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Solid Rocket Propellant Initiation Via Particle Beam Heating

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SOLID ROCKET PROPELLANT INITIATION VIA PARTICLE BEAM HEATING

Introduction

Solid rocket propellants are designed to burn at a controlled rate, and to generate hot gases which escape from a nozzle to provide thrust to the rocket for an extended period of time. Composite rocket formulations are mixtures of a fuel and an oxidizer. The fuel can be an organic polymer binder, powdered aluminum, or an explosive such as HMX. The oxidizer is often powdered ammonium perchlorate (AP). Small amounts of other materials are often added to moderate the burn rate, improve physical properties, etc. Under proper operating conditions, an ignitor starts exothermic reactions in the fuel-oxidizer mixture, which converts from the solid phase to the gas phase at a controlled rate without exploding or detonating. However, there have been several incidents in which solid propellants have ignited and exploded accidentally, with loss of life. It is therefore of importance to investigate the conditions under which a solid propellant will explode violently.

In this work, we report experiments in which solid propellants are confined (so that reaction gases cannot escape), and are heated uniformly by a penetrating electron or proton beam to initiation and explosion. The explosion is due to a runaway exothermic reaction, producing a rapid pressure buildup which ruptures the confining chamber. The data yield values for the explosion temperature, and the thermal initiation threshold, i.e., the minimum deposited energy per gram required to produce explosion of the confined propellant. This work is an extension of similar experiments with confined explosives reported previously^{1,2,3}. We have studied two propellants based upon AP, and three propellants which use a mixture of HMX, AP, Al powder and NG (nitroglycerine).

EXPERIMENTAL PROCEDURE

The experimental technique was basically the same as has been described previously^{1,2,3}. The samples consisted of two disks of solid propellant, each about 6 mm in diameter and 5 mm thick, with an iron-constantan thermocouple junction (0.127 mm wire diameter) sandwiched between them. The propellant samples (about 0.5 gm) were thermally insulated and confined via aluminum windows and indium or O-ring seals in a confinement cell as shown in Fig. 1. The exit window was 0.794 mm-thick aluminum, which will generally blow out before the 3.18 mm-thick aluminum entrance window, thus protecting the accelerator. The rubbery composite propellants were obtained from the Naval Ordnance Station, Indian Head, Md.; the disks were easily cut from stock material. In the case of pure AP the disks were pressed from powder.

Most of the confined samples were exposed to electron beam pulses at the NRL Linac, with the following beam pulse parameters: 40 MeV energy, 1.5 μ s width, 300 mA peak current, 360 Hz repetition rate, and 1 cm diameter Gaussian spot size (full width at half maximum). When properly aligned, the beam heated the samples almost uniformly in profile and depth at a rate of about 15 cal/(g.s). The uniformity of heating provided by this technique greatly simplifies analysis of the data. Typically, data were obtained on about eight samples to yield average results for initiation thresholds and explosion temperatures. The beam passed through an aluminum disk with an imbedded thermocouple which served as a calorimetric beam monitor (dosimeter) before it entered the sample. Data on temperature vs time were obtained for both the propellant sample and the calorimeter. Data acquisition was by both a computer and a chart recorder. All

experiments were performed in air, with the confinement cells placed about 12 cm from the Linac exit window to provide an approximately uniform beam profile at the propellant sample. The Linac exit window was protected from blast by a 1.6 mm thick Al plate, or by bending the beam away from the window with a deflection magnet.

One set of experiments (on VRP propellant) was performed at the Brookhaven National Laboratory Radiation Effects Facility (REF). This was an H^- beam which became a proton beam since the atomic electrons were stripped off in the exit window foil. The beam pulse parameters were: 180 MeV energy, 420 μs width, 20 mA peak current, 5 Hz repetition rate, and 1.4 cm Gaussian spot size diameter (full width at half maximum). A 1.27 cm thick Al plate protected the accelerator from blast.

RESULTS

A. AP-Based Composite

This is a rubbery propellant containing 84% AP (NH_4ClO_4), 9.9% Hycar gum rubber binder, and smaller amounts of plasticizer, catalyst, curative, bonder and stabilizer materials. Nine confined samples were exposed to the electron beam at the Linac until explosion occurred. A typical thermocouple record is shown in Fig. 2. The plateau which appears at about 250°C is due to a crystalline phase transition in AP at about this temperature⁴. The data are tabulated in Table 1.

Table 1. AP-Based Composite Propellant Initiation Data

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
<u>Propellant*</u>	<u>Calorimeter</u>		<u>Propellant*</u>	<u>Calorimeter</u>	
15.0	15.7	6.61	99.0	103.7	290
12.9	13.8	8.40	108.3	116.2	300
11.4	11.7	9.34	106.0	109.6	---
12.7	14.3	7.75	98.1	111.0	275
12.4	10.7	8.42	104.6	90.4	290
14.3	14.8	7.90	112.9	116.9	305
13.2	13.3	8.26	109.3	109.6	300
13.9	14.2	7.60	105.9	107.5	335
6.1	6.2	16.04	97.3	99.3	---
Averages:			104.6 ± 1.8*	107.1 ± 2.8	299 ± 6

* Using $\bar{C}_v = 0.36 \text{ cal/(g.}^\circ\text{C)}$

The beam heating rate is obtained from the slope of the initial portion of the curve multiplied by the average specific heat over this temperature range. Heating rate data are obtained from both the calorimeter and from the propellant itself. The product of these heating rates and the observed times to explosion then yield values of thermal initiation threshold. Unfortunately, the average specific heat of this composite propellant has not been measured, although for its major constituent, AP, the specific heat is known to be about 0.31

cal/(g.°C) below the transition temperature.⁴ We have chosen the value $\bar{C}_v = 0.36$ cal/(g.°C), because it yields an average value of the initiation threshold in agreement with that obtained from the calorimeter. The last run was done at half (180 Hz) the normal beam pulse rate to look for a possible heating rate dependence; the threshold value obtained is compatible with the other data. Explosion temperatures were not obtained for two runs because thermocouple contact with the propellant deteriorated near explosion. The uncertainties quoted in Table 1 are statistical, i.e., standard deviations of the mean.

B. AP

Pellets were pressed from pure ammonium perchlorate powder, and seven confined samples (~ 0.6 g each) were exposed to the Linac electron beam until explosion in order to obtain data for a material with a known specific heat.⁴ A response curve is shown in Fig. 3, which looks very similar to that of Fig. 2, as expected. The data are listed in Table 2 below. Uncertainties are standard deviations of the mean.

Table 2. Initiation Data for Pure AP.

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
AP	Calorimeter		AP	Calorimeter	
5.88	5.85	16.08	94.6	94.1	325
8.62	8.09	11.76	101.4	95.2	300
9.93	9.56	9.67	96.0	92.5	330
12.28	11.39	8.02	98.5	91.4	330
11.51	10.93	8.36	96.3	91.4	315
11.72	10.76	8.35	97.9	89.9	310
4.83	4.25	19.94	<u>96.2</u>	<u>84.7</u>	<u>315</u>
Averages:			97.3 $\pm$ 0.8	91.3 $\pm$ 1.3	318 $\pm$ 4

The last run was done at 180 Hz (half the usual pulse rate); it yields a threshold and explosion temperature consistent with the other data. The 7% difference between thresholds obtained from the AP and the calorimeter is probably due to specific heat uncertainty.

C. VRP

This is a high-thrust Navy propellant which contains the explosives HMX and NG (nitroglycerine) as well as AP, Al powder, binder and other ingredients. It has a rubbery consistency. Two series of measurements were made on this material. In the first series, eight confined samples (~ 0.5 g each) were irradiated to explosion with the Linac electron beam. In Fig. 4, we show initiation of this material by a deflected beam, which is of reduced intensity because of dispersion. The other runs were done in the direct beam, at faster heating rates. The temperature rise is uniform up to explosion at about 210°C. The data are listed in Table 3. Uncertainties are standard deviations of the mean. In calculating heating rates for the propellant, we have used an average specific heat $\bar{C}_v = 0.28$ cal/(g.°C), which was calculated from the major constituents. This yields threshold values for the propellant which are in reasonable agreement with those from the calorimeter.

Table 3. Thermal Initiation of VRP at the NRL Linac

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
VRP	Calorimeter		VRP	Calorimeter	
3.06*	2.87*	15.67*	48.0	44.9	210
15.9	16.6	3.18	50.6	52.7	200
15.2	16.4	3.40	51.7	55.7	215
17.1	17.8	3.04	52.0	54.2	225
5.83	6.31	8.11	47.3	51.2	200
6.72	7.55	6.76	45.4	51.1	205
7.37	7.60	6.44	47.5	48.9	215
6.05	7.33	7.88	47.6	57.8	215
Averages:			48.8 ± 0.8	52.1 ± 1.4	211 ± 3

* Exposed to deflected beam

The second series of data on this material was done by exposure of six confined samples to the proton beam at the Brookhaven REF. Initiation of VRP by this beam is shown in Fig. 5, which appears to be very similar to initiation via the electron beam (Fig. 4). The data for six runs are given in Table 4 for a variety of heating rates. The last run was done under an unusual condition: aluminum energy absorbers of carefully calculated thickness were placed in the 180-MeV beam (upstream from the target) so that the protons stopped in the propellant sample, producing a sharp increase in energy absorption at the end of the proton range⁵ (sometimes called the Bragg peak). This results in explosion at a higher temperature and with a larger threshold. We have excluded this run in obtaining the average values given in Table 4, which are in rather good agreement with the electron beam results (Table 3). This illustrates the

fact that the initiation is due simply to heating by the beam, and is independent of the type of beam particle (with the exception of the Bragg peak case).

Table 4. VRP Thermal Initiation Data at the REF

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
VRP	Calorimeter		VRP	Calorimeter	
4.43	3.98	9.82	43.5	38.8	175
9.12	8.06	6.10	55.7	49.2	230
3.55	3.19	12.06	42.8	38.5	175
7.33	7.34	6.68	49.0	49.1	190
3.46	3.44	14.20	49.2	49.0	240
19.22*	10.01	3.60	69.2*	36.0	260*
Averages, excluding last run:			48 ± 2	45 ± 3	202 ± 14

* Bragg peak initiated

D. VTQ-2 and VTG-5A

These are propellants with formulations similar to that of VRP, differing only in the relative percentages of energetic ingredients, binders, etc. Confined samples were exposed to the direct electron beam at the NRL Linac. Typical data plots are shown in Figs. 6 and 7 for VTQ-2 and VTG-5A, respectively. They both show evidence for exothermic activity above about 170°C, with explosion

at about 220°C. Data for the individual runs are given in Tables 5 and 6. We have again taken the average specific heat to be $\bar{C}_v = 0.28 \text{ cal/(g.}^\circ\text{C)}$, which yields excellent agreement between the propellant and calorimeter results. Again, the uncertainties are standard deviations of the mean.

Table 5. Thermal Initiation of VTQ-2

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
<u>VTQ-2</u>	<u>Calorimeter</u>		<u>VTQ-2</u>	<u>Calorimeter</u>	
14.2	14.5	3.92	55.7	57.0	240
19.5	18.0	3.01	58.7	54.1	235
15.8	15.1	3.36	53.0	50.8	220
7.84	8.10	6.03	47.3	48.8	208
7.65	8.10	5.99	45.8	48.5	215
6.83	7.83	6.48	44.3	50.7	192
9.41	8.61	4.92	46.3	42.4	210
7.89	8.85	6.12	48.3	54.1	222
7.00	6.67	7.20	50.4	48.0	---
Averages:			50.0 ± 1.6	50.5 ± 1.4	218 ± 6

Table 6. Thermal Initiation of VTG-5A

Heating Rate (cal/g.s)		Time to Explosion (s)	Thermal Threshold (cal/g)		Explosion Temperature (°C)
<u>VTG-5A</u>	<u>Calorimeter</u>		<u>VTG-5A</u>	<u>Calorimeter</u>	
14.5	14.2	3.72	53.9	52.7	253
14.8	13.8	3.41	50.6	47.0	225
20.2	18.8	2.82	57.0	53.1	242
16.8	15.6	3.23	54.3	50.4	245
7.65	8.07	6.11	46.8	49.3	225
7.80	7.42	6.46	50.4	47.9	215
4.26	5.37	10.87	46.3	58.4	210
5.36	6.72	8.74	46.9	58.7	2.03
Averages:			50.8 ± 1.4	52.2 ± 1.6	227 ± 6

CONCLUSIONS

The major results of this investigation are summarized in Table 7 below:

Table 7. Thermal Initiation Thresholds and Explosion Temperatures for Five Solid Propellants.

<u>Propellant</u>	<u>Major Ingredients</u>	<u>Thermal Threshold (cal/g)</u>	<u>Explosion Temperature (°C)</u>
AP-Based	AP/Hycar binder	105 ± 10	299 ± 6
AP	AP	97 ± 6	318 ± 4
VRP	HMX/AP/NG/Al/binder	49 ± 5	211 ± 3
VTQ-2	HMX/AP/NG/Al/binder	50 ± 5	218 ± 6
VTG-5A	HMX/AP/NG/Al/binder	51 ± 5	227 ± 6

It is seen that thresholds for propellants in which AP is the major energetic ingredient are about twice those for the more sensitive propellants containing HMX, AP and NG. It seems probable that the low thresholds for the latter group are due to the NG ingredient, since they are lower than the thresholds for AP or for HMX.² The uncertainties given in Table 7 for threshold values are larger than the statistical errors given in the other tables, because they include uncertainties in the specific heats, and the spread between the calorimeter and propellant results. The results given in this report all refer to an initial temperature of 22°C (room temperature).

The essential agreement between the results for the AP-based composite propellant with those for pure AP indicate that the thermal behavior of the composite is dominated by that for AP. The three more sensitive propellants

yield very similar thermal initiation results, indicating that the most sensitive common ingredient (probably NG) is dominant.

The agreement between data obtained with electron and proton beams (for VRP) shows that specific radiative effects (knock-on molecular breakup) must be quite small. Almost all of the absorbed energy goes into ionization, and ends up as heat. Thus, beam irradiation is a convenient way to deposit heat uniformly in a confined target material.

The AP materials do not show any evidence for a dependence of thresholds or explosion temperatures on the beam heating rate. An examination of the data for the more sensitive HMX/NG-containing materials (Tables 3-6) does indicate such a dependence. We have previously seen this effect in other sensitive materials.^{2,3} The effect is probably due to exothermic activity before explosion. This self heating contributes more to the temperature rise when the beam heating rate is low.

These data are useful in assessing the stability and relative safety of propellant materials when exposed to a thermal source (e.g., near a fire). Clearly, the AP-based propellants are considerable safer than the HMX/AP/NG/Al propellants.

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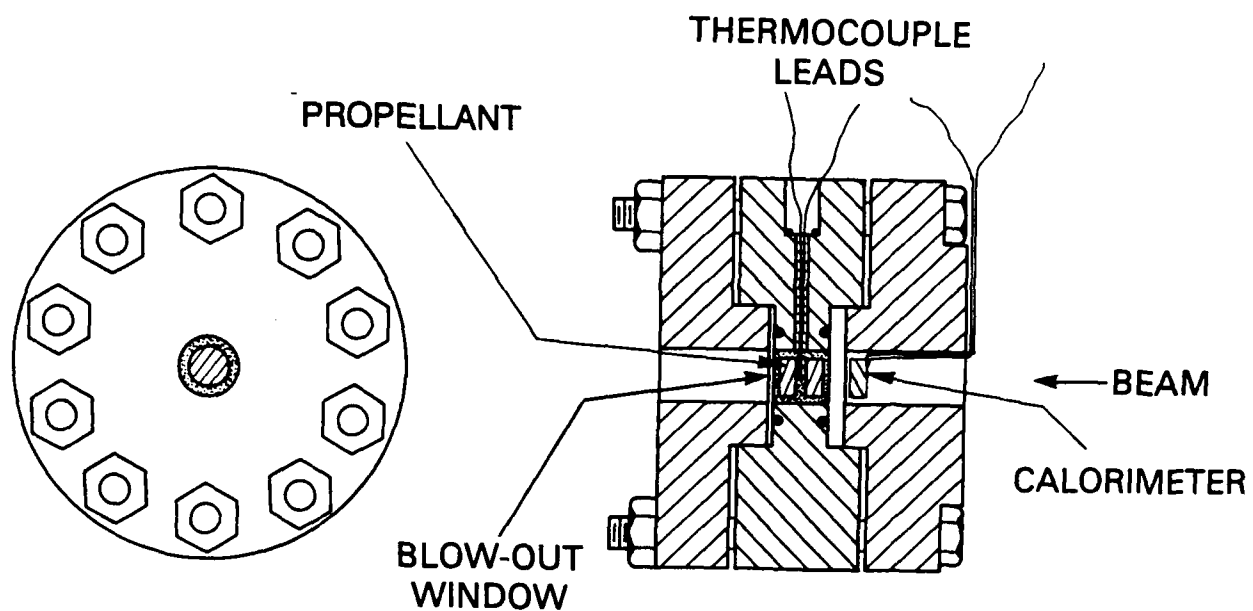


Figure 1. Front and side views of confinement cell for thermal initiation experiments on solid propellants with an electron beam. The outer diameter of the cell is 7.0 cm.

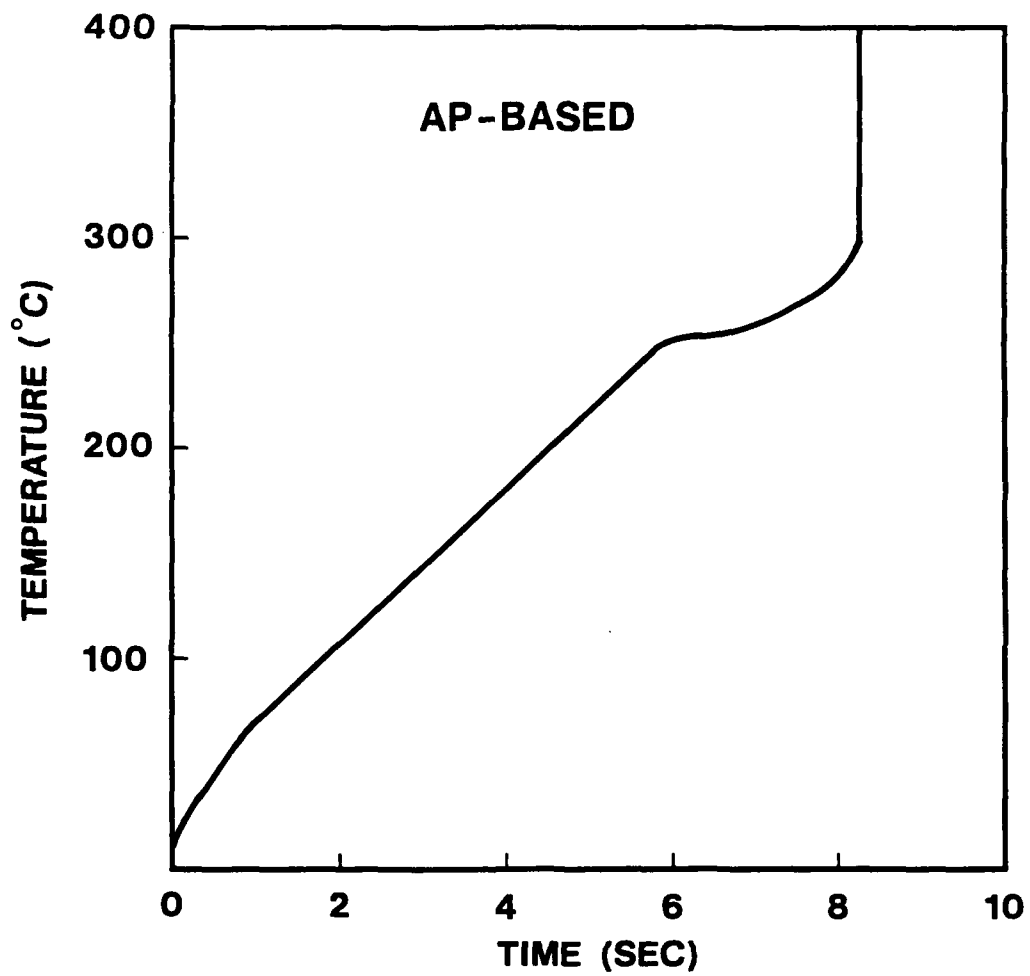


Figure 2. Thermal behavior of a composite propellant with AP as its major ingredient when uniformly heated to explosion by an electron beam.

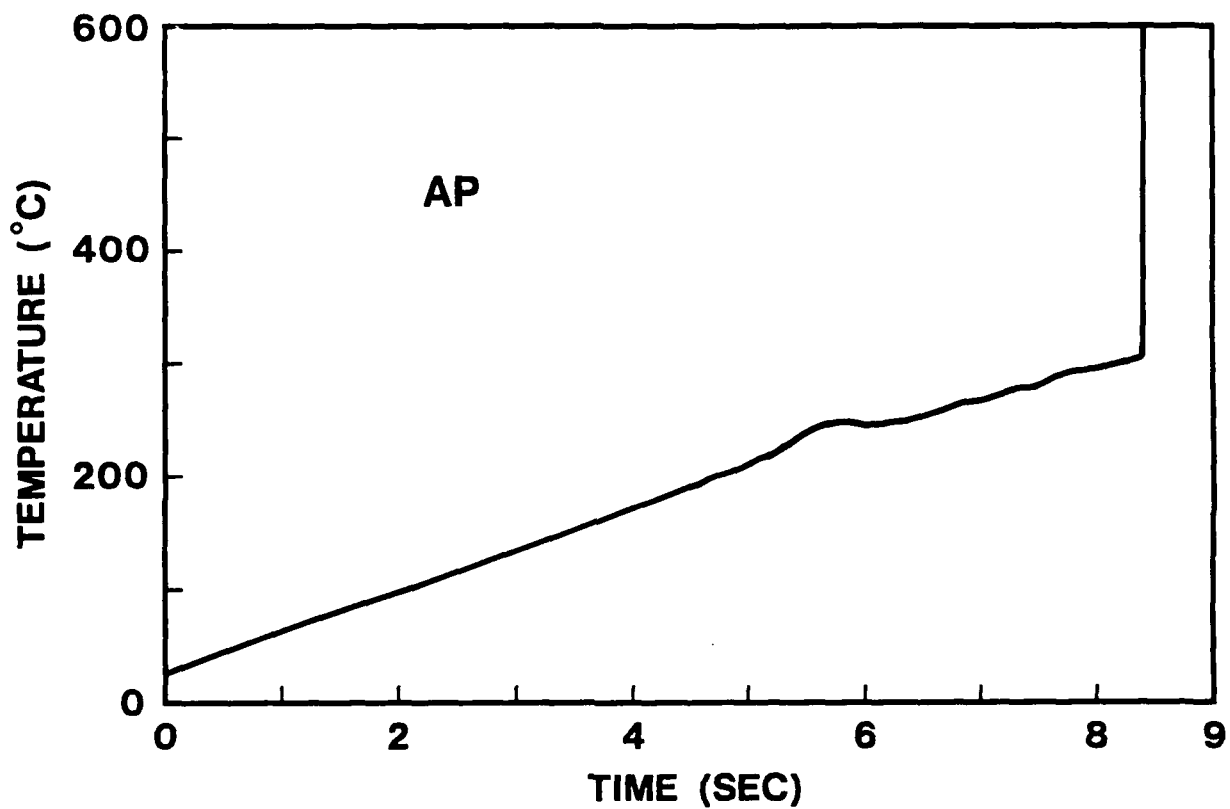


Figure 3. Thermal behavior of pure AP heated uniformly by an electron beam to explosion.

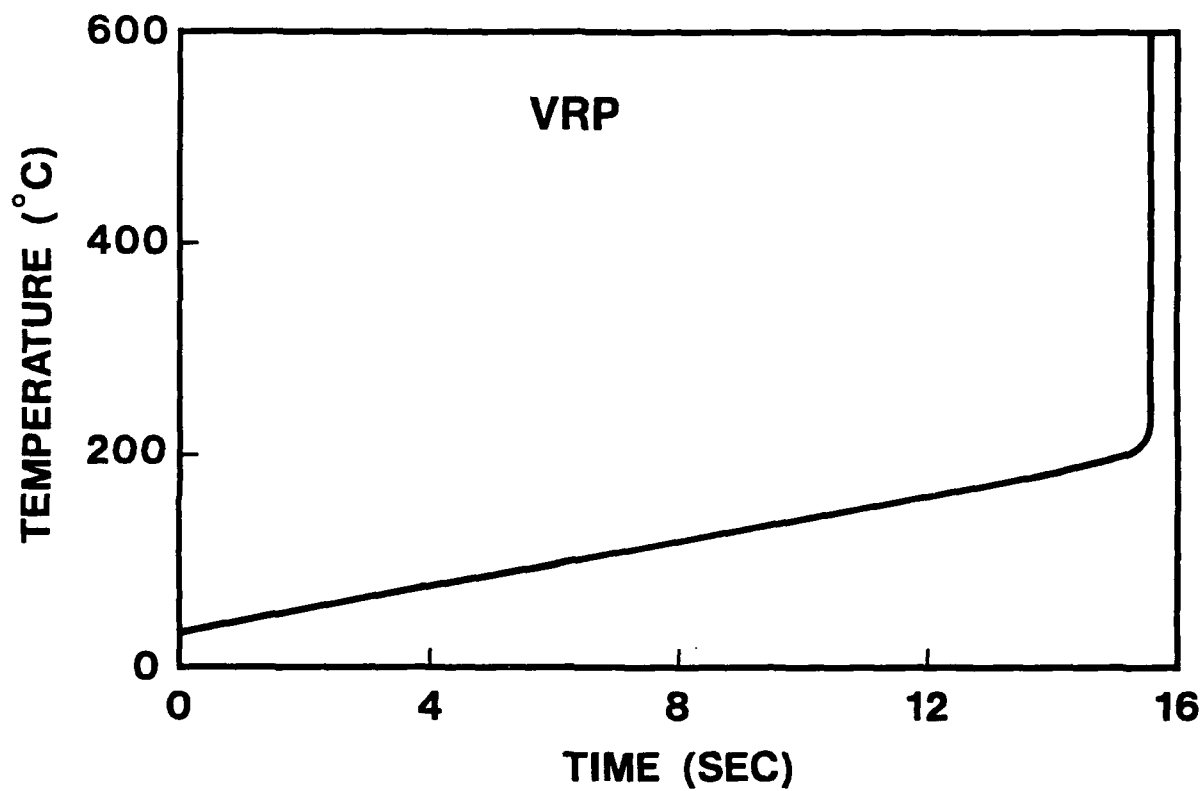


Figure 4. Thermal behavior of VRP propellant heated uniformly and slowly to explosion by an electron beam.

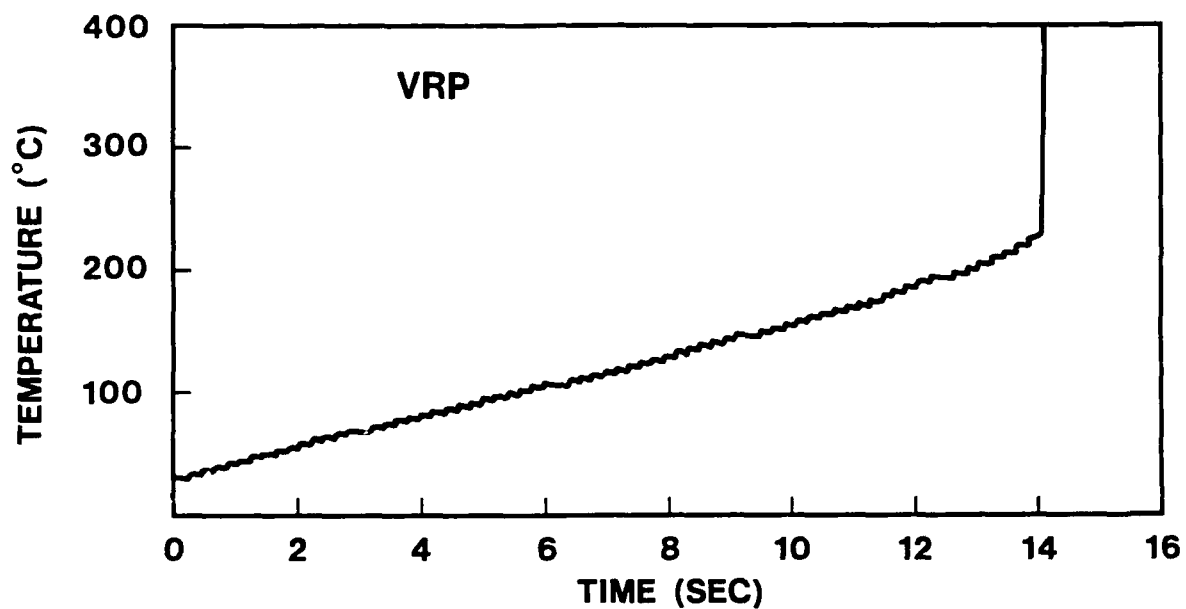


Figure 5. Thermal behavior of VRP heated uniformly and slowly to explosion by a proton beam at the REF. The effect of the individual pulses, at 5 Hz (every 16th pulse missing) can be seen as a staircase pattern.

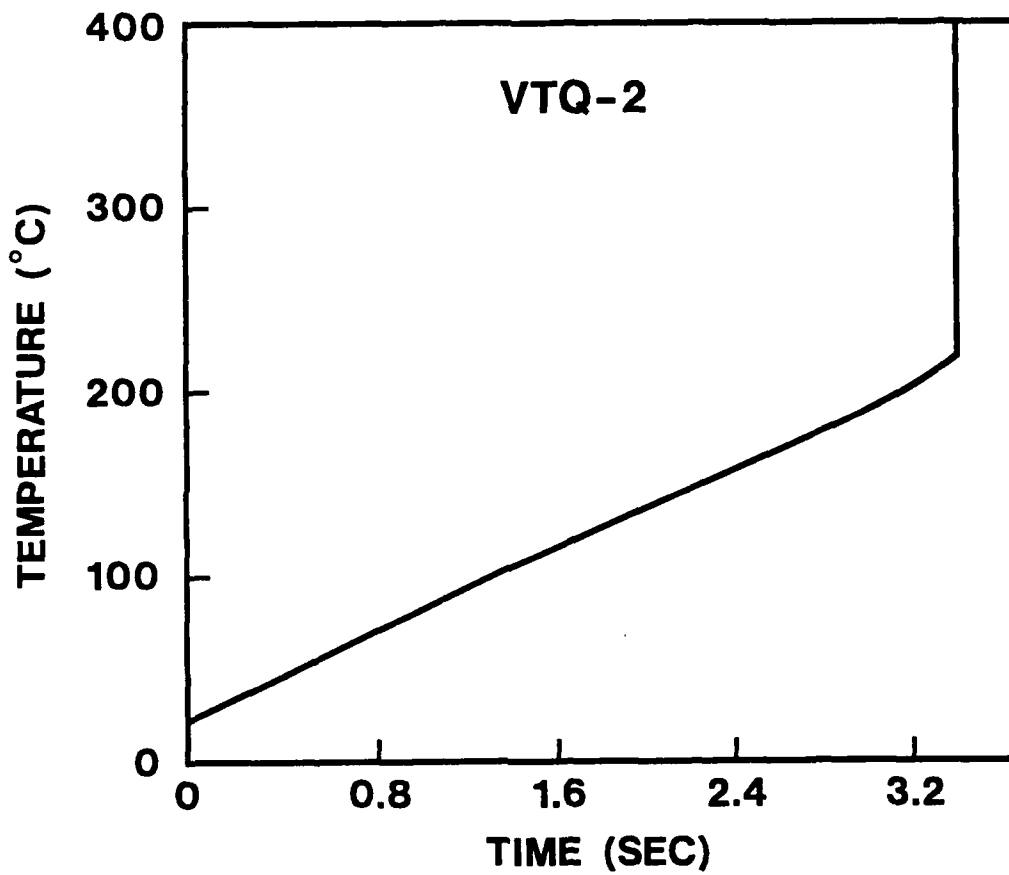


Figure 6. Thermal behavior of VTQ-2 propellant heated uniformly and rapidly to explosion by an electron beam.

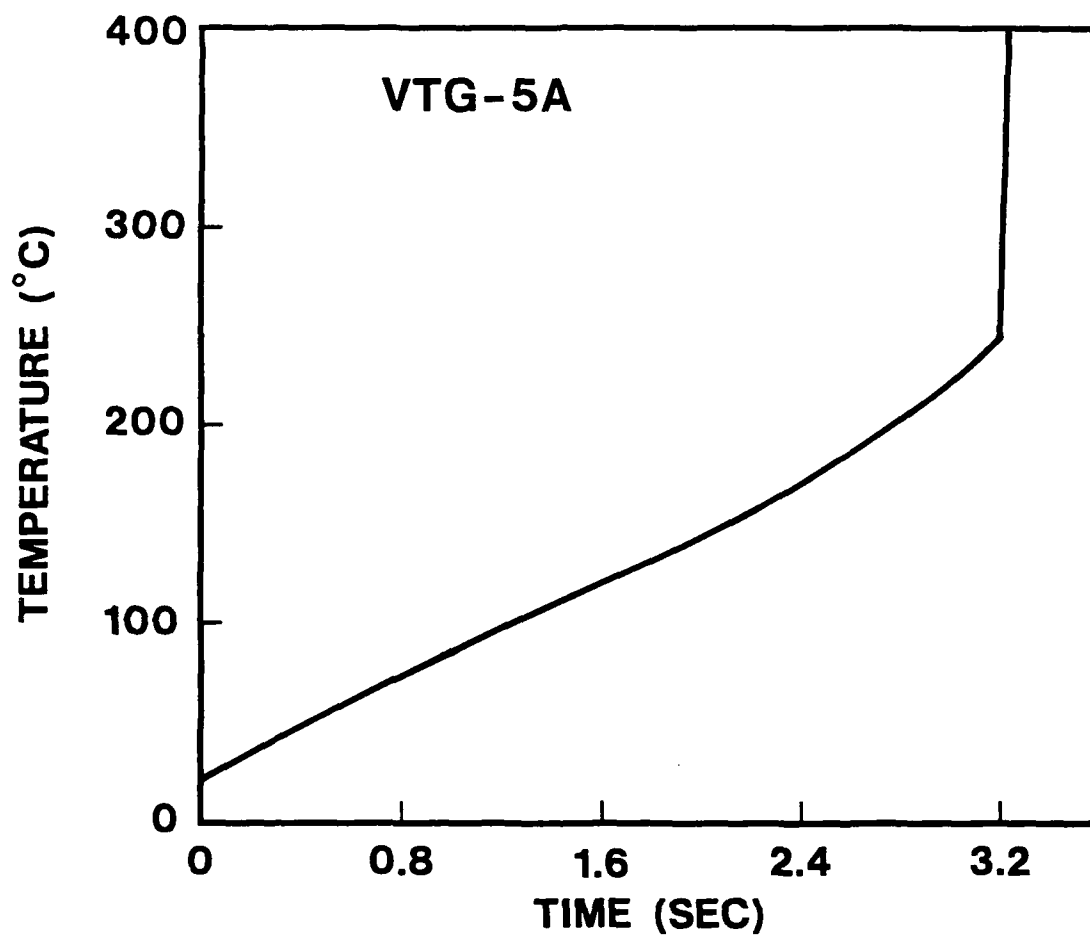


Figure 7. Thermal behavior of VTG-5A propellant heated uniformly and rapidly to explosion by an electron beam.