Modeling, simulation and test of the heat flow in a liquid nitrogen cryostat for Mössbauer spectroscopy

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Abstract. In this work, we present a heat flow analysis for a conceptual design of an open cycle liquid nitrogen cryostat with application in Mössbauer spectroscopy. Initially, we made a study of the different configurations of Dewar type cryostats reported in the literature. These configurations are based on existing designs and the Mössbauer spectrometer requirements of the Instrumentation and Spectroscopy Laboratory at Universidad EAFIT in Medellín. From this study, we establish our own conceptual design. A first heat flow analysis in stationary state was calculated to obtain the dimensions of the reservoir vessel. The heat flow model obtained predicted an adequate evaporation time of the liquid nitrogen for the acquisition of Mössbauer spectra. For this model we employed the Fourier’s law, the heat flow by radiation in grey bodies and the properties of 304 stainless steel, 6061 aluminium alloy and OFHC copper. In consequence, the mechanical design of the cryostat was defined and the new heat loads associated with it were added to the initial model. Finally, the prototype was built, the model was tested and some final corrections were made.

1. Introduction

Since the beginning of Mössbauer spectroscopy, cryostats have been used to lower the temperature of the sample, to increase the probability of having Mössbauer effect; and to determine, among other parameters, the temperature of transition from a paramagnetic to a magnetically ordered state [1][2]. The Mössbauer spectrometer developed in the Instrumentation and Spectroscopy Laboratory at Universidad EAFIT [3][4] in Medellín, allows us to analyze iron containing samples, which can present paramagnetic or superparamagnetic behavior at room temperature. Therefore, it is our objective to develop a low cost and good performance cryostat to study the magnetic and structural properties of the iron containing samples at low temperatures. Looking for a good understanding of their magnetic relaxation mechanisms as well as variations in their structural and electrical properties at temperatures below the room temperature.

A cryostat is a system that allows keeping the sample at a low and stable temperature. Many cryostats have been built in laboratories [5][6] by following the schemes of commercial cryostats [7] or by making adaptations of existent systems [8][9]. Although these systems satisfy the requirements of the users, they could be improved by designing a system that considers since the beginning the heat flows and heat loads into the cryostat as it is done for some specific cases [10][11][12]. We went through the above mentioned process and evaluated our proposed design to ensure that it will perform as expected.

In this work, the evaporation of liquid nitrogen method was chosen to reduce the temperature of the
sample, given that nitrogen evaporation temperature is enough to provide the desired temperature range for Mössbauer spectroscopy applications [13]. Cryostats have different configurations [14][7]. Open cycle cryostats are based on the Dewar vessel and are considered open because after the cryogen evaporation it cannot be recovered. In this configuration, the sample can be immersed in the liquefied gas or attached to a cold finger that has contact with the liquefied gas. For some analyses and measurements, a termed-tail vessel extension is added, which can have windows to carry out radiation experiments [7]. In other configurations, the evaporated gas is used to cool the sample.

The proposed design for an open cycle cryostat consists of three cylindrical coaxial vessels. The inner vessel works as the liquid nitrogen reservoir and is made of 304 stainless steel. Furthermore, the middle vessel is also made of 304 stainless steel and operates as a vacuum jacket for the reservoir. Finally, the outer vessel is made of 6061 aluminum alloy. This provides a vacuum jacket to the sample and the cold finger, and holds a tail with two windows for the entry and exit of the radiation coming from the radioactive source of the Mössbauer spectrometer. Figure 1 presents some schemes of the cryostats aforementioned.

Figure 1. Schemes of the open cycle cryostats with a) immersed sample and tail with windows [7], b) cold finger and tail with windows [7] c) gas immersed sample and the [7] d) proposed design.

2. Heat flow analysis

For designing a cryostat, it is important previously to analyse its heat loads to ensure that the system will perform as expected. There are many causes of heat leaks into a cryostat. Several of these causes are related to the cryostat’s structure, such as, conductive heat flow through the walls of the vessels and supports, heat flow by radiation between surfaces at different temperatures, and the convective heat flow due to the residual gas in the cavities between the vessels [15]. Therefore, the design, dimensions, and selection of materials of the cryostat must be based on a heat flow analysis.

In the proposed design, the liquid nitrogen in the reservoir vessel is the only heat sink to compensate all the heat flows and heat loads of the cryostat and its evaporation time is limited by the evaporation rate due to the heat flow, then the inner vessel was chosen to be the design starting point.

2.1 Heat flow analysis to obtain the dimensions of the inner vessel

Considering that low pressure in the vacuum jacket reduces significantly the heat flow by convection, this initial analysis will consider only the heat flow by conduction through the vessel walls and the radiation heat flow on its lateral surface.

To calculate the heat flow by conduction we used the Fourier’s law for unidirectional heat flow, this equation can be written as follows: [16]
\[ \dot{Q} = \frac{A}{L} \int_{T_1}^{T_2} K(T) dT \]  

(1)

Where \( \dot{Q} \) is the heat flow in Watts, \( A \) is the cross section area to the heat flow, \( L \) is the length between the points at temperatures \( T_1=77 \text{ K} \) and \( T_2=300 \text{ K} \) and \( K(T) \) is the thermal conductivity of the material as a function of temperature.

According to equation (1) there are three variables to be considered that decrease the heat flow; these are cross section area of the vessel \( A \), distance \( L \) between the vessel opening at 300 K and the liquid nitrogen surface at 77 K, and the thermal conductivity of the vessel material. As it is mentioned by different authors [7][17], 304 stainless steel is used in cryogenics due to its good mechanical properties and for its low thermal conductivity. The coefficients for calculating thermal conductivity in the range of 77 K to 300 K were taken from the NIST studies. The thermal conductivity for a specific temperature \( T \) is calculated through the following expression [18]:

\[
K = 10^a \\
\quad a = -1.4087 + 1.3982(\log_{10} T) + 0.2543(\log_{10} T)^2 - 0.6260(\log_{10} T)^3 + 0.2334(\log_{10} T)^4 + 0.4256(\log_{10} T)^5 - 0.4658(\log_{10} T)^6 + 0.165(\log_{10} T)^7 - 0.0199(\log_{10} T)^8 
\]

(2)

The integral of expression (2) has no analytical solution, then the thermal conductivity was numerically calculated through simulations using MATLAB. In the calculations the area \( A \) and length \( L \) were replaced as arrays of values to evaluate the conductive heat flow for various dimensions simultaneously. The range studied for the depth \( L \) of the liquid nitrogen measured from the opening of the vessel is between 0-80 cm and the range studied for the thickness \( t \) of the vessel wall is between 1-5 mm. The general results appear in figure 2.c.

As observed in figure 2, the conductive heat flow increases one order of magnitude with the variation of thickness \( t \) of the cryogenic vessel wall of the cryogenic vessel and decreases as the liquid nitrogen depth \( L \) increases.
Afterward, we calculated the radiative heat flow by applying the gray bodies’ equation for cylindrical vessels, and later we made the correction for the heat flow direction; resulting in the following expression [16]:

\[
\dot{Q}_{i\rightarrow e} = \frac{\sigma A_i (T_e^4 - T_i^4)}{\varepsilon_i + \frac{A_i}{A_e} (\frac{1}{\varepsilon_e} - 1)} + \frac{A_e A_i}{A_e A_i} - 1
\]  

(3)

Where the \(i\) subscript indicates that the value belongs to the inner vessel and the \(e\) subscript refers to the outer vessel, \(A\) indicates the lateral surface, \(\varepsilon\) is the emissivity of the material [16][17] and \(\sigma\) is the Stefan Boltzmann constant (5.6704 × 10^{-8} \text{ W K}^{-4} \text{ m}^{-2}). There are two main contributions to the radiation heat flow. First is the vessel surface temperature, this surface surrounds the liquid nitrogen column, in consequence the surface temperature remains constant and the term \(T_e\) is equals to 77 K. The second contribution is given by the conductive portion of the vessel, in which the term \(T_e\) is assumed to change by following a linear temperature profile between 77 K and 300 K, and it is dependent on the analyzed portion height. The radiation heat flow variation with the liquid nitrogen level, after adding the two contributions mentioned, is shown in figure 3:

![Figure 3](image)

Figure 3. Curve of the radiation heat flow emitted by the middle vessel and absorbed in the outer lateral surface of the inner vessel, as a function of the level of liquid nitrogen.

The radiation heat flow graph is a one-dimensional graph due to the dependence of the radiation heat flow only on the surface area where it insides; it changes with the liquid nitrogen height column. As observed in the graph, the higher the liquid nitrogen column higher the radiation heat flow. This is due to the greater contribution of the radiation heat flow on the surface surrounding the column at constant temperature.

The convective heat flow between the room temperature and the vessel opening was neglected because the gaseous nitrogen at lower temperature being released from the vessel counteracts the heat transference between the warm air and the vessel walls.

To obtain the liquid nitrogen column evaporation time, the column was split in disks of equal volume. Then, to calculate the column’s evaporation time, we compute the individual evaporation times of these disks and add them together. The time of evaporation for each disk is the quotient of heat of vaporization of the volume of the disk by the sum of the heat flows obtained for the height of the analysed disk. Knowing that each disk has approximately 45 ml and that the liquid nitrogen latent heat of vaporization is 161 J/ml, then the energy required to evaporate each disk is 7293.3 J. The graph in Figure 4 shows that the maximum evaporation time of 32.8 h is obtained for \(L= 1 \text{ cm}\), \(r = 1 \text{ mm}\) and \(r = 0.038 \text{ m}\). The maximum evaporation times for various radii are presented in Table 1.
Figure 4. Graph of the evaporation time as a function of the liquid nitrogen level and the wall thickness of the cryogenic vessel.

Table 1. Evaporation time of the liquid nitrogen for various radiuses and thicknesses of the cryogenic vessel for a length L of 80 cm.

| Radius (m) | Evaporation time at 1 mm thick (h) | Evaporation time at 2 mm thick (h) | Evaporation time at 3 mm thick (h) |
|------------|-----------------------------------|-----------------------------------|-----------------------------------|
| 0.025      | 21.53                             | 15.34                             | 12.09                             |
| 0.03       | 25.86                             | 18.47                             | 14.59                             |
| 0.04       | 34.54                             | 24.73                             | 19.59                             |
| 0.05       | 43.22                             | 30.99                             | 24.59                             |
| 0.06       | 51.91                             | 37.25                             | 29.6                              |
| 0.07       | 60.59                             | 43.51                             | 34.6                              |
| 0.08       | 69.27                             | 49.78                             | 39.6                              |

Table 1 shows that as the radius of the vessel increases, the evaporation time increases. However, we could not choose a vessel too wide since the vessel’s radius determines the radius of the external vessels used as vacuum jackets. Since the external vessels need to be adjusted to commercially available values with a convenient separation between vessels, we selected a radius of 0.04 m for the cryogenic vessel. This ensures an adequate evaporation time and allows us to obtain cylinders with radii larger than commercially available devices for the outer vessels used as vacuum jackets.

The results obtained from this analysis were useful for doing a research on the commercially available pipes and materials to build the vessels. Based on the researched supplies, we made a mechanical design and we added new heat flows and heat loads to the model.

2.2 Heat flow in the cryostat

The final design, which is based on the conceptual design and the geometries of some commercially available supplies, counts with a middle vessel, a tail with two windows placed face to face, among other features that are presented in Figure 5. The new heat loads added to the cryogenic vessel had to be calculated and added to the initial heat flows to determine the final evaporation time of the liquid nitrogen.

The new heat loads and their respective values are the conduction heat flow through the middle vessel of 3.961 W, the radiation heat flow in the cold finger 0.159 W, and 77.7 kJ that must be extracted from the cold finger to lower the temperature of the sample from 298 K to 77 K. These 77.7 kJ of energy
correspond to 482 ml of liquid nitrogen or 10.7 cm of the liquid nitrogen column in the inner vessel; then the minimum level of liquid nitrogen in the inner vessel is 10 cm.

The heat required to reduce the temperature of the cold finger was calculated by using the following expression [17]:

$$Q = m \int_{T_1}^{T_2} c(T) dT$$  \hspace{1cm} (4)

Where $Q$ is the heat extracted from the copper cold finger in J, $m$ is the mass of the cold finger and $C(T)$ [18] is the specific heat function dependent on the temperature in the range between $T_1 = 77$ K and $T_2 = 298$ K.

Due to the similarity of the function of the specific heat of the OFHC copper to the thermal conductivity of the 304 stainless steel, it was calculated by numerical procedures in MATLAB.

The new evaporation time for each liquid nitrogen level was obtained by the same procedure used before; but in this occasion, the heat flows were modified for the specific case of $r = 0.038$ m and $t = 1.8$ mm, the radiation heat flow in the cold finger and the conduction heat flow through the middle vessel were also included. Finally, the chart obtained was displaced to reduce the 482 ml consumed to cool down the cold finger to 77 K.
According to the graph shown in Figure 6(b) and the results presented in Table 1 and Figure 4, the final time of evaporation (~15 h) is about half of the time obtained considering only the heat flows in the inner vessel (~33 h). One of the main sources of this effect is the heat by conduction through the middle vessel, which is very close to the dominant heat flow: the radiative heat flow.

After prototype was built, we performed some measurements of the liquid nitrogen evaporation time, by using a 30 cm liquid nitrogen level and two vacuum pumps of references Duoline Rotary vane pump from Pfeiffer and a Turbomolecular pump from Pfeiffer. Under these conditions, we obtained an average evaporation time of 6.5 h, due to the changes of the evaporation caused by the measurement of the liquid nitrogen level. According to the figure, 6(b) an evaporation time approximate of 7 h would be expected. The discrepancy observed in the calculated and measured evaporation time is explained because the pressure inside the cavity between the cryogenic vessel and the jackets was not low enough (~10^{-2} Torr), being necessary at least a value of the order of 10^{-4} Torr to disregard the heat flow by convection. This is caused by the limitations of the vacuum pumps and the volume of the cavities. Another condition necessary to reach better results, similar to the ones obtained by the model is to provide low pressure, around 10^{-4} Torr, in both vacuum jackets, the one for the inner vessel and the other for the cold finger and the sample.

3. Conclusions and future work

We performed a heat flow model to determine the dimensions and evaporation time of liquid nitrogen in an open cycle cryostat with concentric cylindrical vessels, resulting in parameters that we could obtain commercially. We simulated and analyzed the design of the cryostat, finding that the model predicts an evaporation time of 15 hours for an 80 cm liquid nitrogen level. This time is adequate for the acquisition of Mössbauer spectra by programming periodical refills of the cryogenic vessel. The evaporation time predicted by the model by using a level of 30 cm of liquid nitrogen was 93% in agreement with the evaporation time measured experimentally.

The tests carried out revealed that the model needs the addition of heat flows by convection when the pressure reached in the vacuum jackets is not low enough, and more heat flows need to be added if electric components such as sensors, heaters or magnetic sources are added to the system, due to the thermal loads these introduce.

References
[1] J van Lieron and D H Ryan, 2002, Physical Review B, vol 65, pp. 104402-1, 104402-9.
[2] A A Velásquez and J P Uriquijo, 2015, Hyperfine Interactions, vol 232, pp. 97-110.
[3] A A Velásquez, A Carmona, D Velásquez and L Ángel, 2011, Hyperfine Interactions, vol 202, pp. 63-71
[4] A A Velásquez and M. Arroyave, 2014, Hyperfine Interactions, vol 224, pp. 65-72.
[5] M Shinohara, A Ishigaki and Kazuo Ono, 1968, Japanese Journal of Appl. Physics, vol 7, pp 170-173.
[6] W Wiedmann, W A Mundt and D Kullmann, 1965, Cryogenics, pp 94-100.
[7] M N Jirmanus and Janis Research Company, Introduction to laboratory cryogenics, (Willmington, Janis) pp 5-37.
[8] A Janoshka, G Svenconis and Schüinemann, 2010, Journal of Physics: Conference Series, vol 217, pp 1-4.
[9] P Novak, J Pechousek, O Malina, J Navarik and L Machala, 2014, AIP Conf. Proc., vol 1622, 67, pp 67-71.
[10] V Parma, 2013, Cryostat Design, Proc. of the CAS-CERN Accelerator School: Superconductivity for Accelerators, vol. 005, pp 353-399.
[11] I Sekachev, M Meekins, C Sborchia, G Vitupier, H Xie and C Zhou, 2014, Physics Procedia, vol 67, pp 294-301.
[12] F Bai, X Niu, X Wang, J Hu, Y Hu and Y Zhang, 2018, Cryogenics, vol 95, pp 29-35.
[13] F Pobell, 2007, Matter and Methods at Low Temperatures, (Hidelberg, Springer) pp. 1-6 and 33-
40.

[14] N H Balshaw, 2001, *Practical Cryogenics*, (Oxon England, Oxford Instruments Suérconductivity Limited) pp. 21-30.

[15] G K White, 1979, *Experimental techniques in low temperature physics* (Oxford, Clarendon Press) pp. 129-149 and 171-174.

[16] O Levensielp, 1993, *Engineering Flow and Heat Exchange*, (New York Plenum Press) pp. 161-165 and 178-183.

[17] P Lebrun, 2007, *An Introduction to Cryogenics*, (Geneva, CERN) pp 8-11.

[18] E D Marquardt, J P Le, and R Radebaugh, 2011, Cryogenic Material Properties Database, *11th International Cryocooler Conf. Proc.*

**Annexes**

Script in MATLAB all the calculations presented, this script can be copied and pasted to obtain the graphs shown through the article.

%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
%% Program for the proceeding of VI CNIF del cryostat BOREAS
%% NATALIA GUTIERREZ ANDRADE and ALVARO ANDRES VELASQUEZ TORRES
%% UNIVERSIDAD EAFIT
%% TESI FÍSICA 2018
%% VARIABLES: R(RADIUS VESSEL IN METERS), T(TEMPERATURE KELVIN),
%% K1(THERMAL CONDUCTION OF SS 304)
%% L(LENGTH OF THE VESSEL METERS), Q(CONDUCTIVE HEAT FLUX %
%% WATT),CN(EVAPORATION HEAT OF LIQUID NITROGEN J)
%% Q2(RADIATION HEAT FLUX WATT) QT(TOTAL HEAT FLUX)
%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%%
clc,clear;
R=0.038;\Comment{INTERNAL RADIUS OF THE INNER VESSEL}
T=[77:1:300];\Comment{TEMPERATURE VECTOR}
\Comment{Thermal Conductivity of 304 Stainless Steel from NIST}
K1=[-1.4087 1.3982 0.2543 -0.626 0.2334 0.4256 -0.4658 0.165 -0.0199];
\Comment{COEFFICIENTS TO CALCULATE THE THERMAL CONDUCTIVITY}
K=zeros(1,224);
for i=0:1:223
  \Comment{for j=0:1:8}
  K(1,i+1)=10^(-1.4087+1.3982*(log10(T(1,(i+1))))+0.2543*(log10(T(1,(i+1)))^2)-
  0.626*(log10(T(1,(i+1))))^3)+0.2334*(log10(T(1,(i+1)))^4)+0.4256*(log
  10(T(1,(i+1)))^5)-
  0.4658*(log10(T(1,(i+1)))^6)+0.165*(log10(T(1,(i+1)))^7)-
  0.0199*(log10(T(1,(i+1)))^8));\Comment{condutividad térmica por}\n  temperatura\n\Comment{end}
end
figure (1)
plot(T,K),xlabel('Temperature [K]'),ylabel('Thermal conductivity [J/m*K]')
Kt=0;
for i=1:1:223
    Kt=K(1,i)+Kt; % TOTAL THERMAL CONDUCTIVITY AFTER THE INTEGRAL
end
%% CONDUCTIVE HEAT FLUX FOR 304 SS VESSEL
L=[0.01:0.01:0.80];% LENGTHS VECTOR
A=[0.001:0.0001:0.005]; %THICKNESS VECTORS
[Q,CN]=meshgrid(A,L);
B=R+A;

for i=1:1:size(L,2)
    for j=1:1:size(A,2)
        Q(i,j)=((B(1,j)^2)-(R^2))*pi*Kt/L(1,i); % CONDUCTIVE HEAT FLUX W
    end
end
figure(2)
surf(A,L,Q),xlabel('Thickness t [m]'),ylabel('Length L [m]'),
zlabel('Conductive Heat Flow [W]')

%% eVAPORATION ENERGY PER VOLUME OF LIQUID NITROGEN
for i=1:1:size(L,2)
    for j=1:1:size(A,2)
        CN(i,j)=R^2*pi*(0.8-L(1,i))*1000*161000+1; %VOLUME OF LIQUID NITROGEN FOR AN SPECIFIC L TIMES THE ENERGY PER mL
    end
end
figure (3)
surf(B,L,CN),xlabel('THICKNESS [m]'),ylabel('LENGTH [m]'),
zlabel('EVAPORATION ENERGY [J]')

%% OFHC COPPER SPECIFIC HEAT
ct=0;
for i=0:1:223
    for j=0:1:8
        K(1,i+1)=10^(-1.911844-0.15973*(log10(T(1,(i+1))))+8.61013*(log10(T(1,(i+1)))^2)-18.996*(log10(T(1,(i+1)))^3)+21.9661*(log10(T(1,(i+1)))^4)-12.7328*(log10(T(1,(i+1)))^5)+3.54322*(log10(T(1,(i+1)))^6)-0.3797*(log10(T(1,(i+1)))^7));
    end
end
ct=0;
for i=1:1:224
    ct=K(1,i)+Kt; %SPECIFIC HEAT IN THE RANGE FROM 77K TO 300K OF COPPER OFHC
end

%% RADIATION HEAT FLUX
A1=2*pi*R*(0.80-L);
A2=2*pi*(2*R)*L;
Q2=zeros(1,size(L,2));
for i=1:1:size(L,2)
    Q2(1,i)=(5.670e-8*A1(1,i)*(300^4-77^4))/((1/0.3)+(A1(1,i)/A2(1,80))*(1/0.061));
end
Q3=zeros(size(Q));
for i=1:1:size(Q3,1)
    Q3(i,:)=Q2(1,i);
end

figure (4)
plot(L,Q2),xlabel('LEVEL OF LIQUID NITROGEN [m]'), ylabel('RADIATION HEAT FLUX [W]')

%% ADDITION OF HEAT FLOWS
Q4=Q3+Q;
figure (5)
surf(A,L,Q4),xlabel('TICKNESS [mm]'),ylabel('LEVEL OF LIQUID NITROGEN [m]'), zlabel('TOTAL HEAT FLUX [W]')
figure (6)
surf(A,L,I2),xlabel('thickness t [m]'), ylabel('Length L [m]'), zlabel('time [h]')
figure (7)
I2=CN(79,1)./Q4;%CALCULATION OF STORAGE TIME FOR EACH LEVEL OF LIQUID NITROGEN
surf(A,L,I2),xlabel('TICKNESS [mm]'), ylabel('LEVEL OF LIQUID NITROGEN [m]'), zlabel('TIME [SECONDS]')
figure (8)
I3=I2./3600;
surf(A,L,I3),xlabel('TICKNESS [mm]'), ylabel('LEVEL OF LIQUID NITROGEN [m]'), zlabel('TIME [HOURS]')
I4=I3;
for j=1:1:41
    for i=1:1:79
        I4(80-i,j)=(I3(81-i,j)+I4(80-i+1,j));
    end
end
figure(9)
surf(A,L,I4),xlabel('thickness t [m]'), ylabel('Length L [m]'), zlabel('time [h]')
h = rotate3d;
h.Enable = 'on';
figure (10)
contour(A,L,I4),xlabel('Espesor [mm]'), ylabel('Longitud [m]'), zlabel('tiempo [h]')