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Thermal Initiation of Confined Primary Explosives with a Proton Beam

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The thermal initiation characteristics of the primary explosives lead azide and lead styphnate have been studied by heating confined samples with a 200-MeV proton beam. Values were obtained for thermal initiation thresholds, explosion temperatures, and average specific heats. Thresholds and explosion temperatures were found to depend strongly on the beam heating rate.					
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THERMAL INITIATION OF CONFINED PRIMARY EXPLOSIVES WITH A PROTON BEAM

INTRODUCTION

We have developed techniques over the past decade for studying the thermal initiation of confined energetic materials via uniform heating with a high-energy charged particle beam. The beam deposits energy in the target principally by ionization, which results in a temperature rise due to increased molecular motion. A large amount of work has been done with electron beam heating of confined high explosives [1, 2, 3, 4]. These data have yielded values of thermal initiation threshold (dose required to initiate a runaway chemical reaction) and explosion temperature for a variety of explosives. In the work reported here, we have extended these measurements to the study of initiation of primary explosives by a proton beam.

Primary explosives are used as ignitors for propellants and high explosives. It is difficult to obtain data on these materials for samples which weigh about 1 gm or larger since they are subject to accidental ignition by electrostatic spark, friction, or impact. Using elaborate safety precautions, we successfully assembled confinement cells containing about 1-gm samples of lead azide or lead styphnate, each with an imbedded thermocouple. Tests were performed on a total of 14 confined samples by placing them in the 200-MeV proton beam at the Brookhaven National Laboratory (BNL) Radiation Effects Facility (REF). All of the samples exploded violently, yielding data on thermal initiation thresholds, explosion temperatures, and average specific heats.

EXPERIMENTAL PROCEDURE

Samples consisting of two pressed powder disks of primary explosive were assembled in Al confinement cells and sealed with O-rings and Al windows as shown in Fig. 1. Special safety precautions taken during assembly and testing include: use of a 1-inch thick lucite shield, face mask, grounded wrist straps (straps), grounded Al work table, wearing Kevlar gloves, cotton overalls, and conducting plastic shoe covers. The diameter of the sample disks was 1/4-inch and the total mass was about 1.16 gm for the lead azide samples, and about 0.88 gm for the lower density lead styphnate samples. The disks were pressed to about 70% theoretical maximum density at the Naval Surface Warfare Center. About 1% Teflon by weight was added to the lead styphnate powder to improve cohesion. A 5-mil diameter chromel-alumel thermocouple junction is sandwiched between the two disks, which are surrounded by thermally insulating material. The exit window is 1/32-inch thick Al, and is designed to blow out first. In practice, these primary explosives always go into detonation after initiation, usually blowing out the entrance window (1/8-inch thick Al) as well as the exit window. The accelerator exit window is protected by a 1/4-inch thick Al plate. All tests were performed inside a cubic containment box, with 2-foot sides.

Data on temperature vs time were obtained by both a computer and a chart recorder. The principal dosimeter was an Al calorimeter disk placed in the re-entrant hole as shown in Fig. 1, so that the beam passes through it before entering the sample. Additional dosimetry (provided by BNL personnel) came from Al activated foils placed before each sample. Unfortunately, all but one of these foils were destroyed by the violence of the detonations. Beam profile data were taken for 5 and 10 beam pulses by exposing Al foils for activation analysis

by BNL personnel), and also radiachromic films which were scanned by an optical densitometer. The results were in excellent agreement, showing a rather tight beam, about 1.2 cm horizontal and 1.8 cm vertical, full width at half maximum. A three-dimensional picture of the beam profile with 0.5 cm resolution, obtained from the foil activation data, is shown in Fig. 2. Alignment was done with respect to a laser beam. However, the particle beam was not always coincident with this position, so some of the samples were not well aligned with the beam. The REF beam parameters were as follows: energy, 200 MeV; pulse current, 22 mA; pulse width, 320 μ sec; repetition rate, 5/sec (every 12th pulse missing). The beam actually consists of H^+ ions, but the 2 atomic electrons are stripped off when the beam enters a solid material, yielding a proton beam into the explosive target.

RESULTS AND DISCUSSION

A. Lead Azide

Eight identical confined samples of lead azide were exposed to the REF beam until explosion. An example of the temperature vs time data is shown in Fig 3. The upper curve is for the explosive, and the lower one is for the Al calorimeter. In this case, where the alignment was good, detonation occurs 3.64 sec after the beam is turned on, or after 17 pulses. The effect of the individual pulses produces the staircase pattern observed, with cooling between the pulses. The straight lines are least-squares fits to the data to obtain the average slopes. The results of analysis of the 8 runs are tabulated in Table 1.

Table 1. Lead Azide Initiation Data at the REF

Average Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
Explosive ^a	Calorimeter		Explosive ^a	Calorimeter	
4.96 ^b	5.97 ^b	4.25	21.1	25.4	188
6.95	6.85	4.30	29.9	29.5	265
8.40	8.15	3.86	32.4	31.5	290
8.92	9.36	3.64	32.5 ^c	34.1	290
8.02	8.21	4.12	33.1	33.8	290
8.72	8.15	4.00	34.9	32.6	300
7.82	7.94	4.21	32.9	33.4	290
8.13	8.30	3.27	<u>26.6</u>	<u>27.1</u>	<u>240</u>
Averages (Excluding First Run):			31.7±1.0	31.7±1.0	281±8

a. Assuming $\bar{C}_v = 0.136 \text{ cal/(g.}^\circ\text{C)}$.

b. Poor alignment.

c. Al foil activation dose = 31.3 cal/g.

Heating rates are obtained by multiplying slopes by average specific heats over the temperature range measured. Thermal initiation thresholds are then obtained from the product of average heating rates and observed times to explosion. The specific heat of lead azide as a function of temperature is not well known. An average value of $\bar{C}_v = 0.136 \text{ cal/(g.}^\circ\text{C)}$ was chosen, which yields a result for the average threshold in agreement with the value obtained from calorimetry. This constitutes a measurement of the average specific heat of lead azide for the temperature range between ambient and explosion. The first run was excluded from these averages since it exploded early, yielding unusually low values for threshold and explosion temperature. This may be associated with

the low heating rate for this run due to poor alignment with the beam center. Only one activated foil survived explosion to allow dosimetry analysis (run 4). It yielded a dose to explosion of 31.3 cal/g, in excellent agreement with the threshold results obtained from the explosive and calorimeter (32.5 and 34.1 cal/g, respectively). These low thresholds found for lead azide mean that it is very sensitive to thermal initiation as well as to other stimuli.

B. Lead Styphnate

Six confined samples of lead styphnate were tested at the REF. An example of the data is shown in Fig. 4. Clearly, this material is less thermally sensitive than the azide, requiring many more pulses for ignition. Again, the upper curve is for the explosive and the lower curve is for the calorimeter. Note the absence of every 12th pulse, and the cooling between pulses. The straight lines are again least-squares fits to the data to obtain average slopes. The results of analysis of these data are given in Table 2.

Table 2. Lead Styphnate Initiation Data at the REF

Average Heating Rate (cal/g.s)		Time To Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
<u>Explosive^a</u>	<u>Calorimeter</u>		<u>Explosive^a</u>	<u>Calorimeter</u>	
8.82	8.57	7.11	62.7	60.9	310
8.31	8.34	6.80	56.5	56.7	310
8.76	9.41	6.79	59.5	63.9	310
4.56 ^b	4.22 ^b	10.23	46.7	43.2	250
5.42 ^b	5.07 ^b	9.73	52.8	49.3	275
3.78 ^b	4.22 ^b	10.82	<u>40.9</u>	<u>45.6</u>	<u>230</u>
Averages (First 3 Runs):			59.6±1.8	60.5±2.1	310±0
Averages (Last 3 Runs):			46.8±3.5	46.1±1.8	252±13

a. Assuming $\bar{C}_v = 0.224$ cal/(g.°C)

b. Poor alignment.

The specific heat of this material at higher temperatures is not known. The average value $\bar{C}_v = 0.224 \text{ cal/(g.}^\circ\text{C)}$ yields good agreement with the calorimeter results. A strong heating rate dependence of the threshold was found for this material. The last three runs had lower heating rates (due to misalignment with the beam), and they all exploded at lower temperatures, yielding lower thresholds than the samples which were heated more rapidly. Thus, separate average values were obtained for the first three runs and the last three runs. The heating rate dependence of the thermal threshold is clearly illustrated in Fig. 5.

CONCLUSIONS

The thermal initiation thresholds of primary explosives appear to vary widely. The threshold for lead azide is about half that for lead styphnate. Previously [5], we found that the sensitive explosive PETN exhibits a range of thresholds (from 50 to 110 cal/g). These materials are unstable, and can occasionally explode at a lower threshold for no apparent reason.

The results are in very good agreement with results obtained with a 40-MeV electron beam at the NRL Linac [5]. When the same values of specific heat are used, the threshold results agree within 8%.

A strong heating rate dependence was found for these materials; thresholds are considerably lower for lower heating rates. For lead styphnate, a reduction of the heating rate by a factor of two results in about a 50% reduction in the thermal threshold. The explanation may be due to exothermic activity well below explosion temperature. Self-heating from this exothermic activity makes a larger contribution at lower beam heating rates. This property of primary explosives is helpful in that, to some extent, it reduces the necessity for perfect beam alignment to initiate the explosive.

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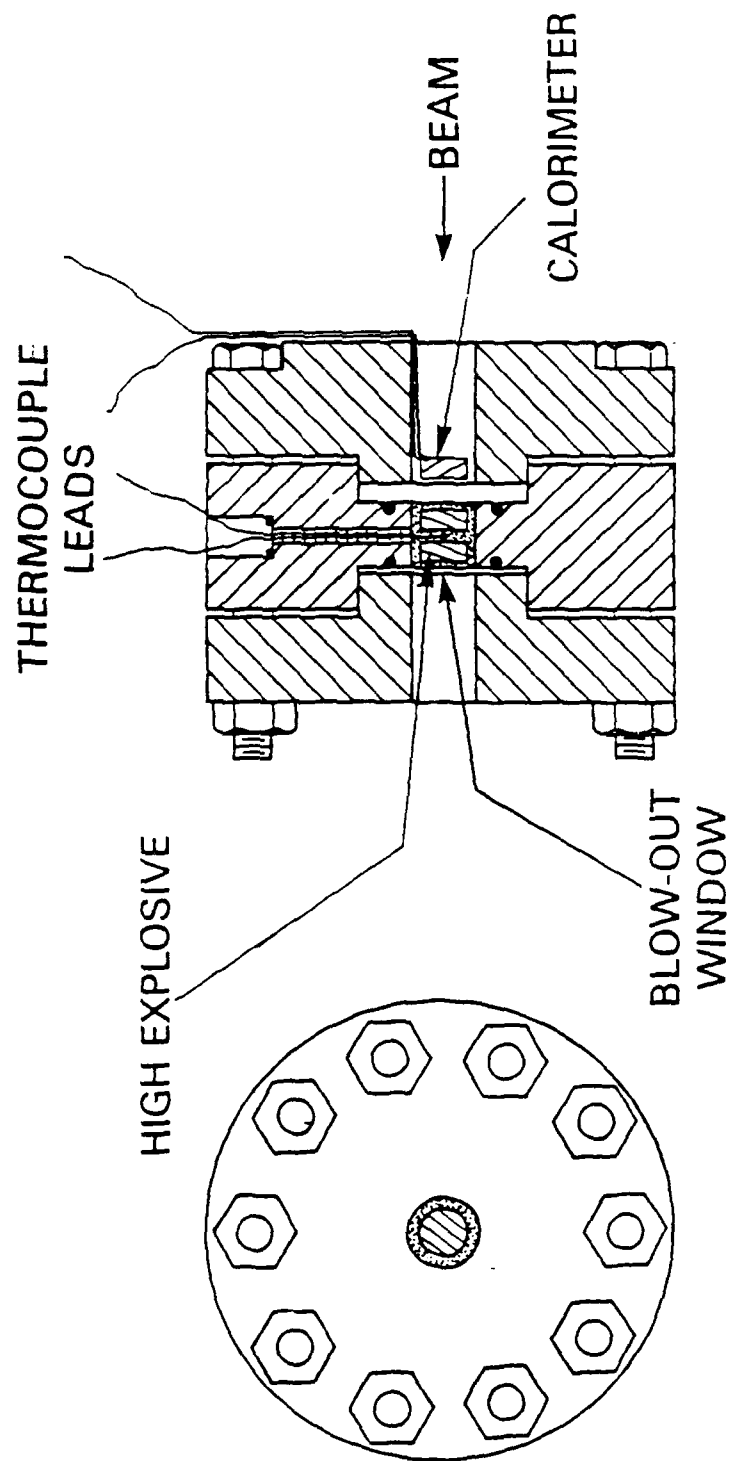


Figure 1. Aluminum confinement cell for thermal explosion experiments on primaries with a proton beam. The outer diameter of the cell is 7.0 cm.

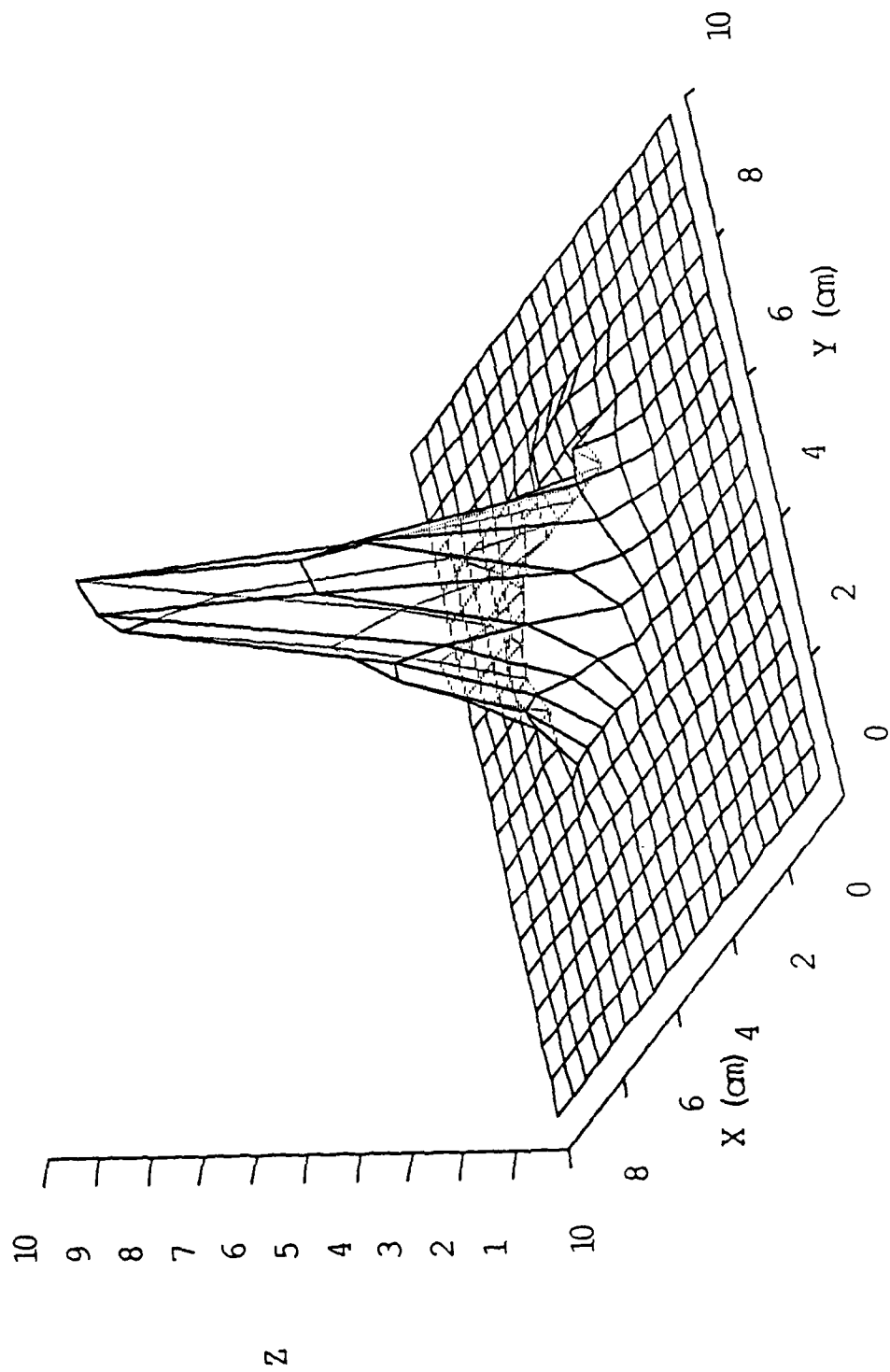


Figure 2. Beam profile obtained by aluminum foil activation analysis. The resolution is 0.5 cm in the x and y directions.

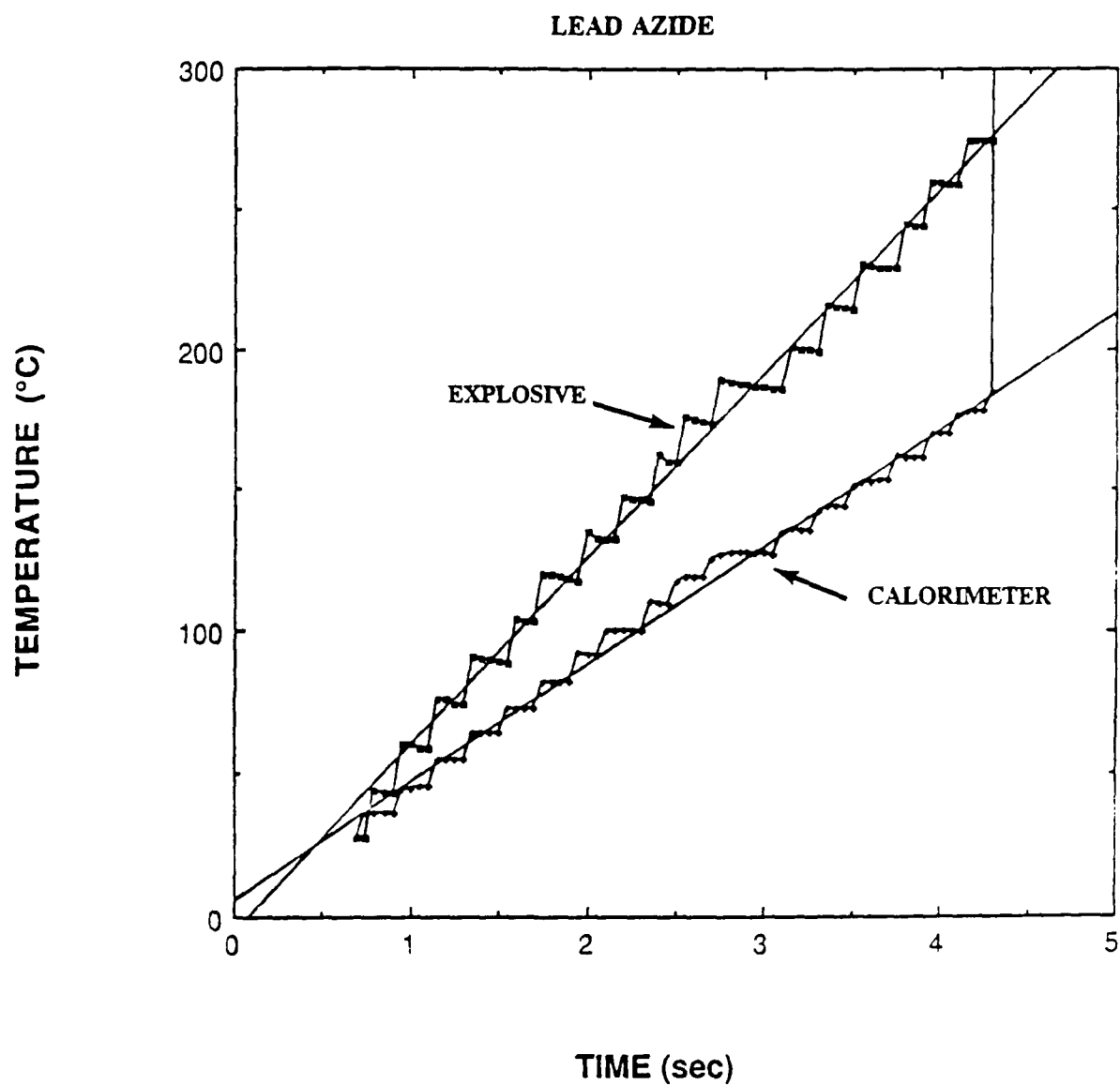


Figure 3. Thermal behavior of lead azide heated uniformly by proton beam pulses to explosion (upper curve). The lower curve shows the response of an Al calorimeter.

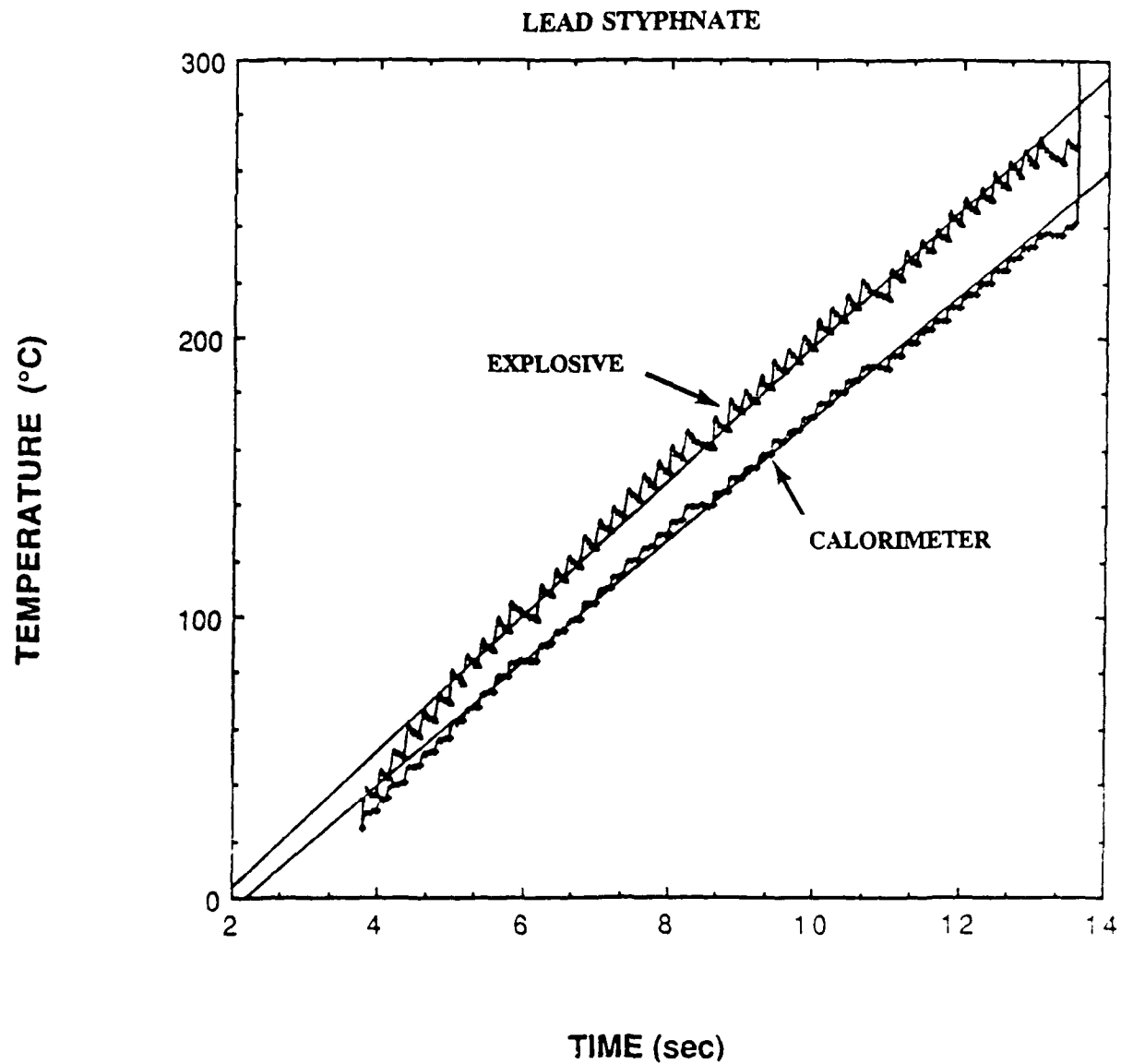


Figure 4. Thermal behavior of lead styphnate heated uniformly by proton beam pulses to explosion (upper curve). The lower curve shows the response of an Al calorimeter.

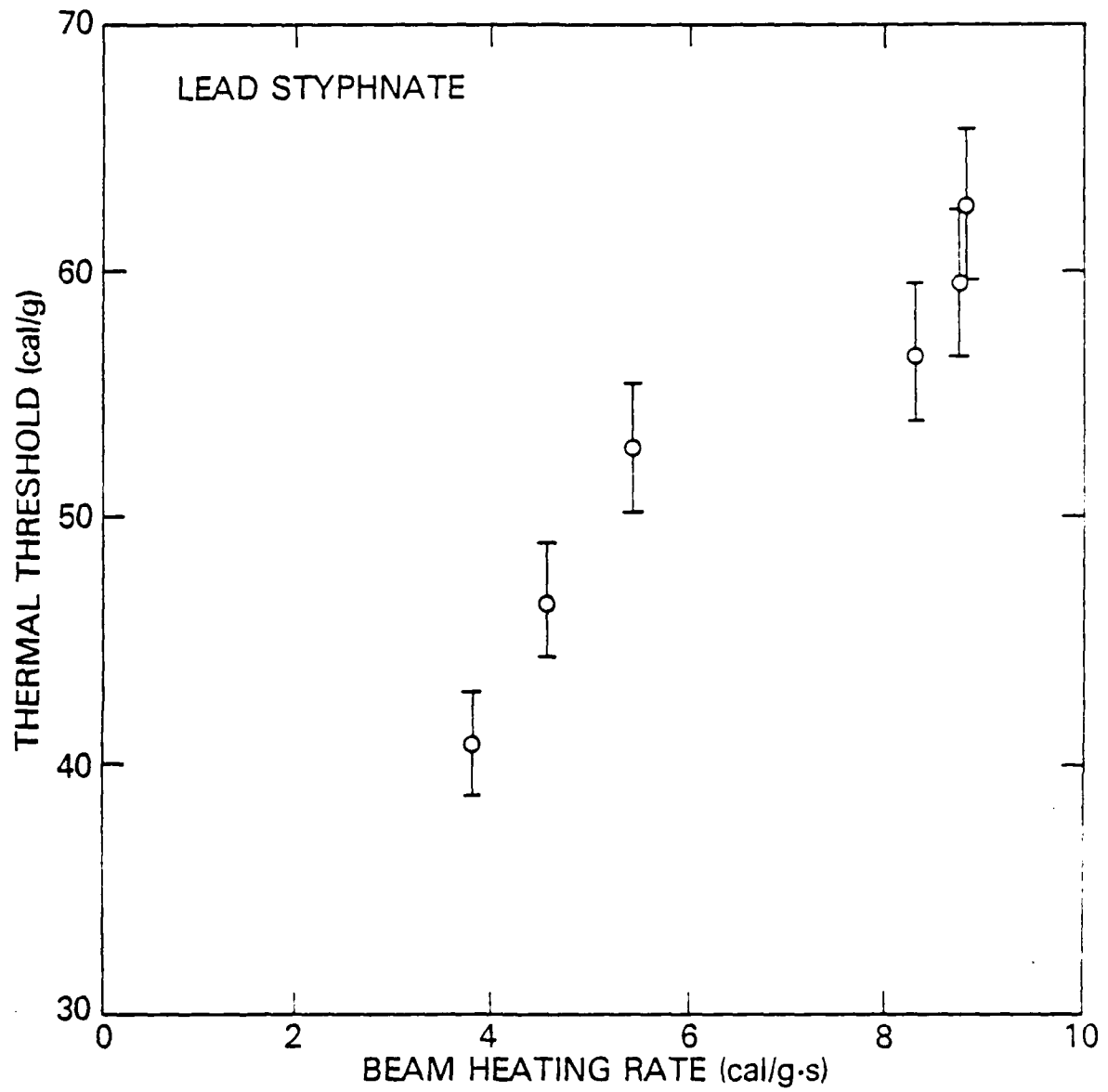


Figure 5. Thermal initiation thresholds of lead styphnate vs beam heating rate.