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TITLE CALORIMETRY AT 10.6 μm USING ORGANIC VOLUME ABSORBERS

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Calorimetry at 10.6 μm using organic volume absorbers*

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Abstract

Copper-Kapton laminate foils will be utilized as isoperibol calorimeters for pulsed energy measurements at Antares with apertures up to 1.5 m. These are volume absorbing calorimeters in which the radiation is absorbed in a layer tens of wavelengths thick, thereby reducing surface temperature and increasing damage threshold as compared to surface absorbing calorimeters. Kapton was chosen for its high absorption coefficient, 330 cm^{-1} at [illegible] in the 10 μm band at [illegible], and its useful short pulse damage threshold, 2.0 J/cm^2. The copper backing integrates the absorbed heat pulse. The resultant temperature pulse is sensed by [illegible] thermocouples [illegible] bonded to the copper. Time vs voltage traces of the thermocouples can then be analyzed to obtain the energy. [illegible] calorimeters can be fabricated to virtually any size or shape [illegible] techniques [illegible] elements or several in a thermopile [illegible], the [illegible] of this technique.

Introduction

[illegible]

[illegible]

[illegible]

[illegible]

Theory

[illegible]

[illegible]

[illegible]

[illegible] The higher the temperature rise [illegible] the more [illegible] losses.

Choice of Materials for the Absorber

A search was made of various materials. Inorganic solids and crystalline materials were surveyed by Lyman and Schmidt.[1] These have the disadvantage of high cost and difficulty in handling and fabrication. Also, large sizes are prohibitive. Plastic

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absorbers are used commercially.[5] A spectrophotometric survey of plastic materials was done to locate an optimum absorber. The thermal properties of all plastics are approximately the same. The thermal diffusivity, which is the ratio of the thermal conductivity to the product of the density and specific heat, is approximately 10^{-3} cm^{2}sec^{-1} for all plastics. Therefore, the choice of material is made on the basis of the absorption. Table 1 shows some of the materials measured. The values for absorption length are representative but may vary. Also be warned there are at least seven things called "Teflon" and they are all different in their absorption characteristics. The Table shows two different kinds, an FEP and a PTFE. The Kapton we use is DuPont Type V, polyimide.

Table 1. Absorption Coefficients at 10.6 μm

Material	Absorption Coefficient (cm^{-1})
Polyethylene	10
Teflon® PTFE	62
Polystyrene	39
Teflon® FEP	121
Lexan® (Polycarbonate)	130
Mylar® (Polyester)	140
Saran® (Polyvinylidine Chloride)	170
Kapton® (Polyimide type V)	330

An absorbing material should absorb the radiation in many wavelengths. For Kapton, 99% of the energy is absorbed in 15 wavelengths in the material. If the stopping range is much shorter, the damage threshold will drop. For Kapton, the damage threshold was measured to be [illegible] J/cm^2 for a 1 ns, 10.6 μm pulse. The design goal was [illegible] J/cm^2. Table 2 shows some damage threshold data.

Table 2. Damage Threshold Measurements (Courtesy of [illegible] Thomas and Robert Harrison)

Material		Lowest Flux to Damage (J/cm^2)	Highest Flux to Not Damage (J/cm^2)	Mean	Std. Dev.	Number of Shots
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	0.26	[illegible]

[illegible] shows a spectrophotometer scan of the transmission characteristics done on a [illegible] Spectrophotometer with a resolution of [illegible] cm^{-1}. To be sure there was no fine absorption structure in the 10 μm band where the CO_2 laser operates, a more detailed absorption spectrum was run using a [illegible] Fourier Transform Spectrometer with a resolution of [illegible] cm^{-1}. The pressure-broadened line width of the laser [illegible], the output of which will be measured, is [illegible] cm^{-1}. Hence, structure [illegible] is irrelevant.

Table [illegible] presents refractive index and absorption values obtained from the absorption [illegible] for the lines which will run in [illegible]. The greatest thickness of Kapton [illegible] (in of material) of the energy is [illegible] μm. The predominant line is [illegible], for which [illegible] is sufficient. This assumes the metallic copper substrate to which the Kapton is bonded is a 100% reflector. If, in fact, it is an absorber [illegible] the [illegible], the heat is deposited in the Kapton and collected by the copper.

Table [illegible]. Kapton 10 μm Data

Line	Wave No.	Wavelength	Index of Refraction	α (cm^{-1})	1/α (μm)
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

Below are estimates of the various loss mechanisms for a typical copper Kapton laminated calorimeter design. Of course the actual values for the loss mechanism depend on the construction details, but these calculations give first order guidelines.

Radiation. Assuming all the energy is deposited in one absorption length the temperature rise in a very worst case sense may be estimated as

$$\Delta T = \alpha E / \rho c_p$$

where α is the absorption coefficient (330 cm^{-1})

E is the energy flux (Joules cm^{-2}) design goal [illegible] J cm^{-2}

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ρ is the Kapton density (1.42 gm cm^{-3})
C_p is the heat capacitance at constant pressure, (1.092 J $°K^{-1}$ gm^{-1})

This gives a temperature rise of 426°K. Rather high, but it cannot incandesce or flash.

The time it takes the heat pulse to pass into the material is given by

$$\tau = \frac{1}{2\pi^2 K} = 5 \text{ milliseconds} \quad ,$$

where k is the thermal diffusivity (1.004×10^{-3} $cm^2 sec^{-1}$). The reradiation is estimated assuming an emissivity, ϵ, of 1 and that the material radiates at ΔT for 3 into 2π steradians. The ratio, F_1, of the reradiation to incident flux is

$$F_1 = \frac{3\tau\epsilon\,\sigma(T_0 + \Delta T)^4}{\phi} = \frac{20 \text{ mJ/cm}^2}{2 \text{ J/cm}^2} = 0.01 \quad .$$

This is an acceptable level.

Convection

The convection losses may be estimated by considering the heat flux into the air caused by the calorimeters at the elevated temperature, ΔT, in comparison to the incident flux. This is given by

$$F_2 = \sqrt{\frac{\rho' C' k'}{\rho C k}} = 1.23 \times 10^{-4} \quad .$$

where k is the thermal conductivity (watt cm^{-1} $°K^{-1}$) and the primed quantities refer to the values for air.

Conduction. The conduction may be estimated by the heat flux through the thermocouple wires and the polyester supporting threads. A single thermocouple services a collecting area, A. The size varies. A representative integrating area, such as is used in one of the large disc calorimeters, is 3.6 cm^2. At ambient temperatures or slightly above, this will radiate into 2π steradians about 2.2 mW/°K. One pair of thermocouple wires (chromel-constantan), 10 mils in diameter and 13 cm long, could only bleed off 1.7×10^{-5} W/°K of the heat. Thus we can use as a figure of merit for the conduction losses F_3: the ratio of the conduction losses to the radiation losses,

$$F_3 = 7.6 \times 10^{-3} \quad .$$

Clearly, radiation is the dominant cooling mechanism, as desired.

The conductive loss paths could be reduced by a factor of 100 by going to 0.001" wires. However, 0.010" is much easier to work with and wire-wrap techniques may be used. Wire-wrap connections are of special alloy to produce the proper holding power. Consequently, to assure good constantan joints, solder is applied over the wire wrap. Constantan was by far the better choice for this application. The chromel is more spring-like and does not wrap as well.

When the energy is absorbed by the calorimeter in a time that is short, relative to the thermal response time, no integral corrections are necessary. The response time for the copper-Kapton laminate calorimeters is on the order of 5 milliseconds. The laser pulse width is approximately 1 ns. Hence, the calorimeter sees essentially a delta function input and the response function is that of the calorimeter only, requiring no extensive and complicated deconvolution. Deconvolution would be required in the case where the energy is input in a time comparable to or longer than the calorimeter response time.

If the measure of a performance of a calorimeter (or any measurement device) is the degree to which its behavior follows the theoretical model for the device, then these are rather good calorimeters. They behave as nearly perfect isoperibol calorimeters. The measured sensitivity for an input calorimeter prototype was 1810 μV cm^2 J^{-1} $couple^{-1}$ and the theoretically predicted sensitivity based on the heat capacitance was 1860 μV cm^2 J^{-1} $couple^{-1}$.

The effective absorption was deduced from the collective effects of Fresnel reflection (a function of the index of refraction) and the bulk absorption.

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Sensor Construction

The elements of the thermopiles are constructed to produce a 10-μV signal as a minimum in order to avoid difficulties with dissimilar metals in the connections (a solder-copper junction will produce ~3 μV/°K). Using careful construction, smaller voltages and signals could be used, however, the greatest interest lies in measuring large energies and fluxes rather than small ones.

The temperature is sensed using type E thermocouples which are chromel-constantan with a responsivity of 60.5 μV °K^{-1}, the highest of any metallic couple.[6] The couples can be conveniently piled by directly soldering the sensing couple to the copper integrating plate. Electrical isolation is accomplished by etching. The copper is removed in any pattern desired using photo-lithographic techniques. This eliminates the need for silver filled epoxies which in the past have always caused problems.

Figure 3 shows the typical construction of such a device. The Kapton in commercially available prelaminated material is Type V, a sort of sanforized or preshrunk version having superior dimensional stability. The copper can be either electrodeposited, or rolled and annealed. The electrodeposited copper is cheaper, but rolled and annealed copper is more rugged and can stand more bending and forming. For application in calorimeters electrodeposited copper is sufficient. Table 4 shows the range of Kapton thicknesses (a), and copper thicknesses (b), that are readily available, along with the sensitivities in μV cm^2 J^{-1} couple^{-1} and time constants for the various combinations.

Table 4. Copper-Kapton Laminate Data

mils of Cu	Oz. Cu	1/2	1	2	3	5	mils of Kapton
0.5	3/8	9212	7087	4849	3686	2491	
0.7	1/2	7448	5994	4312	3367	2341	
1.4	1	4237	3724	2997	2508	1891	
2.8	2	2279	2121	1864	1662	1366	
4.2	3	1557	1482	1351	1242	1069	
5.6	4	1193	1139	1061	992	879	

Sensitivities
(μV cm^2 J^{-1} couple^{-1})

mils of Cu	Oz. Cu	1/2	1	2	3	5	mils of Kapton
0.5	3/8	10.71	13.93	20.35	26.78	39.63	
0.7	1/2	13.25	16.46	22.89	29.31	42.15	
1.4	1	23.29	26.50	32.93	39.35	52.20	
2.8	2	43.31	46.53	52.95	59.37	72.23	
4.2	3	63.39	66.60	73.03	79.45	92.30	
5.6	4	83.41	86.63	93.05	99.47	112.13	

Time Constants
(seconds)

Because the integrating elements (the copper parts) can be easily isolated electrically they can be soldered directly, thereby eliminating the need for thermal epoxies. In order to assure uniform solder joints, preweighed and goldcoated solder balls are used. They are goldcoated to avoid problems with the oxide coating that would otherwise form. The oxides cause nonuniform fluxing and also tend to load the tin of the soldering iron. The soldering pens are controllable both in duration and intensity of the heat pulse applied.

The thermocouples are produced by tungsten-inert-gas (TIG) welding. Spot welding, twisting pairs of wires together using an acid dip, and constructing intrinsic thermocouples were also considered as fabrication techniques.

As far as performance, there seems to be no difference from one technique to the next. However, it is possible that over a period of time the strains introduced in twisting may relax and subtly alter the response. TIG welding was the easiest and most reliable technique that we found.

The use of a twin in the construction of the calorimeters is an excellent way to avoid drift when building small sensors. The idea here is to have two identical sensors looking at the same radiation environment. Only one sensor can see the signal of interest. Their outputs are combined with opposing polarity so the background is subtracted and only the signal is sensed. Unfortunately, for large calorimeters, say 30 cm in diameter or so, one seldom has the space to add an additional 30 cm element alongside. What is often done is to add the twin immediately behind the sensing element, but looking in the opposite direction. With an appropriately designed optical cavity around the pair this is effective. However, it does become bulky. In many situations a twin is impractical because of the

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this is true. In general, however, the ambient radiation environment is not rapidly varying and a twin is often not necessary. In those situations a heavy copper plate is used to provide the reference temperature.

A typical design in use on Antares is illustrated in Fig. 4, the input calorimeter. There are 48 sensing elements and a heavy reference plate. A heavy aluminum sheet metal shroud is provided to isolate this sensor as it works in air. All other Antares calorimeters work in vacuum.

Calibration

A calibration facility is maintained at Los Alamos National Laboratory which provides a National Bureau of Standards (NBS) traceable calibration. The facility is very similar to that at the Bureau. A transversely excited atmospheric (TEA) laser with a smoothing tube provides up to 2 J of 10.6 μm radiation in 100 ns pulses. This is used in a beamsplitter arrangement to calibrate each element of each calorimeter which is used in the system. The tertiary standards in the facility are twinned Los Alamos National Laboratory designs using beryllium oxide (BeO) as the absorber. These are recalibrated against the NBS secondary standards twice a year. This procedure eliminates any question about equivalence of sources in electrical calibrations.

Data Reduction

Several methods of data reduction were evaluated. The curve-fitting and integration technique,[7] a standard exponential fit,[8] a weighted exponential fit,[9] peak-sensing,[10] and a delayed sampling method[11] were compared. As noted by Johnson,[12] if the calorimeter is well behaved and can be characterized by, at most, two time constants, four points are sufficient to sample the calorimeter response. Similarly, if the calorimeter is well-behaved and exhibits substantially a single time constant, one sampled point appears sufficient to provide results which are reproducible to better than 1%. Figure 5 shows a typical response curve for the input calorimeter. Sample runs for the input calorimeter operated in air are shown in Table 5.

Table 5. Typical Input Calorimeter Test Data

Shot Number		Calorimeter Sensitivity	
1		668 μV/J	
2		651	
3		639	
4		630	
5		625	
	Mean	643	2.7%
	Std. Dev.	17	

It is clear from the Table that there is a systematic error, which possibly could be eliminated. If considered as random errors the standard deviation in this set of 5 is only 2.7%. These data were taken using a 100 J gain-switched 100 ns pulse over a 150-mm-diameter beam directed into an 8" Scientech calorimeter. A salt beam splitter provided 8 J into the prototype input calorimeter. It is not clear which calorimeter is responsible for the drift. The absolute accuracy of the laser calibration via NBS is about 5%. Therefore, it does not seem practical to agonize over 0.1% in precision of reduction techniques.

Summary

Copper-Kapton laminated absorbing materials for 10 μm radiation allow the construction of calorimeters of 5% absolute accuracy in virtually any configuration and size.

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The authors wish to thank Steve Levings, Joyce Dock, and Valerie Adams for their assistance in the construction and evaluation of the calorimeters reported here. Chris Shera made valuable contributions in computer programming and automated data acquisition while a summer student and casual employee for the Laboratory. Others too numerous to mention have helped in carrying out these experiments, the design of the actual Antares calorimeters, and the preparation of this manuscript and illustrations. Their help is greatly appreciated.

References

1. E. D. West and K. L. Churney, "Theory of Isoperibol Calorimetry for Laser Power and Energy Measurements," J. Appl. Phys. 41, 2705 (1970).
2. Rob E. Watt, "Calorimeter for Picosecond Laser Pulses," Appl. Opt. 12, 2373 (1973).

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3. D. L. Franzen and L. B. Schmidt, "Absolute Reference Calorimeter for Measuring High-Power Laser Pulses," Appl. Opt. 15, 3115 (1976).
4. Stuart R. Gunn, "Volume-Absorbing Calorimeters for High-Power Laser Pulses," Rev. Sci. Instrum. 45, 936 (1974).
5. The Scientech volume absorbing calorimeters use Teflon(TM).
6. Based upon the 1971 revised NBS reference tables superseding NBS Circular No. 561.
7. E. D. Watt and W. E. Case, IEEE Trans. Instrum. Meas. IM-23, 422 (1974).
8. A linear regression method used on the natural log values of the voltage response vs. time.
9. A. Picot, Am. J. Phys. 48, 302 (1980).
10. Simply one value -- the peak of the response is taken.
11. In this method the calorimeter response is sampled $(10\epsilon)^{-1}$ after the peak. This avoids spiking problems that can occur on the peak signal due to a peculiar radiation distribution.
12. Eric G. Johnson, Jr., Appl. Opt. 16, 2315 (1977).

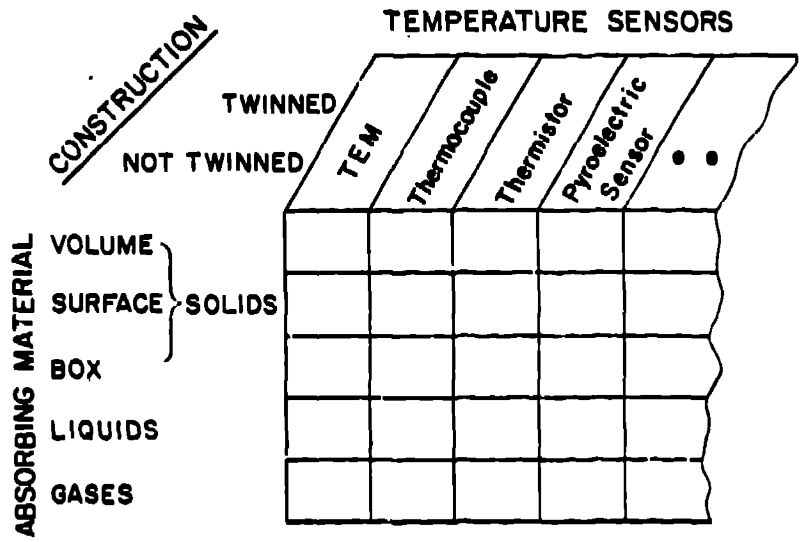
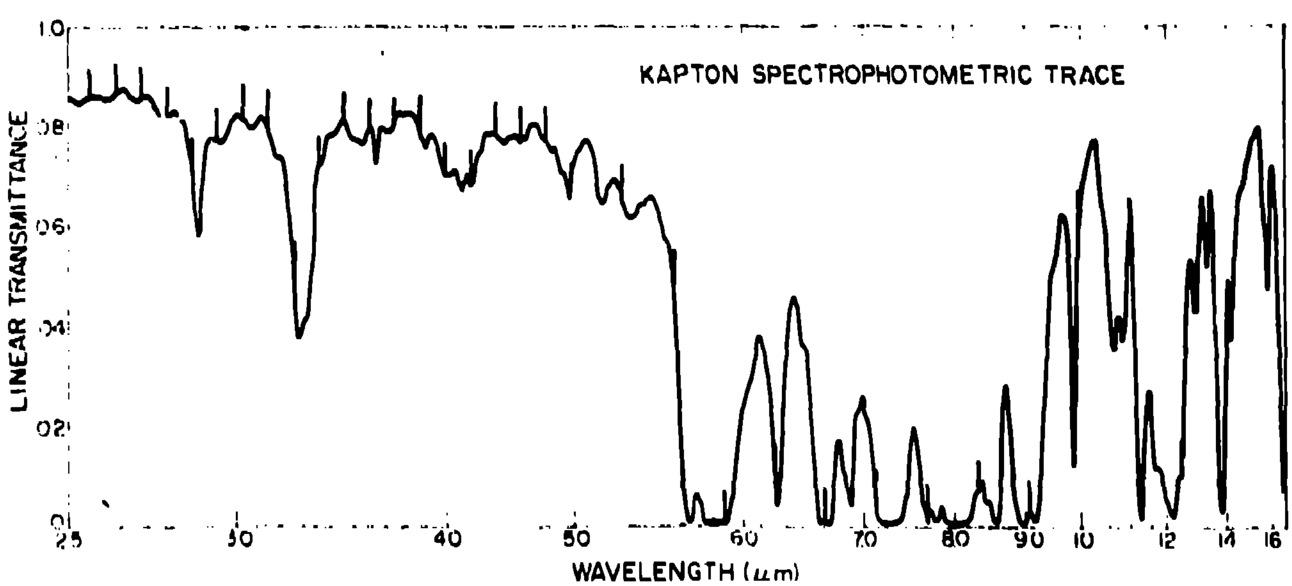


Fig. 1. Thermal energy sensing device matrix.



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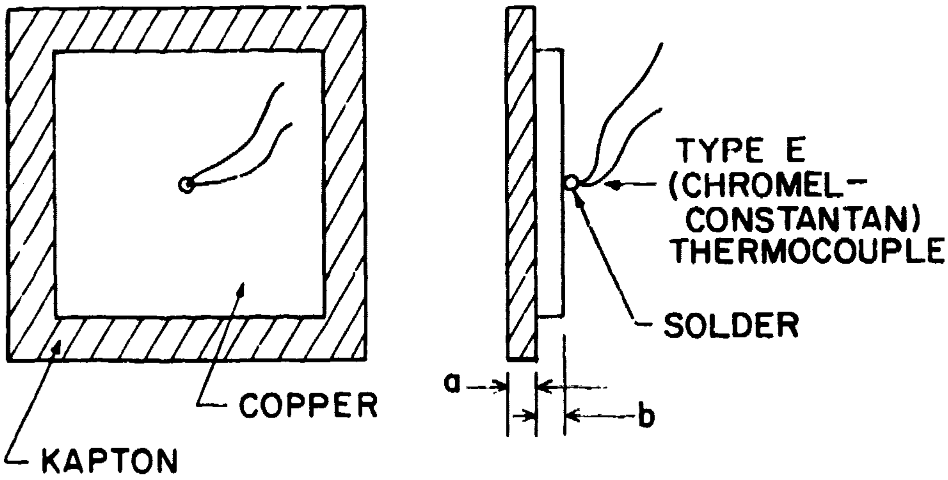

Fig. 3. Typical copper-Kapton calorimeter element.

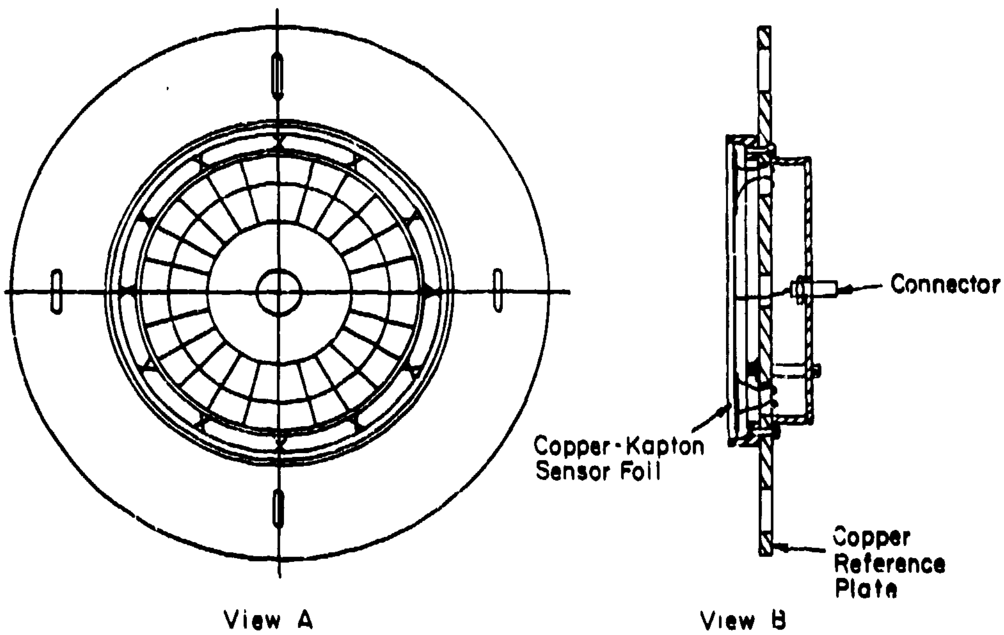

Fig. 4. Input calorimeter.

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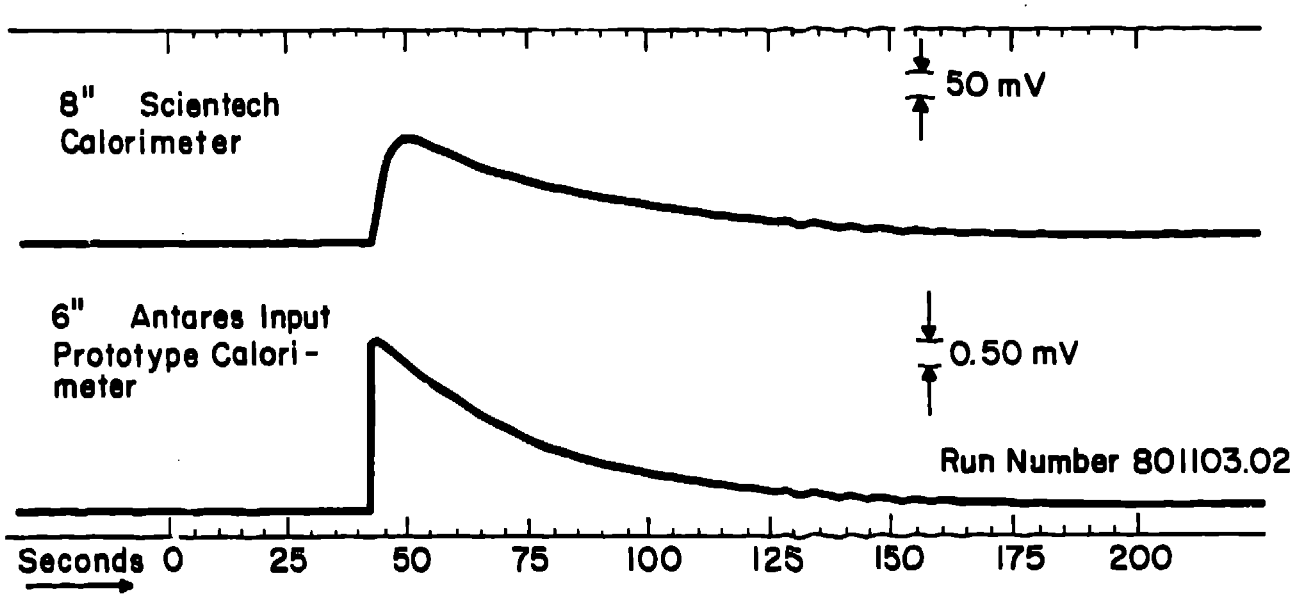

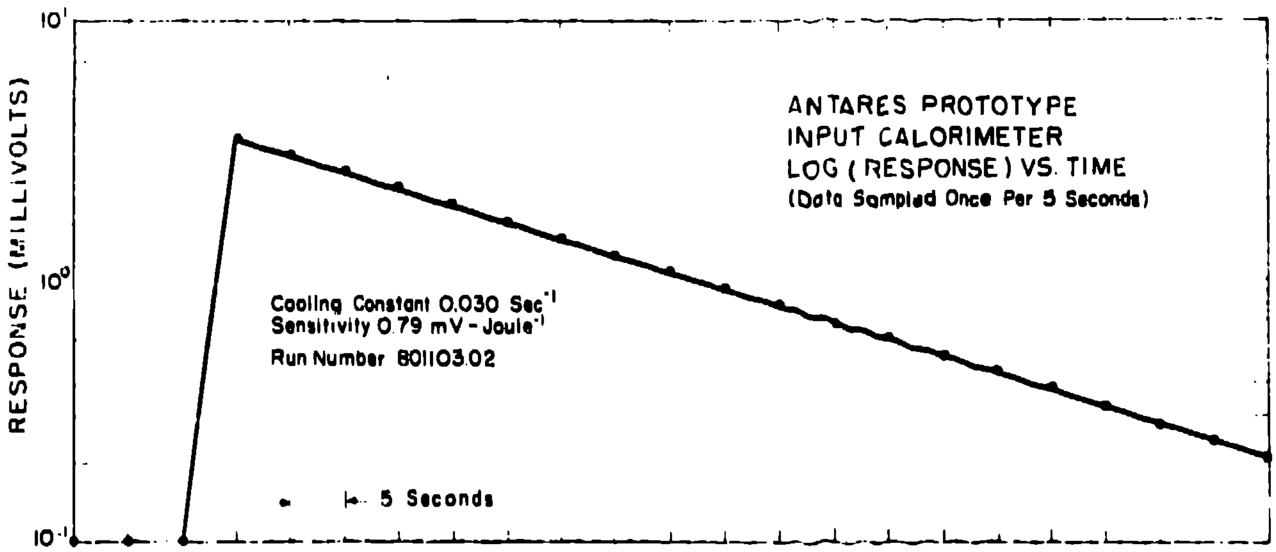

Fig. 5. A typical response curve of the input calorimeter.