean seed (at 1.5 second after immersing)

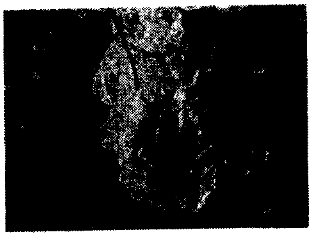
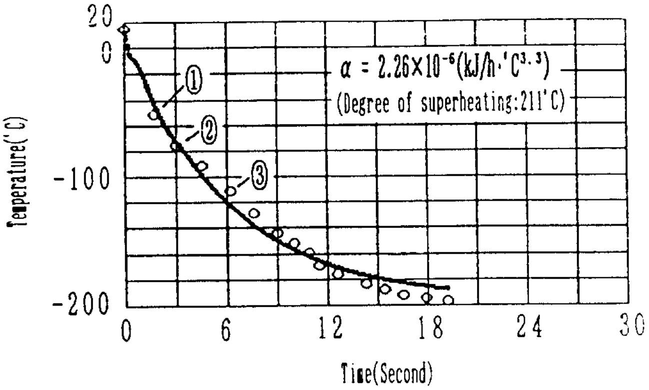


Fig.4 Boiling status on the surface of soybean seed (at 2.9 second after immersing)

Fig.5 Boiling status on the surface of soybean seed (at 6.2 second after immersing)



T_{SO} = 15.00 (°C) W = 3.0 ×10^{-4}(kg)
T_L = -195.8(°C) A = 4.00×10^{-4}(m^2)
$\Delta\theta$ = 0.2 (s) m = 12.1 (%, wet basis)

Fig.2 Freezing simulation for soybeans (at moisture content of 12.1%)

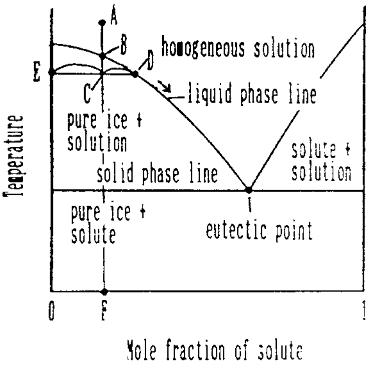


Fig.6 Scheme of phase changes for one component system

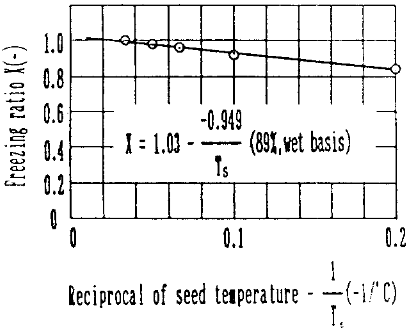


Fig.7 Relationships between freezing ratio and seed temperature

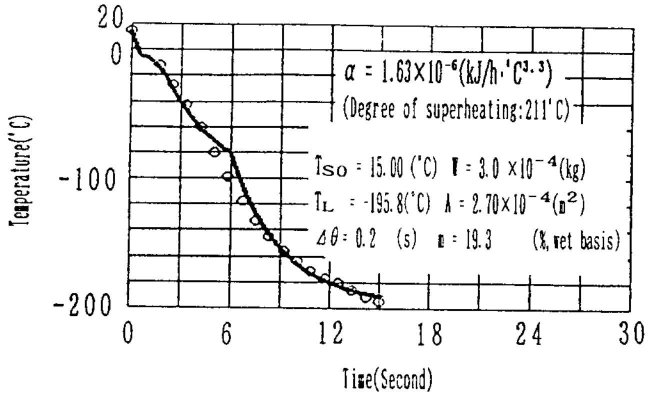


Fig. 8 Freezing simulation for soybeans
(at moisture content of 19.3%)

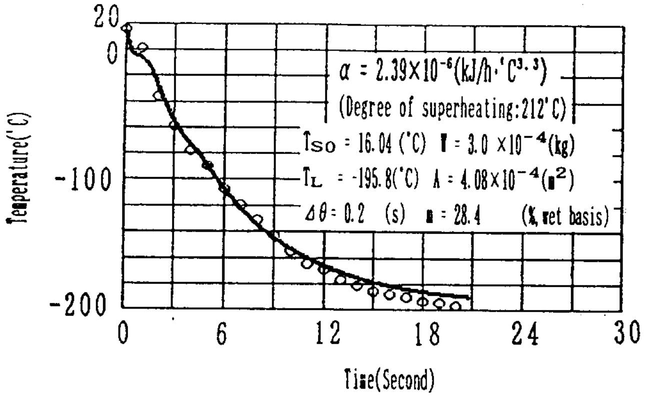


Fig. 9 Freezing simulation for soybeans
(at moisture content of 28.4%)

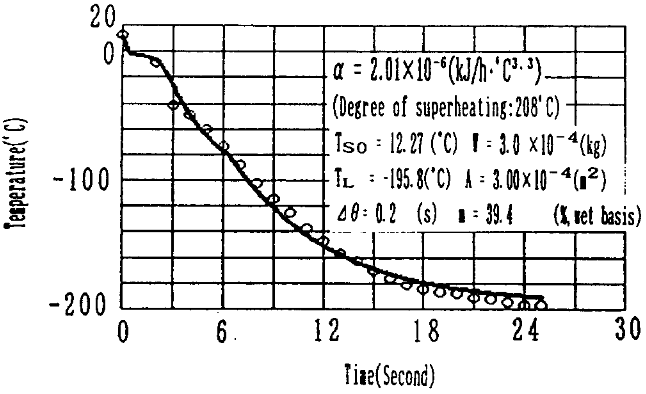


Fig. 10 Freezing simulation for soybeans
(at moisture content of 39.4%)