Final Report: A Calorimeter for Neutron Flux Measurement

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Abstract

A calorimeter for absolute neutron flux measurement has been built and tested. The calorimeter measures the heat produced in a 10°K thick LiPb target when neutrons are captured via the ${}^6\text{Li}(n, {}^3\text{H}){}^4\text{He}$ reaction. The sensitivity achieved was 1.3×10^6 n/s for a 1 hour measurement. Separate flux measurements with the calorimeter and a ${}^{238}\text{U}$ fission chamber are in agreement and show that systematic errors are less than 3%. An improved calorimeter has been built which is sensitive to 10^5 n/s for a 1 hour measurement.

1 Introduction

Several techniques have been employed for neutron flux detection. Black detectors measure the entire flux. Thin, $1/v$ detectors that measure a fraction of the flux with efficiency proportional to the time that the neutrons spend in the fiducial volume, for example one in which a decay can be detected. A technique for absolute calibration of these different methods of flux measurement and neutron beam measurements employing such an absolute flux measurement will improve the precision of flux detection. In this report, we describe the performance of the Harvard/LANL calorimeter built to demonstrate that a precise absolute measurement of neutron flux can be achieved using calorimetry of the heat produced when neutrons are absorbed via the ${}^6\text{Li}(n, {}^3\text{H}){}^4\text{He}$ reaction.

The design goal of the calorimeter is to make an absolute determination of a neutron flux that is itself on the order of 10^8 n/s to an accuracy of 0.1% or better. This requires an absolute sensitivity of 10^5 n/s. The exothermic reaction

has a large thermal cross section (940 b), with few competing processes and only the ground-state exit channel energetically allowed. The Q value corresponds to 7.6615×10^{-13} J/n or $76.615 \mu\text{W}$ for 10^8 n/s. The required absolute sensitivity goal of the calorimeter is therefore 77 nW.

2 The Calorimeter

The calorimeter, described in detail in this section, is shown in Figure 1. Cooling for the calorimeter is provided by a closed cycle He refrigerator. Two radiation shields are maintained at 40K (outer shield) and 8K (inner shield). A temperature controlled reference heat bath is incorporated into the inner shield as the lid of the cylindrical can. The target containing ${}^6\text{Li}$ is suspended within the inner shield.

2.1 The Target

The ideal target has a high fraction of ${}^6\text{Li}$, but pure ${}^6\text{Li}$ is not suitable due to a low temperature phase transition. It is therefore necessary to use a material consisting of a mixture of ${}^6\text{Li}$ and other elements. The properties of this material that we seek are: 1) that the remaining target components absorb neutrons negligibly, 2) that it has a high thermal conductivity, 3) that it has a low specific heat at the operating temperature, and 4) that it is able to absorb as heat the energy the triton (${}^3\text{H}$) and alpha particle (${}^4\text{He}$). LiPb metal is a good choice. The material is easy to machine and safe to handle. It does, however, slowly degrade in air, turning yellow and brittle after several months.

We started with a disk of LiPb, 5.715 cm diameter disk and 0.635 cm thick. The target was faced off, and a groove was machined around the outside edge, and for heater wire to be wound into the groove. Thermal contact of the heater wire was improved with silicone grease. Grease allows for changes to be made more easily than, for example, epoxy. Finally, 0.0127 cm of pure aluminium foil was wrapped around the front of the target and secured to the back using more grease and six aluminium screws. The purpose of the aluminum is to contain a recoil ${}^3\text{H}$ or ${}^4\text{He}$ that would otherwise escape from the front of the target.

The absorption length for thermal neutrons in LiPb is 0.025 cm, whereas the range for the triton (of energy 2.7MeV) is 0.004 cm. The minimum thickness of aluminium that would be acceptable is 0.0064 cm. An alternative target assembly consists of a box of pure Si loaded He gas for heat transfer from the target.

The Pb and contaminantns can also capture neutrons in reactions producing different amounts of heat. Natural lead (used in target preparation) contains four isotopes, listed below along with the natural abundance and thermal neutron capture cross section:

- ^{204}Pb (1.42%, $\sigma_c = 0.66$ b),
- ^{206}Pb (24.1%, $\sigma_c = 0.03$ b),
- ^{207}Pb (22.1%, $\sigma_c = 0.71$ b) and
- ^{208}Pb (52.3%, $\sigma_c = 0.0005$ b).

There is also an unstable isotope: ^{205}Pb ($t_{1/2} = 1.4 \times 10^7$ y , $\sigma_c = 3.8$ b). that is produced in negligible quantities over the target's lifetime. Since lead is a good gamma-ray absorber, the target will absorb energy from gamma-rays that contaminate the neutron beam. This is a problem, even when the neutron beam is chopped because the neutron shutter may also attenuate the gammas. The best way to avoid such a problem is to condition the neutron beam in such a way as to avoid gamma contamination entirely.

The effect of the aluminium in the path of the neutron beam is negligible since aluminium captures very few neutrons, ($\sigma_c = 0.23$ b), making it a good material for neutron entrance windows. It does, however, become activated by the reaction

Aluminium of thickness of 0.0127 cm (5 mil) absorbs 0.02% of the neutrons. Each capture is accompanied by the emission of a gamma, and a beta. The contribution due to this process is less than 0.02%. Other materials present in the target (manganin, copper, and conductive grease) are kept out of the way of the neutron beam and will not be activated.

2.2 Temperature Control

The calorimeter employs two thermal control systems, one to maintain a reference heat bath (the lid of the inner radiation shield) at 8K, and one to control the target at 10K. We measure the change in the feedback power to the target when neutrons are incident on the target.

The beam-off feedback power minus the beam-on feedback power is the power produced by the ${}^6\text{Li}$ reaction.

We measure the feedback power by monitoring the heater voltage and current. The heat dissipated in the leads to the heater wire introduces a systematic error since an unknown fraction of that heat is absorbed by the target. This error is no larger than the ratio of the resistance of the leads to that of the heater, and it is likely to be much less since we measure the change in power needed to maintain the target's temperature. In the present design this is 0.7%. The design can be improved by lowering the relative electrical resistance of the leads.

Thermometry is accomplished through semi-conductor diodes placed in thermal contact with various parts of the heat bath and target. When forward biased, the voltage across these diodes increases with decreasing temperature. Each diode is biased by a $10\text{ }\mu\text{A}$ constant current source. At 8K, the diodes have approximately a 2V drop, and change 50mV per degree. Each diode therefore supplies $20\text{ }\mu\text{W}$ of the heat to the target. In order that the diodes themselves are not a source of error, the current supplies must be stable to 0.01%. In order to measure the temperatures to 1mK, we must measure the diode voltages to $50\text{ }\mu\text{V}$ (i.e. 25 ppm). The voltage measurement is accomplished with a DVM interfaced to a micro-computer and has a resolution of only 75ppm. Since we average many readings, our effective resolution is improved.

The goal of the design is to isolate the target from all fluctuating sources of heat. The target is therefore surrounded by two radiation shields, the inner one at 8K, and the outer one at 45K. The volume between the shields is evacuated. The inner radiation shield consists of an aluminium can bolted to the "cold finger" of the CTI closed-cycle helium refrigerator. The lid of the can serves as the reference heatbath and is held at 8°K by a Palm Beach model 4025 thermometer/controller. A heater wire is wound onto the can just below the lid. The "heatbath control" diode is placed at the center of the lid, near to where the "weak link" is connected. A second diode (the "heatbath sense" diode) is used to measure the temperature at a different point on the lid. The temperature fluctuations of the reference heat bath are described by white noise of $1\text{mK}/\sqrt{\text{Hz}}$.

The prototype design suffered from the effects of thermal fluctuations of the inner shield. These arise from the thermal noise of the cold finger itself: it contains a mechanical piston that produces a constant 2.7 Hz thermal oscillation. Another serious problem is the variation of the cooling power of the refrigerator as the temperature of the cooling water changes. The heat bath is not affected by this slow wandering; it is held with sufficient stability at 8K by the temperature control loop. The base of the can that changes slowly, varying as much as a degree. The can will radiate onto the target according to the Stefan-Boltzmann law with power variations due to temperature change of ΔT of

$$\Delta P_R = 4\sigma_B \epsilon A T^3 (\Delta T) \quad (3)$$

For our target, the varying power in μW is given by

$$\Delta P_R = 0.8(\Delta T) \quad (4)$$

where ΔT is in degrees Kelvin. From this we can see the importance of controlling the entire inner shield (not just the lid) with effective variations of 1 mK or less.

The target is suspended from an aluminum bracket attached to the reference heat bath by nylon threads. The thermal connection from the target to the heat bath is through a "weak link." All leads to the target (including the target heater leads) are thermally connected ("lagged") to the reference heatbath as well. The weak link is manganin. The weak link is formed of 30 parallel lengths of 0.032 cm wire each 3 cm long. The thermal resistance is 8711 $^\circ\text{K}/\text{W}$. Knowing this we can measure the thermal time constant of the target/weak link system, thus inferring the heat capacity of the target itself. At 8K we have measured the LiPbto have a specific heat of 0.008 +/- .001 J/K-g.

Target temperature control was accomplished with a Lake Shore temperature controller. The ideal target set point requires the feedback loop to supply just over the maximum power one expects from the neutrons. Running at a higher temperature increases the target's heat capacity which is proportional to T^3 at these temperatures. The target temperature was controlled to 1 mK, and has a theoretical sensitivity (assuming perfect thermometry) of 0.1mW.

3 The NIST Calorimeter Test

The calorimeter was tested with a neutron beam at the reactor facility of the National Institute of Standards and Technology, NIST in Gaithersburg, Maryland. A schematic of the set up is shown in Figure 2. The neutron beam was collimated by a pair of ^6LiF apertures separated by 60 cm as shown. A ^6LiF shutter was used to chop the neutron beam.

The main objectives of the test were:

1) To test the calorimeter with calibration heat source. In this way we assign the error due to calorimetry alone. (There could be additional error due to unforeseen nuclear interactions of the beam and target or poor thermalization of the decay products by the target.)

2) To show that the calorimeter was effected by neutrons; show that this effect did not change over time; and show that the response time of the calorimeter to neutrons was consistent with that for calibration input.

3) To compare to another flux determination method, in this case the ^{235}U fission chamber of Gilliam and Lamaze.

We achieved a 3% accuracy for a 1 hour measurement of a 4×10^7 n/s beam.

Figure 3 shows a measurement of the long term stability of the target feedback signal when there is no other heat input. Each point is the average of 3600 one-second measurements of the feedback power. The error bars indicated ($0.75\mu\text{W}$) is the standard deviation of the 3600 measurements that make up each point. The $1\mu\text{W}$ drift that is still present in the feedback signal is related to changes of the cooling power of the refrigerator that cause a change of the temperature of the bottom of the inner shield that is coupled to the target either through radiation or through the residual gas.

To test the ability of the calorimeter to measure heat and to assign an error, several calibration runs have been made. A second heater wire is wound onto the target and monitored by a circuit similar to the one used to measure feedback power. Using this heater, one can add a measured amount of heat to the target, thus simulating the effect of the neutrons. The data for one calibration run is shown in Figure 4. Each point shown is the difference of two 1 hour averages, one with no input, one with an input of $13.75\mu\text{W}$. The average of the ten pairs of heater on - heater off runs is $13.63 \pm 0.1\mu\text{W}$ where the assigned error is the standard deviation of the ten data points. Using a similar data set (not shown), we determined that chopping the beam with a period of 1 hour (as described below) yields an error per cycle of $1\mu\text{W}$.

Figure 5 shows the power spectrum of the feedback signal for a static eight hour period. Each data point is the average of the feedback signal for one minute. The data set was smoothed by averaging pairs of points, limiting the bandwidth to 8.33 mHz so that the data could be Fourier transformed without aliasing and the power spectrum determined. The $1/f$ noise is plainly visible in the data, and an optimum chopping frequency can be chosen that is higher than the corner of the $1/f$ contribution.

The effect of the chopped neutron beam is shown in figure 6. The value 2.7×10^7 n/s may be inferred from the change in the feedback signal with an error of 2.6%. Each point was originally the average of the feedback signal for one minute but the data were later smoothed with a 10 minute running average to make the effect visible to the eye. This running average does not accurately represent the response time of the calorimeter (less than one minute) and the 10 transition points are not displayed in figure 6.

The reproducibility of the calorimeter was then tested by doing an automated run in which the beam was chopped at a frequency of one cycle per hour. Figure 7 shows the results of this test with a comparison to a single fission chamber measurement performed at the beginning of the run. The fission chamber is assumed to have 5% accuracy and the reactor itself is reported to vary less than 5% peak-to-peak over the course of one day. The

fission chamber measurement was $(4.34 \pm 0.2) \times 10^7$ n/s; the average of all the neutron runs $(4.33 \pm 0.03) \times 10^7$ n/s. The expected statistical error of each 1 hour cycle for the calorimeter data is 0.13×10^7 n/s and the standard deviation of all measurements was 0.17×10^7 n/s. The close agreement of these two figures shows that the systematic errors are smaller than the statistical errors associated with calorimetry.

We also performed a test to see if there were any long time constant transients due to such sources as induced radioactivity. We signal averaged 10 successive runs as shown in figure 8. There are no measurable transients.

4 Improvements

The limitations of the prototype calorimeter developed for the tests described here are thermal stability, heat leakage to the target and the target's heat capacity. We are currently building a calorimeter which addresses these problems by using a liquid He cooled system. There are two advantages provided by liquid He cooling: better stability and lower temperatures. Improved stability comes from the elimination of the variations in cooling power that are present in the closed cycle refrigerator. We have also developed improved temperature control. With these improvements, described in more detail below, a sensitivity of 10^5 n/s should be attainable.

The new ^6Li neutron flux-calorimeter has been built and is currently running at NIST along with a black $^{10}\text{B}(n,\gamma)^4\text{He}$ counting technique. The new calorimeter employs a liquid helium dewar to cool the ^6Li -Pb target to 4.5K. Calorimetry with the astounding sensitivity of 4 nanowatts for each 20 minute measurement has become routine. This sensitivity will allow the calorimeter to be used for comparison to thin detectors at a monochromatic neutron beam. The monochromatic beam has three orders of magnitude lower flux than the white beam at which the black detectors are being tested. The ultimate goal of this work is to establish a convenient standard for neutron flux detection at the 0.1% level with the particular application to measurement of the neutron lifetime.

5 Conclusion

We have built a prototype neutron flux calorimeter that demonstrates the utility of this method of absolute neutron flux determination. Detailed understanding of the limitations of the prototype have led to a design of a liquid He cooled calorimeter and improved temperature control with a sensitivity goal of 10^5 n/s for relatively short measurements (several hours).

This requires calorimetry with sensitivity of 77 nW as well as further consideration of systematic effects not apparent in the tests of our prototype.

Publication

A Calorimeter for Neutron Flux Measurement, J. M. Richardson, T.E. Chupp, R.G.H. Robertson, J.F. Wilkerson, Nucl. Inst. and Meth. in Phys. Res. A306, 291 (1991).

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6 Figures

Figure 1: The calorimeter as it was tested at NIST

Figure 2: Run geometry during the test at NIST.

Figure 3: Long term plot of target feedback signal. Each point is the average of 3600 one-second measurements. The error (0.75mW) is the standard deviation divided by root n.

Figure 4: Results of a calibration run. The points shown are the difference of a one hour measurement with no input followed by a one hour measurement with an calibration input of 13.75mW.

Figure 5: Fourier transform of target feedback signal. The data was first digitally filtered to limit the bandwidth to that shown.

Figure 6: Calorimeter response to a 4 hour exposure of 2.7×10^7 n/sec.

Figure 7: Results of an overnight run. The points shown are the difference of a half hour measurement with no input followed by a half hour measurement with the neutron beam present. The predicted error was 1.3×10^6 n/s. The standard deviation of the above measurements is 1.6×10^6 n/s. The mean value of the above is $4.33 \pm 0.03 \times 10^7$ n/s. The fission chamber value was $4.34 \pm 0.2 \times 10^6$ n/s.

Figure 8: Average of 10 identical runs. By averaging the runs, the noise is reduced allowing us to visually check for transient behavior. None is apparant in the data.

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