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EXPERIMENTAL DETERMINATION OF SURFACE HEAT TRANSFER COEFFICIENT IN A DRY ICE-ETHANOL COOLING BATH USING A NUMERICAL APPROACH

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Abstract

BACKGROUND: Dry ice-ethanol bath (-78ºC) have been widely used in low temperature biological research to attain rapid cooling of samples below freezing temperature. The prediction of cooling rates of biological samples immersed in dry ice-ethanol bath is of practical interest in cryopreservation. The cooling rate can be obtained using mathematical models representing the heat conduction equation in transient state. Additionally, at the solid cryogenic-fluid interface, the knowledge of the surface heat transfer coefficient (*h*) is necessary for the convective boundary condition in order to correctly establish the mathematical problem. **OBJECTIVE:** The study was to apply numerical modeling to obtain the surface heat transfer coefficient of a dry ice-ethanol bath. **MATERIALS AND METHODS:** A numerical finite element solution of heat conduction equation was used to obtain surface heat transfer coefficients from measured temperatures at the center of polytetrafluoroethylene and polymethylmetacrylate cylinders immersed in a dry ice-ethanol cooling bath. The numerical model considered the temperature dependence of thermophysical properties of plastic materials used. **RESULTS:** A negative linear relationship is observed between cylinder diameter and heat transfer coefficient in the liquid bath, the calculated *h* values were 308, 135 and 62.5 W/(m^2K) for PMMA 1.3, PTFE 2.59 and 3.14 cm in diameter, respectively. **CONCLUSION:** The calculated heat transfer coefficients were consistent among several replicates; *h* in dry ice-ethanol showed an inverse relationship with cylinder diameter.

Keywords: heat transfer coefficient, dry ice-ethanol cooling bath, unsteady state heat conduction

INTRODUCTION

Dry ice-ethanol bath (-78ºC) is widely used in biological research to attain rapid cooling of samples below the freezing temperature. The cryopreservation of biological materials is a cornerstone tool in biomedical research and has numerous applications in cell banking and therapeutic treatments (16, 26). Although most studies on reproductive cells have been made using liquid nitrogen baths, several studies have also reported cryopreservation of animal gametes in liquid dry ice-ethanol cooling baths (17, 20) or in the cryopreservation of plant genetic resources (14). In 1973, Bank and Mazur (3) used a dry ice-ethanol bath to study the effect of cooling rates on the survival of yeast cells subjected to freezing. Studies of cryopreservation of human cells and tissues have also utilized dry-ice ethanol baths (27). Holovati et al. (12) reported the use of a dry ice-methanol bath for studying cryopreservation of human red blood cells. Albrecht and Schumacher (1) studied the viability of frozen measles virus. In all cases, the knowledge of cooling rates is essential to achieve maximum cell survival and resumption of tissue function.

Recently, several studies reported the use of mathematical models to predict cooling rates of small freeze devices immersed in liquid nitrogen (13, 23, 24). The numerical simulation has been performed by solving the non-stationary heat transfer partial differential equation considering the geometry of the devices (2, 9, 23). Those authors have emphasized the importance of knowing the surface heat transfer coefficient in order to solve the heat conduction equation with the convective boundary conditions. The surface heat transfer coefficient is an important parameter that enables the estimation of cooling rates (23). However, reports for heat transfer coefficients in dry ice-ethanol slurry are scarce or simply not available, preventing the prediction of cooling rates of biological samples immersed in the liquid bath. Surface heat transfer coefficient depends on several factors such as the geometry of the system, the type of cryogenic fluid and its properties, the fluid dynamics conditions around the solid etc (2, 4, 10, 11, 18, 19). The knowledge of *h* values is very important for the mathematical modelling of heat transfer; without this information it is impossible to predict accurate cooling rates.

The objective of the present study was to perform transient heat transfer experiments measuring time-temperature histories and to apply the finite element method to calculate the surface heat transfer coefficients in plastic cylinders of different diameters, immersed in dry ice-ethanol cooling bath (-78ºC). The *h* values were compared with literature correlations.

MATERIALS AND METHODS

Cooling bath

Ethanol (98%v/v) was poured into the insulation box and chips of dry ice were slowly added until temperature was stabilized at -78ºC. The insulation box consisted of two polystyrene foam boxes, one inside the other. Additional insulation was provided by foam peanuts and an air cushion between two boxes. Insulation box was covered with a polystyrene foam lid which had been perforated to introduce plastic cylinders used in measurements.

Experimental setup

Three cylinders of different diameters were utilized in this study. Two cylinders were made of polytetrafluoroethylene (PTFE), 2.59 cm and 3.14 cm in diameter and 10.0 cm long, and one of polymethylmetacrylate (PMMA), 1.30 cm in diameter and 11.0 cm long. The cylinders were drilled at the center to insert a small thermocouple (diameter = 0.14 cm) allowing to record the time-temperature history. Once the temperature of the cooling bath stabilized, the cylinders containing the thermocouple were immersed vertically in the systems and temperatures were recorded every 10 seconds. Measurements were conducted in four to eight replicates. Temperatures were recorded using a Testo 735-1 measuring instrument (Testo AG, Lenzkirch, Germany), fitted with a type T copper-nickel immersion probe (-200 to 40°C). The thermocouples were pre-calibrated. Figure 1 shows the experimental setup.

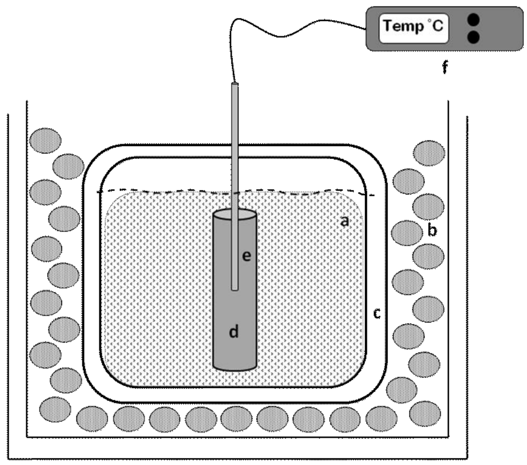


Figure 1. a) The dry ice-ethanol liquid bath, b) air/styrofoam pellet insulation, c) styrofoam insulation, d) PTFE or PMMA cylinder, e) type T copper-nickel probe fit snug into the drilled cylinder, f) data acquisition system. Dry ice-ethanol bath was prepared by addition of dry ice chips during stirring until system reached a stable temperature of -78°C.

Thermophysical properties of PTFE & PMMA

In cooling experiments, the temperature of plastic cylinders changed from room temperature to -70ºC, indicating thermo-physical properties (i.e., specific heat and thermal conductivity) of PTFE and PMMA may be temperature dependent. A literature survey was performed (5, 21, 22, 29) on this temperature effect (Table 1). The numerical model used to calculate heat transfer coefficients utilized the values of these thermophysical properties.

PTFE density and thermal conductivity ρ= 2170 kg/m^3 and k=0.32 W/m K were considered constant in the temperature range used. However, the specific heat is temperature-dependent (also shown in Table 1) (5, 22). For PMMA the density ρ=1202 kg/m^3 was assumed constant in the temperature range where the experiments were conducted, and the thermal conductivity as well as specific heat were considered temperature-dependent (21, 29).

Numerical modeling

Heat conduction equation in transient state describing the cool bath is given in Eq. 1 (23, 24)

$$\rho(T)Cp(T)\frac{\partial T}{\partial t} = \nabla \cdot (k(T)\nabla T) \qquad (1)$$

where: T is temperature, ρ is density, C*p* specific heat, k thermal conductivity.

A convective boundary condition was considered at the interface solid-cryogenic fluid.

$$q = -k(T)\nabla T * n = h(T - T_{ext}) \quad (2)$$

where *h* is the surface heat transfer coefficient, k is the solid thermal conductivity, T is the variable surface wall temperature of the interface solid -cryogenic liquid, T_{ext} is the external temperature (the temperature of dry ice-ethanol system), n is the normal outward vector, and ∇T is the temperature gradient evaluated at the surface.

The non-linear mathematical problem was solved by using the finite element method considering the temperature dependence of thermal properties. The software COMSOL was implemented to predict the temperature evolution in the geometrical center of each cylinder.

Calculation of surface heat transfer coefficients

Different heat transfer coefficients at the boundary condition were used to simulate temperature profiles by solving Eq 1 and Eq 2 numerically using the finite element analysis; variable thermal properties (density, specific heat and thermal conductivity) of materials were introduced in the program. The measured and predicted temperatures for each proposed *h* coefficient were compared. The heat transfer coefficient that minimized the residual sum of squares (RSS) as given by Eq 3 was selected.

$$RSS = \sum(T_{exp} - T_{pred})^2 \qquad (3)$$

Table 1. Thermal properties of PTFE and PMMA at various temperatures. Adapted from Warfield (29), Pradhan (21), Blumm (5), Rae (22).

T (ºC)	PMMA		PTFE
	K (W/m² K	*Cp (J/kg*	*Cp (J/kg*
30	--	--	1050
23	0.195	1470	--
25	--	--	1030
10	--	1388	--
0	0.192	1357	990
-13.2	--	1350	--
-23.2	--	1325	--
-25.0	--	--	910
-33.2	--	1290	--
-43.2	--	1265	--
-50.0	0.188	--	840
-53.2	--	1234	--
-63.2	--	1190	--
-73.2	--	1143	--
-75.0	0.182	--	780

RESULTS

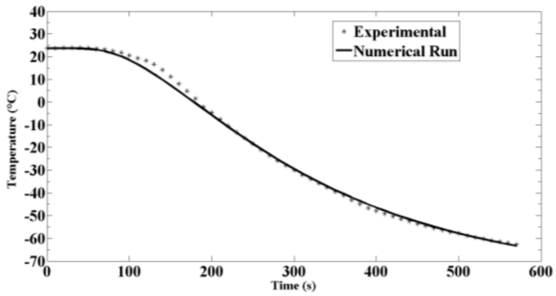
Heat transfer coefficients of the cooling bath

Fig 2a and 2b show the close agreement of the predicted temperatures with the measured temperatures. The prediction was made via the simulation with the calculated values of heat transfer coefficients. The similar agreement was obtained for all experiments.

The values of heat transfer coefficients for cylinders immersed in dry-ice ethanol are shown in Table 2. The consistence among different measurements (i.e., replicates) is very good as indicated by the small values of deviation (95% confidence interval). It is noteworthy that Torres-de María et al. (28) measured the heat transfer coefficient of a cooper cylinder 2 cm diameter immersed in a static brine solution at -7ºC and derived a value of 204 W/(m²K), which is in the good agreement with present study.

The effect of the cylinder diameter on heat transfer coefficient in a bath of dry ice-ethanol is

(a)



(b)

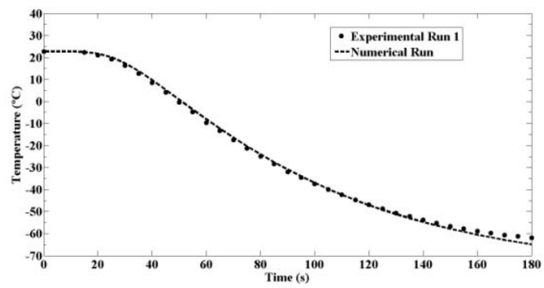


Figure 2. Predicted and measured center temperature of PMMA cylinder 1.30 cm diameter **(a)** and of PTFE cylinder 2.59 cm diameter **(b)** immersed

Table 2. Heat transfer coefficients (h) for dry ice-ethanol cooling bath. (mean±95% C.I.)

Diameter (cm)	No of replicates	*h* W/(m² K)
1.30 (PMMA)	8	309 ± 22
2.59 (PTFE)	4	135 ± 8
3.14 (PTFE)	4	62.5 ± 7

shown in Fig 3. A liner relationship with negative slope is observed between cylinder diameter and heat transfer coefficient in the liquid bath.

DISCUSSION

Comparison of h with literature correlations

The *h* values from the present work are compared with literature correlations of Nusselt number (Nu=h*L/k) for natural convection and also forced convection at low Reynolds numbers.

In the case of natural convection for vertical cylinders, the correlations valid for vertical plates can be applicable (7):

$$Nu_L = 0.68 + 0.67 * Ra^{0.25} * \left[1 + \left(\frac{0.492}{Pr}\right)^{9/16}\right]^{-4/9} \quad (4)$$

where R_a is the Rayleigh number defined as G_r*P_r; Gr_L is the Grashof number defined as follows:

$$Gr_L = \frac{\beta * (Ts - T\infty) g L^3}{\upsilon^2} \quad (5)$$

where *L* is the length of the vertical surface, *β* the volumetric thermal expansion coefficient, Ts surface temperature of the solid, T_∞ temperature of cryogenic fluid, *g* the acceleration of gravity,

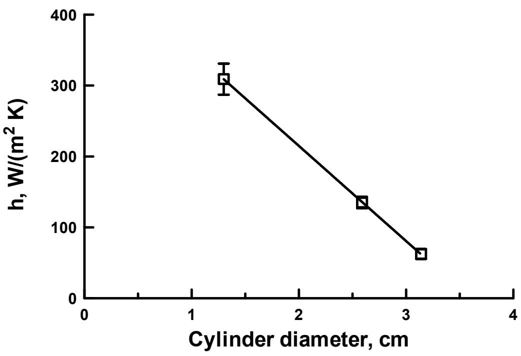


Figure 3. Effect of cylinder diameter on heat transfer coefficient in dry ice-ethanol bath. The error bars are not shown smaller than the symbols.

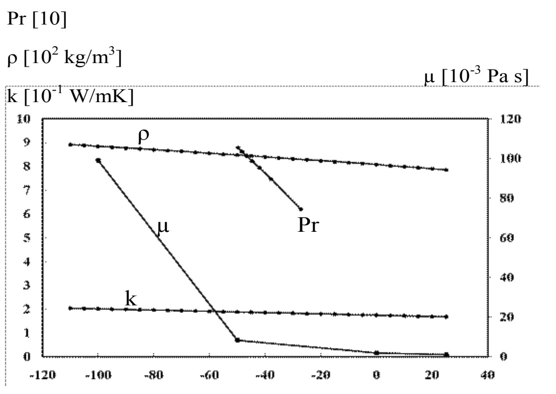


Figure 4. Thermophysical properties of EtOH and the Pr dimensionless number at different temperatures.

and ν the kinematic viscosity.

Pr is the Prandtl number $Pr = Cp\mu/k$ where *Cp* is the specific heat, *μ* the viscosity and *k* the thermal conductivity of the cryogenic fluid.

All the thermo-physical properties used in the correlations have to be evaluated at the film temperature $T_f = (Ts+T\infty)/2$.

As the temperature T_s at the cylinder surface changes with time during cooling, a Matlab program was implemented to calculate the thermo-physical properties at T_f as time elapses.

Sparrow and Gregg (25) suggested that natural convection from a vertical cylinder can be treated as a vertical flat plate when the diameter to length ratio meets the criteria:

$$\frac{D}{L} > \left(\frac{35}{Gr_L^{0.25}}\right) \quad (6)$$

However, Eq. 6 was not satisfied by the three tested cylinders; therefore a correction factor considering the effect of the cylinder curvature proposed by Cebeci (6) was included (18). In spite of using the correction factor for vertical cylinders there was a lack of agreement between these correlations and the experimental heat transfer coefficients, since the correlation for vertical tubes (Eq. 4) is strongly dependent on the height of the cylinders and show dependence on the diameter, as was noted in the *h* values obtained in the present work.

Considering that the cryogenic fluid surrounding cylinders is not completely stagnant due to CO2 bubbling (sublimation of dry ice in a viscous ethanol bath), the correlation to calculate external *h* coefficients around cylinders under forced convection was applied (18, 19).

$$Nu_D = 0.3 + \frac{0.62 * Re^{1/5} * Pr^{1/3}}{\left[1 + \left(\frac{0.4}{Pr}\right)^{2/3}\right]^{1/4}} \quad (7)$$

where *Re* is the Reynolds number (*Re=ρVD/μ*), with ρ is density of ethanol and *V* the fluid velocity around cylinders. The equation is valid for low *Re* numbers.

The characteristic velocity of surrounding fluid used in the calculation of the *Re* number, was assumed to be in the range 1-5 cm/s since it is highly influenced by the presence of CO_2 bubbles that detach and rise to the surface producing a mild mixing effect in the cryogenic fluid. These assumptions led to the best results in terms of agreement between experimental and literature correlations and they confirm that the cryogenic bath cannot be considered a stagnant fluid. This conclusion is based on the calculations carried out considering *Nu*=0.3 (the asymptotic value of the correlation when *Re*→0); in this case *h* values from the correlations were lower than experimental coefficients for three cylinders.

Fig 4 shows the properties of the cryogenic bath and the Prandtl number as functions of temperature (8, 15). The information is very important because of the marked influence of temperature on the thermal properties which affect the dimensionless numbers.

CONCLUSION

The numerical model allowed to determine the heat transfer coefficients for the prediction of cooling rates of samples immersed in dry-ice ethanol cooling bath (-78ºC). The obtained *h* values represent the thermal and fluid dynamic phenomena occurring at the interface of the system in terms of heat flow exchanged between the dry-ice ethanol and the cooled objects.

An inverse lineal relationship between *h* and cylinder diameter was observed. Literature correlations for forced convection at low Reynolds numbers gave the best fit with the *h* values derived from experimental measurements and numerical predictions for the estimation of *h* in vertical cylinders. Cryogenic vials that store biological material, human or animal cells, have diameters which are in the range of those reported in the present work.

The *h* values calculated are useful when predicting cooling rates of cylinder shaped objects in an ethanol-dry ice cryogenic bath. This knowledge is important for the mathematical modelling of heat transfer in the cryo-technology field.

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