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Heat transfer to Liquid Nitrogen Droplets during Cyrogenic Freezing of Foods

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ABSTRAK

Kajian ini dilakukan untuk menyelidiki fenomena pemindahan haba yang terjadi apabila titik-titik cecair nitrogen jatuh ke atas permukaan makanan. Kepingan gelatin digunakan sebagai bahan makanan. Nitrogen cair dijatuhkan ke atasnya daripada sistem penjatuh/bakungan. Suhu kepingan serta masa penyejatan titik tersebut diukur bagi suatu jangkamasa tertentu. Daripada nilai-nilai ini, koefisien pemindahan haba dikira. Ia dibandingkan dengan nilai ramalan teori dan didapati nilai yang diperolehi adalah tiga hingga empat kali lebih tinggi. Walau bagaimanapun dengan mengambilkira bahawa titik tersebut menjadi semakin kecil semasa penyejatan, nilai purata koefisien pemindahan haba di dalam julat tersebut menjadi lebih hampir kepada nilai teori, iaitu kurang daripada dua kali ganda lebih tinggi.

ABSTRACT

This work investigates the heat transfer phenomena that occurs when liquid nitrogen droplets fall onto a food surface. A gelatin slab was used as the food material. Liquid nitrogen was dropped on it from a dropper/reservoir system. The temperature of the slab and the droplet evaporation time were measured over a period of time. The heat transfer coefficients were calculated from these values. These are compared with the theoretical predicted values. It is seen that the experimental values are three to four times higher than the predicted values. However, if the average value of heat transfer coefficient is taken over the whole size range as the droplet evaporates, a closer agreement is obtained — the experimental values being less than twice higher.

INTRODUCTION

Industrially, liquid nitrogen is used to cyrogenically freeze foods at it gives very high freezing rates due to its low boiling point of −195.8°C at atmospheric pressure. An added advantage is that it gives individually quick frozen items. Liquid nitrogen is one of the few refrigerants which can come into intimate contact with foodstuffs without causing adverse effects — it is tasteless, chemically inert, sterile and has a very low liquid viscosity.

Liquid nitrogen is sprayed onto the foods to provide the cooling effect. Freezing by immersion of foodstuffs in liquid nitrogen is generally regarded as an inefficient method despite a high heat transfer coefficient in the nucleate boiling region. This is due to the fact that the spheroidal heat transfer coefficient is a hundred-fold lower, i.e. the coefficient of heat transfer when a solid sphere or food material is in contact with the liquid refrigerant causing vigorous boiling as a result of the large temperature difference (Anon, 1969).

It is a normal practice to have an initial precooling section in a cryogenic freezer where the product is cooled by an air/nitrogen mixture at a temperature of around −100°C. In this section, the product is brought down to a temperature of around 0°C. This chilled product is then subjected to a spray of liquid nitrogen droplets. When the liquid nitrogen hits the food surface, it immediately enters into the film boiling regime, due to the large temperature difference between the product and the liquid nitrogen. Film boiling of discrete masses of liquid is commonly referred to as the Leidenfrost phenomena (Keshock and Bell, 1970); this is in constrast with pool film boiling where the liquid is of essentially infinite extent. A well known manifestation of the phenomenon is the action of water droplets splashed on a frying pan whose surface is so hot that nucleate boiling cannot occur.

In the Leidenfrost region, since there is considerable vapour formation, the droplets are surrounded by a vapour blanket. When they meet the food surface, the liquid evaporating from the bottom part of the drop produces a "vapour cushion" with a slight positive pressure (Wachters and Bonne, 1966) — enough to keep the drop suspended above the food surface (See *Fig. 1*). Hence the drops do not actually touch the food surfaces, and this has the effect of reducing the overall heat transfer, since heat transfer then occurs by conduction through a vapour phase (Keshock and Bell, 1970).

Fig. 1. Liquid nitrogen drop on a food surface.

From the boiling heat transfer curve shown (Flynn *et al.*, 1962) *(Fig. 2)*, the region of nucleate boiling (region B) gives high heat flux, i.e. high heat transfer coefficients. Unfortunately, due to the Leidenfrost boiling (film boiling of liquid nitrogen drops) which involves a blanketting vapour film, the heat transfer coefficient is reduced. Hence a much higher temperature difference is required for the same heat flux (region D).

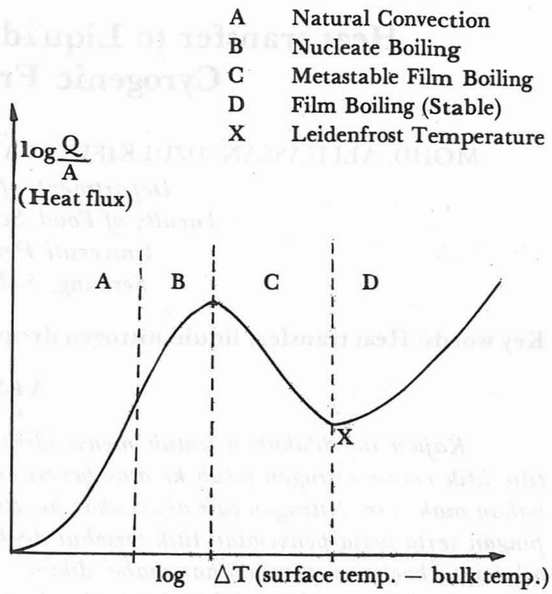


Fig. 2. Boiling heat transfer curve.

In the freezing process, we aim to achieve high heat transfer rates. Since high heat transfer rates are possible with lower values of ΔT (region B), it is therefore evident that the freezing process needs to be closely controlled. If the foodstuffs are immersed in liquid nitrogen, the temperature is high enough to push the point of boiling towards region D. In the spray system however, the process can be controlled by spraying liquid nitrogen droplets onto the food surface and using the cold gaseous mixture of air and nitrogen at around −100°C to provide the precooling. This also has the advantage of minimising bio-physical stresses in the food and reducing damage to the cells.

Heat Transfer Coefficient

Baumeister, Hamill and Schoessow (1966) used dimensionless parameters for volume, area, latent heat and vapourisation time to predict the Leidenfrost heat transfer coefficients. Different correlations are given for different drop sizes. The size range is classified by using the dimensionless volume V* given by:

$$V^* = \frac{V}{(\sigma/\rho_L)^{3/2}}$$

The classification is given in Table 1.

In the present work, the drop diameter used is 2.37 mm. When the values of 0.0827 N/m and 804 kg/m^3 are substituted for the surface tension and density of liquid nitrogen at atmospheric pressure respectively (Weast, 1973), a value of 6.9 for V* is obtained. Hence we are in the large drop domain.

Keshock and Bell (1970) pointed out that the parameters are 30% below the theoretical curve for liquid nitrogen Leidenfrost phenomenon although the experimental data conforms with the general trend of the theory. They also suggested that the actual heat transfer coefficient in the Leidenfrost boiling region was 30% higher than the prediction made by Baumeister *et al.* (1966).

Baumeister's analyses were based upon a drop model in the shape of a cylindrical disk, separated from the solid surface by a constant gap thickness. In his model, drop vapourisation occurs because of energy addition to the lower drop surface by conduction across the constant vapour-film thickness; mass diffusion from the top and sides is assumed negligible and radiation is neglected.

The research work carried out in this project involves measuring the evaporation time associated with single drops dropped onto a gelatin slab and the temperature drop experienced by the gelatin slab. These enable us to calculate the heat transfer rates and the heat transfer coefficient. The results obtained are then compared with the theoretical predictions of heat transfer coefficients due to film boiling of nitrogen drops on horizontal surfaces. These results on single droplet behaviour should give an insight into the problems associated with the spray system in the liquid nitrogen freezing plant.

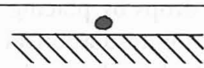
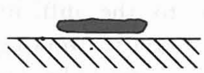
MATERIALS AND METHODS

In order to produce single drops of nitrogen, a dropper system was designed. It consisted of a copper cylinder; 2.5 cm internal diameter and 20 cm long. A piece of 2.5 cm diameter brass with a small spout was welded to the base of the cylinder. The brass had a 3 mm hole drilled at the centre. A small 26 gauge hypodermic needle was attached to the spout. The whole unit was insulated with 7 cm thickness expanded polystyrene, to reduce the rate of heat flow into the reservoir when it was filled with liquid nitrogen. In order to obtain single drops of liquid nitrogen, a very fine wire was inserted through the hypodermic needle and bent to form a very small loop. Single drops of consistent size was formed.

A gelatin slab about 1.5 cm thick in a petri dish was used as the food material. The gelatin was prepared by dissolving 10 grams of gelatin into 100 ml of hot water to obtain the right texture and hardness. The ratio of 1 : 10 was found to be satisfactory.

When the nitrogen droplets fall on the surface, it moves about from side to side, effectively floating on the vapour cushion. In order to keep it stationary, a small indentation was made

TABLE 1
Classification of drop domain

Volume Range	Drop Domain	Representation
V* ≤ 0.8	Small spheroid	
0.8 < V* ≤ 155	Large drop	
V* > 155	Extended drop (constant thickness)	

on the surface and liquid nitrogen was dropped into it.

As mentioned earlier, industrially the freezing of foods using liquid nitrogen is done in a medium consisting of nitrogen and air at about −100°C. However in the present study, with liquid nitrogen droplets falling over a distance of about 3 cm onto the gelatin slab in open laboratory conditions, the air/nitrogen cloud surrounding the slab surface was at −10°C to −20°C. Hence to obtain results at medium temperatures around −100°C, a separate cooling apparatus was required.

A dewar vessel with a small window was tried. Temperatures of −100°C could be easily achieved inside the dewar when it was filled with liquid nitrogen. However, when the gelatin slab was placed in the cold environment at −100°C, there was considerable 'haze', i.e. a foggy environment was produced. It was impossible to see whether the nitrogen drops had fallen exactly into the surface indentation on the slab. A different apparatus which consisted of a reservoir with provision for liquid nitrogen to be circulated around its base (see *Fig. 3*) to provide the cooling effect was tried. This was done in order to reduce the temperature of the surrounding air around the bottom of the reservoir. The reservoir consisted of a copper cylinder of 14 cm internal diameter and 20 cm in height. A similar hypodermic needle was attached to its base to reduce the nitrogen drops. The reservoir was insulated with 5 cm of polystyrene, but this time the base was left uninsulated to produce the cold environment.

When this apparatus was tried out, there was a lot of 'drip' and wetting on the outside at the base of the copper vessel due to condensation of atmospheric oxygen which provided additional cooling on the gelatin slab. An attempt to eliminate the drops by placing a very thin layer of glass fibre for insulation at the base of the reservoir resulted in the temperature of the air around the base of the cylinder being only −30°C. Due to the difficulties involved in achieving the −100°C working environment, it was decided to restrict the experiments on the heat transfer rates to the higher temperature range (from 0°C to −30°C).

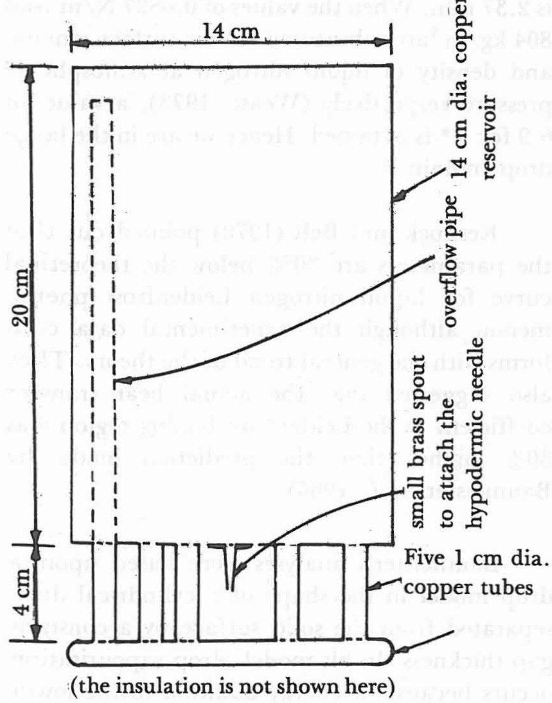


Fig. 3. The modified dropper/reservoir system.

Temperature Measurement

Measurement of temperature was done by placing a thermocouple just underneath the indentation and another at a point 3 cm away. The temperature measurement was recorded on a chart recorder. Hence a comparison could be made between the cooling due to the nitrogen drops and that due to the cold medium.

Evaporation Time of the Droplet

The evaporation time of the drops is difficult to measure visually, for evaporation occurs at a rapid rate and moreover it is difficult to determine when exactly the drop has totally evaporated. However, from the chart recorder measurements of the temperature against time, it was observed that when the droplet fell into the hole, the temperature there started to drop sharply. When the droplet had evaporated, there was again a sharp increase in the temperature. Hence, the evaporation time of the droplet on the gelatin surface can be taken as the time from which the temperature measurement on the recorder starts to fall sharply until it starts to rise sharply again.

Droplet Size

The droplet size was measured by an impact technique. The droplets were allowed to fall onto fine nickel powder, placed in a petri dish at a distance of 3 cm from the droplet formation site i.e. of equal distance between the droplet formation site and the gelatin slab. The droplets evaporated on this powder and left a crater. The crater size diameter was measured for several drops. It was observed that by using the same 26 gauge needle and the fine wire through it, the diameter of the crater was quite consistent.

From the diameter of the crater, the actual size of the drops is given by the formula quoted by May (1950) as

$$d = I_c \times C_t$$

where d is the actual diameter of the drop; I_c is a coefficient which depends on the droplet size range, and C_t is the crater diameter. For drop sizes from 200 to 2000 microns, May suggested a value of 0.86 for C_t.

RESULTS

The instantaneous temperature drop of the gelatin slab due to the heat transfer by the liquid nitrogen droplets were measured at medium temperature of 0°C to −30°C to determine the effect of medium temperature on the heat transfer rate. The other parametres (the droplet size, the initial temperature of the gelatin slab, the frequency of the droplets) were kept constant. The results are shown in *Fig. 4*. The droplet evaporation time at a medium temperature of −20°C is given in *Fig. 5*.

Heat Transfer Rates

Neglecting heat transfer by radiation and convection to the surrounding medium, the rate of heat transfer due to the nitrogen droplets on

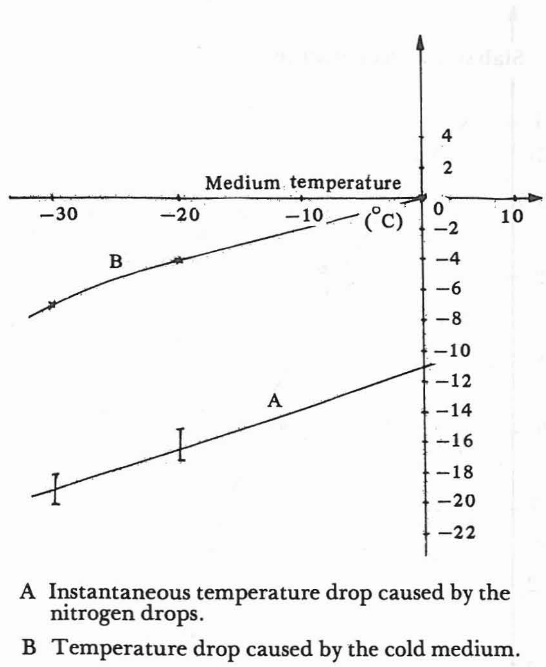


Fig. 4. Temperature of slab surface against medium temperature.

the gelatin surface is given by:

$$Q = h.A. \quad \Delta T \qquad (1)$$

where h is the overall heat transfer coefficient,

A is the effective heat transfer area of the drop on the gelatin surface, and

ΔT is the temperature difference between the droplet and the surface of the gelatin.

The effective area of the droplet, is given by Baumeister *et al.* (1966) as the average of the projected area of the sphere (i.e. πr^2) and the surface area of the lower half of the sphere (i.e. $2 \pi r^2$). So, the effective area is

$$A = 1.5 \quad \pi r^2$$

This is for a small liquid droplet on a flat surface. In the present work, since the droplets actually fall into an indentation on the surface whose diameter is of the same order of magnitude as the drop, it is deemed more reasonable to assume that a larger part of the droplet surface area is involved in the heat transfer to the gelatin slab. The surface area chosen is half the surface

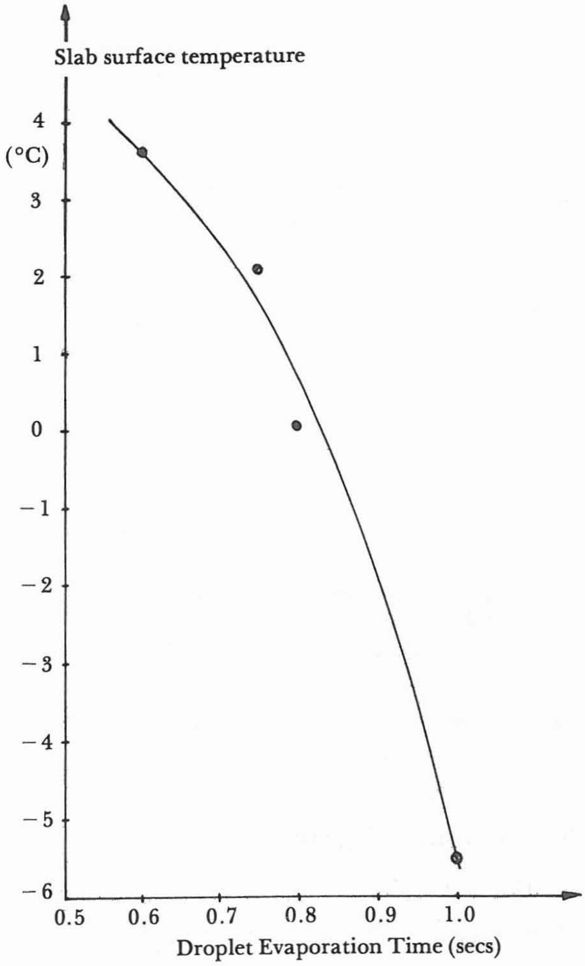


Fig. 5. Graph of slab surface temperature against droplet evaporation time.

area of a sphere, i.e. the surface area of the lower half of the drop. So, the heat transfer area A is given by:

$$A = 2 \pi r^2$$

If we assume that the rate of evaporation of the drops is constant, then the heat transfer rate Q is also given by:

$Q = \lambda x$ (rate of evaporation of drops in kg/s)

$= \lambda x$ (volumetric rate of evaporation of drop $x \rho_L$)

If we assume that the droplets are spherical (justifiable for small drops with $V^* = 6.9$), then the initial drop volume is $V = \frac{\pi}{6} d^3$

when d is the initial drop diameter. The final drop volume is zero since all the drop evaporates.

If we let the time for the droplets to evaporate be t, then the heat transfer rate

$$Q = \lambda x \pi x \rho_L x d^3/6 x t \qquad (2)$$

Heat Transfer Coefficient

Equating equation (1) and (2) and rearranging, we get

$$h = \frac{\pi}{6} . \lambda . \rho_L . d^3/A . \Delta T.t \qquad (3)$$

Table 2 gives the evaporation time of the droplet at various temperatures of the slab and the average temperature difference ΔT. Values of 191500 J/kg and 804 kg/m^3 are used for the latent heat and density of liquid nitrogen at atmospheric boiling point. The initial droplet diameter is 2.37 mm. The average temperature difference ΔT is taken as the difference between the atmospheric boiling point of liquid nitrogen and the average temperature of the gelatin slab due to the liquid nitrogen droplets at the indentation. The droplet evaporation time is obtained from *Fig. 5.*

The heat transfer coefficient between the nitrogen drops and the gelatin surface calculated from equation (3) is plotted against the freezing time in *Fig. 6.*

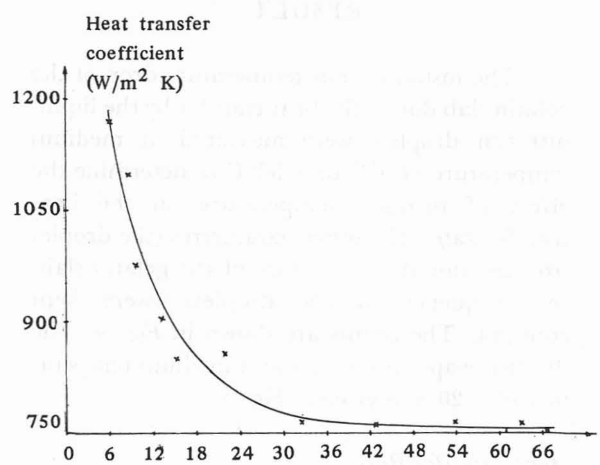


Fig. 6. Experimental heat transfer coefficient against time.

TABLE 2
Droplet evaporation time at Mean Slab Temperatures
(Medium temperature: − 20°C)

Temp. of slab surface (°C)	Average temp. of slab surface due to nitrogen drops (°C)	Average Temp. Diff. ΔT (°C)	Droplet Evaporation Time (secs)
3.5	− 23.5	172.3	0.600
3.0	− 25.0	170.8	0.650
2.0	− 25.0	170.8	0.725
1.0	− 24.5	171.3	0.780
0	− 23.5	172.3	0.825
− 1.0	− 32.5	163.3	0.865
− 2.0	− 20.0	175.8	0.900
− 3.0	− 26.0	169.8	0.935
− 4.0	− 32.0	163.8	0.965
− 5.0	− 35.0	160.8	0.85
− 5.5	− 35.5	160.3	1.000

DISCUSSION

The theoretical value for the heat transfer coefficient is calculated for comparison with the experimental values. In the large drop domain, Baumeister *et al.* (1966) suggested that

$$h = 1.075 \left[\frac{k^3 \cdot g^{1/2} \cdot \sigma^{1/2} \cdot \rho_L{}^{1/2} \cdot \rho_V \cdot \overset{\circ}{\lambda}}{\Delta T \cdot \mu_V \cdot V^{2/3}} \right]^{1/4} \quad (4)$$

The temperature of the vapour in between the drop and the slab is assumed to be between − 195.8°C and − 30°C (slab temperature). The values of the vapour properties used are at − 173°C. The modified latent heat of vapourisation of liquid nitrogen (Baumeister *et al.* 1966) is given by:

$$\lambda^* = \lambda \left[1 + \frac{7}{20} \times \frac{C_p \cdot \Delta T}{\lambda} \right]^{-3}$$

The droplet diameter that is involved is only 2.37 mm. Due to the low surface tension of nitrogen, the dimensionless drop volume V* gives a value of 6.9. Hence it is in the large drop domain. Since the value of 6.9 for V* is at the low end of the range for the large drop, and since the drop actually sits around the identation on the slab, it is deemed reasonable to assume that the droplets acquire a greater degree of sphericity than on a flat surface. Hence it is possible to assume that the droplets are in fact spherical and in the small spheroid drop domain, where the heat transfer coefficient (Baumeister *et al.*, 1966) is given by:

$$h = 1.1 \left[\frac{k^3 \cdot \lambda^* \cdot g \cdot \rho_L \cdot \rho_V}{\Delta T \cdot \mu_V \cdot V^{1/3}} \right]^{1/4} \quad (5)$$

Table 3 gives the value of the heat transfer coefficient for different ΔT for both the large drop and the small drop domain, calculated using equations (4) and (5) respectively.

Hence it is seen that by assuming a large drop domain, the heat transfer coefficient is between 275 W/m^2K and 284 W/m^2K; whereas by assuming the drop to be in the small drop domain, the value of the heat transfer coefficient is between 329 W/m^2K and 339 W/m^2K. However the experimental value is from 1174 W/m^2K to 758 W/m^2K.

The theoretical value of the heat transfer coefficient assuming a small drop domain gives a

TABLE 3
Theoritical heat transfer coefficient at different ΔT

Time (mins)	Average ΔT (°C)	λ^* (J/kg)	Heat transfer coefficient (W/m^2K) h; assuming Large drop	Small drop
6	172.3	82231	275	329
8.5	170.8	82760	276	330
9.5	170.8	82760	276	330
13	171.3	82583	276	330
15	172.3	82231	275	329
21.5	163.3	85478	282	337
32	175.8	81012	273	326
42	169.8	83116	277	331
53	163.8	85293	281	336
62	160.8	86410	284	339
65	160.3	86598	284	339

better agreement to the experimental value compared with the heat transfer coefficient calculated by assuming the large drop domain. This suggests that the drop may actually be more spherical than it appears to be by just considering the V* range.

However the experimental values are higher by a factor of three to four than the theoretical ones. Keshock and Bell (1970) also reported this where they showed that for the larger drops, the difference between experimental and theoretical values is large, but for small droplets the agreement was quite good. In this present work however, the difference is quite large although we are dealing with small drop sizes.

If we consider that the droplets actually decrease in size as they evaporate, we can calculate the theoretical heat transfer coefficient for varying drop sizes (see Table 4).

The values of the heat transfer coefficient in Table 4 is calculated at 168 K, i.e. the arithmetic mean of the temperature difference ΔT given in Table 2.

It is evident from Table 4 that the heat transfer coefficient increases with decreasing drop sizes. In the experimental value, the heat transfer coefficient is calculated using the largest drop size and the evaporation rate was assumed constant. If we compare the experimental value with the average value of the heat transfer coefficient at varying drop sizes, we get a better agreement with the theory.

TABLE 4
Theoretical heat transfer coefficient at different drop sizes

Drop diameter (m)	Drop volume V (m^3)	dimensionless drop Volume V*	Heat transfer coefficient h (W/m^2K)
0.00237	6.970175×10^{-9}	6.5 (large drop)	278
0.001	5.235988×10^{-10}	0.49 (small drop)	413
0.0005	6.544985×10^{-10}	0.06 (small drop)	491
0.00005	6.544985×10^{-14}	0.000061 (small drop)	873

The other factor that is noticeable is that from the theory, the heat transfer coefficient rises slightly with time, i.e. it rises with decreasing ΔT. This is not so in the experimental values which decrease with decreasing ΔT. In the theoretical expressions the only variable is ΔT (since λ^* also varies with ΔT). λ^* increases with decreasing ΔT and the total effect is to increase the heat transfer coefficient. In the experimental expression, the heat transfer coefficient is inversely proportional to the product of ΔT and the evaporation time t. Although ΔT decreases with time, t increases from 0.6 seconds to 1.0 second. The sum effect is to reduce the heat transfer coefficient with decreasing ΔT. This difference in behaviour between the theoretical and the experimental values is believed to have been due to the fact that the evaporation time included in the calculation of the experimental value varies with ΔT, whereas in the theoretical expressions, no evaporation time is included.

The droplet evaporation time increases with decreasing ΔT. This is expected since the decreasing ΔT, the driving force for evaporation to occur is less and hence it takes longer to evaporate.

CONCLUSION

The experimental heat transfer coefficient obtained is about three to four times higher than the theoretical predictions. Keshock and Bell (1970) also found that the theoretical values are lower than the experimental ones, but they reported that for small droplets (less than 0.1 ml) the difference is not excessive. The droplet size that we are working with is much less than this, but still the difference is quite substantial. If however, we taken an average value of the theoretical heat transfer coefficient over varying drop sizes as the droplet evaporates, we get quite a good approximation to the theory — the experimental values being higher by a factor of one and a half to two.

From the V* of 6.9, the droplets fall into the large drop domain. It is believed that since the drops were made to fall into a hole, it takes a more spherical shape. By calculating the theoretical values of the heat transfer coefficients, the values in the smalldrop domain gives a better approximation to the experimental ones. This suggests that the droplets may in fact be spherical.

Another difference between the theoretical and experimental values is that whilst the experimental values of the heat transfer coefficient decrease with decreasing ΔT, the theoretical values show a slight increase. In the expression to calculate the experimental value the vapourisation time is present and has the effect of decreasing the heat transfer coefficient. In the theoretical expression however, the only variable is ΔT.

It is thought essential that for future work in this area, some thought be given to designing a dropper system which would produce consistent drop frequency throughout the entire experiment. Ice deposition can be reduced if the work is done in a more enclosed environment. Also, in order to achieve realistic values of the heat transfer coefficient as in the cryogenic freezers, the medium temperature needs to be around −100°C. More accurate values for the droplet evaporation time can be obtained by using photographic techniques.

It is suggested that future work should include photographic and microscopic studies of the droplet on the food surface. From these we can ascertain what the drop domain is. It might provide further insight into the Leidenfrost boiling phenomena.

The effect of droplet velocity is not included here. Further research could include this variable. It is believed that the droplet velocity in the spray system can have a marked effect on the heat transfer coefficient.

Evaporative cooling effect is also not dealt with here. If the temperature of the droplets can be measured as it evaporates, then this effect can be taken into account. If it is significant, then the temperature difference between the droplet and the slab can be corrected. This would have an effect of reducing the experimental heat transfer coefficient.

NOMENCLATURE

A	surface area of droplet
A^*	dimensionless surface area of droplet
C_p	specific heat of vapour at constant pressure
f	radiation correction factor
g	gravitational acceleration
h	heat transfer coefficient to the drop
h^*	dimensionless heat transfer coefficient to the drop
k	thermal conductivity of vapour underneath drop
t	evaporation time of droplet
t^*	dimensionless evaporation time to droplet
T_p	temperature of food surface
T	temperature difference between food surface and the drop
V	volume of drop
V^*	dimensionless volume of drop
σ	surface tension of liquid droplet
λ	latent heat of vapourisation
λ^*	modified latent heat of vapourisation
ρ_L	liquid density
ρ_V	vapour density
μ	dynamic viscosity of vapour

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