Heating in crystalline solids due to rapid deformation

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We present experimental evidence that during rapid deformation of at least some crystalline and polymeric solids heat is generated at unexpectedly high temperatures beginning essentially at the moment deformation begins. The heat was observed by fast infrared sensors. Both the initial infrared emissions and the apparent blackbody temperatures were far in excess of what can reasonably be accounted for by the conventional picture that distributes the energy due to deformation over the bulk of the solid. It appears that the origins of these high temperatures are associated with mechanical processes that effectively concentrate the energy of deformation into small local regions within the sample. These local hot spot regions are most likely associated with shear bands and possibly fracture sites in the deforming crystal.

I. INTRODUCTION

It is well known that materials undergo heating during impact due to the plastic work done on them by the impactor. The classical theory would distribute this energy in some manner throughout the bulk of the sample. The temperature of the sample would increase with the amount of plastic work done on the sample by the impactor.

Here we present experimental evidence that shows that relatively high-temperature infrared emissions begin immediately on impact for at least several different crystalline and polymeric solids. Both the emission level and the apparent blackbody temperatures recorded within a few microseconds of initial impact are far in excess of what can reasonably be expected from a bulk process.

It has long been necessary to postulate the existence of an energy localization process in order to explain the sensitivity of explosives to low-level impact.[1,2] Almost certainly the same processes occur in inert crystalline solids, but because of their nonreactive nature the existence of small localized hot spots is of little consequence and not an obvious feature in such materials. However, in recent years there has been an increasing interest in accounting for the appearance of shear bands and fracture in inert materials that have undergone rapid deformation from impact or shock.[3,4] The deformation is often found to be localized in shear bands which consequently are the sites of plastic work and therefore must become hot. The mechanisms responsible for this energy localization and shear band formation have been the object of considerable research.[5-9] It has been shown that hot spot formation during impact of explosive materials occurs mainly in the region of high-rate shear deformation.[10] In much the same way, the occurrence of a heterogeneous ignition process in explosives that have been subjected to high strength shocks is indicative of a hot spot forming process.[11]

Time-resolved temperature measurements of shocked nonenergetic materials by infrared radiometry in the low-pressure range have become experimentally feasible and brightness[12,13] and two-color[14,15] temperatures of shock materials have been reported. In addition, a report[16] describing the use of this technique to investigate reactive shocks in explosive materials to provide a new and versatile diagnostic tool in the study of initiation and buildup to detonation has also been published. The latter experiments resulted from shock-induced heating in explosives. We will show in this report that shock conditions are not necessary to observe deformation-induced heating. We will show that induced emission occurs at the moment of impact with loading rates as low as 5 m/s.

II. EXPERIMENTS

An impact machine was used to investigate the deformation-induced heating for low loading rates of 5–15 m/s.

The impact machine, shown in Fig. 1, consists of a reasonably well-guided drop weight with a mass of 0.5 kg and a maximum free-fall drop height of 1.5 m. Through the use of an additional accelerating device, the equivalent free-fall drop height can be increased to approximately 20 m for the 0.5 kg drop weight mass. The anvil, shown in Fig. 1, consists of a sapphire cylinder 1.905 cm in diameter by 1.27 cm high cut from a single crystal. The ends of the sapphire cylinder were polished to optical flatness. The sapphire anvil transmits both visible and infrared emissions out to wavelengths of approximately 6 μm. For longer wavelengths silicon anvils were used, but these results will not be discussed here. Directly below the anvil was a front-faced silvered mirror mounted at a 45° angle to the vertical. As shown in Fig. 1, the infrared emissions were directed through a series of infrared lenses, a beam splitter/filter, and brought to focus on two infrared detectors. The detectors were either the Barnes Engineering Company type A-600 detector or the New England Nuclear Company HgCdTe detector. Both of these infrared detectors were liquid nitrogen cooled. The response time of the New England Nuclear units was approximately 5 ns while that of the Barnes was about 20 ns. The beam splitter/filter passed a different infrared band to each detector. By assuming a black- or graybody spectrum, an estimate of the temperature[7] of the infrared source could be obtained by taking the ratio of the signal amplitudes from both detectors.
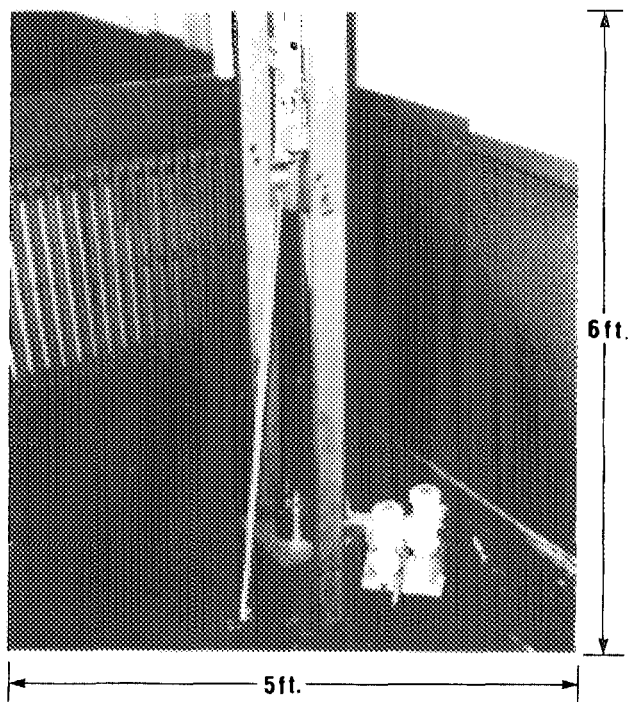
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FIG. 1. Impact machine. Includes guided dropweight of 0.5 kg and maximum free-fall height of 1.5 m and 5.5 m/s velocity. Slings increase the effective height to 20 m and velocity to 18.5 m/s. The sapphire anvil and IR detectors are also shown.

The signals from each detector were passed through fast, broadband amplifiers and recorded simultaneously on dual-beam digital oscilloscopes. Also recorded simultaneously on a separate dual-beam oscilloscope were the signal from an accelerometer and the signal from one of the infrared detectors. The accelerometer was mounted on the drop weight, 3.57 cm from the impact surface, and was used to monitor the deceleration of the drop weight during impact. Because of the distance between the accelerometer and the impact surface, the accelerometer signal was delayed 6.5 μs with respect to the infrared signal. The time resolution of the experiment was limited by the response of the accelerometer which was measured to be approximately 1 μs.

Both before and after a series of impact experiments the infrared detectors were calibrated with a blackbody source and chopper wheel placed against the sapphire anvil in approximately the same position as the material to be impacted. The calibration of the accelerometer was periodically checked throughout the duration of the experiments.

Using the above apparatus, impacts were conducted on NaCl, AP (ammonium perchlorate), RDX (cyclotrimethylene trinitramine), PETN (pentaerythritol tetranitrate), and Teflon, as well as composite materials. The NaCl, AP, RDX, and PETN samples were single crystals approximately 1.5 mm on a side and with masses of about 20 mg. The Teflon samples were in the form of pellets with a mass of approximately 70 mg each. AP, RDX, and PETN are energetic materials which can react violently under impacts of sufficient severity. This did not occur in the data reported here because the drop weight was always prevented from

totally crushing the sample by the presence of metal stops which halted the downward motion of the impactor approximately 0.5 mm above the surface of the anvil. In those cases where reaction was allowed to occur, generally when the stops were removed, the resulting infrared emissions were very different from those when no chemical reactivity occurred; the infrared emissions were very large, with typical apparent blackbody temperatures of 10^3 °C before saturation occurred in the detector-amplifier systems. What is of main concern here is the response of these energetic crystals to subinitiation impacts and this much more closely resembles the heating that occurs in the inert materials NaCl and Teflon. This is in keeping with the hypothesis that at early times after the start of a mild impact, $t < 50\,\mu$s, the response of energetic materials is very likely to be similar to that of inert substances. It is only after the temperature in a local hot spot equals or exceeds the ignition threshold that chemical reaction occurs.

A. Blackbody assumption

Before further discussion of the experimental results, we will briefly discuss the blackbody assumptions used to determine sample hot spot temperature. The response of an infrared detector from blackbody emission may simply be described by

$$W^{obs}_{\Delta\lambda} = G \int_{\Delta\lambda} e(\lambda) W^{bb}(\lambda, T) d\lambda ,$$

where W^{obs} is the observed radiance from the wavelength interval $\Delta\lambda$ obtained from the voltage response of the detector; G is a constant optical geometrical factor representing the light-gathering capability of the optics, $e(\lambda)$ is the emittance, and $W^{bb}(\lambda, T)$ is a blackbody function with the detector responsivity and optical component wavelength dependence incorporated into it.

A brightness temperature for any given experiment may be obtained by calibrating a single detector with a known blackbody source, as long as it is assumed that G remains constant; that the emittance $e(\lambda)$ is the same for both the blackbody source and the sample in question, and of course that the sample emits black- or graybody radiation. By simultaneously measuring the emission using two detectors at two different wavelength intervals, a two-color temperature may be determined. This derived temperature represents an estimate of the true temperature, independent of $e(\lambda)$, provided that $e(\Delta\lambda_1) \simeq e(\Delta\lambda_2)$ and provided that the emitted radiation follows a blackbody function. With the above assumptions, it is possible to evaluate $e(\lambda)$ by comparing the brightness temperature and the two-color temperature. This quantity $e(\lambda)$ is a direct measurement, then, of the fraction of the surface of the sample, as seen by the detector, which is hotter than the surrounding material. This quantity, thus, represents the fraction reacted or fraction hot of the sample being observed. In the impact experiments described here, this becomes an important quantity in that the origin of the induced observed emission relates directly to the mechanism for hot spot formation and growth.

The errors in the temperature measurements, as described here, arise from two main sources. First, experimen-
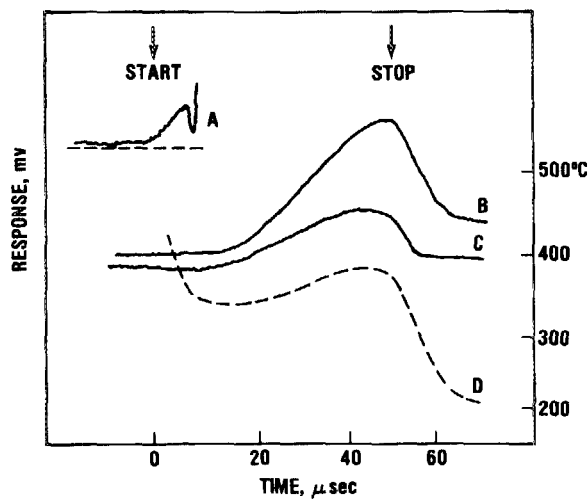
FIG. 2. Infrared emission and two-color temperature of NaCl due to 18.5 m/s impact. Curve (a) is the accelerometer response, (b) and (c) are, respectively, the $> 5.5 \mu m$ and $< 5.5 \mu m$ detector responses, and (d) is the calculated temperature.

FIG. 4. Infrared emission (a) and accelerometer response (b) of RDX impacted with 5.5 m/s velocity.

tal error can result from the nonreproducibility of optical alignment between the calibration and sample runs and also from the signal-to-noise levels of the observed signals. Second, error can arise from the assumptions being used in the blackbody model. For example, in the brightness temperature determination, if the sample acts like a gray body with $e(\lambda)$ less than 1.0, the observed temperature will be a lower limit, or will be less than the true temperature. In the two-color temperature measurements $e(\Delta\lambda_1) \approx e(\Delta\lambda_2)$ is generally a good approximation and the observed temperature determination is a true estimate of the actual temperature. When comparing the brightness temperature to the two-color temperature for a given experiment the fraction of sample

hot or reacted, $e(\lambda)$, also represents a lower limit to the true fraction of sample surface heated. Furthermore, a very important factor that can cause error will be the deviation from blackbody behavior that results from selective molecular emission.

B. Discussion of results

Returning to the experimental results, Figs. 2–5 show the infrared and accelerometer records of impacts on single crystals of NaCl, AP, RDX and PETN, respectively. Figure 6 shows a similar set of records from an impact on a disk at Teflon. In these, as in all of the experiment of this type that we have run to date, the infrared emissions appear essentially simultaneously with the first indication of deceleration from the accelerometer (taking into account the 6.5-μs delay in the arrival of the impact stress wave at the accelerometer). Of particular significance is the immediate appearance of heat at a temperature above the threshold level of about 30 °C which can be detected by the infrared sensors as they
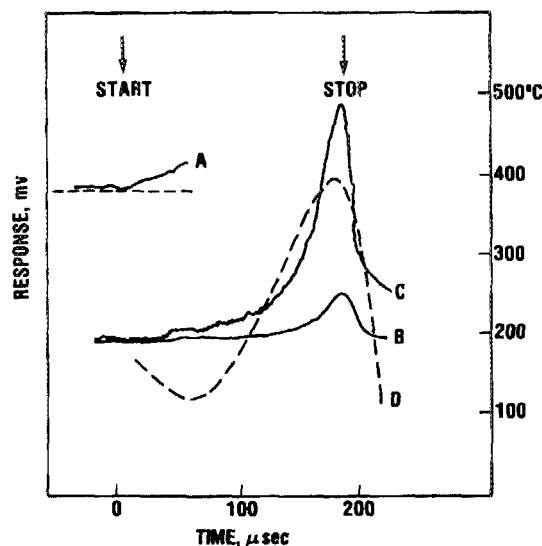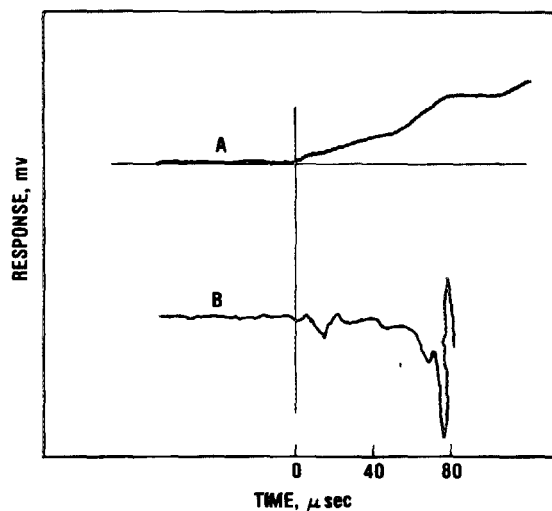

FIG. 3. Infrared emission and two-color temperature of AP due to 5.5 m/s impact. Curve (a) is the accelerometer response (b) and (c) are, respectively, the $> 5.5 \mu m$ and $< 5.5 \mu m$ detector responses, and (d) is the calculated temperature.
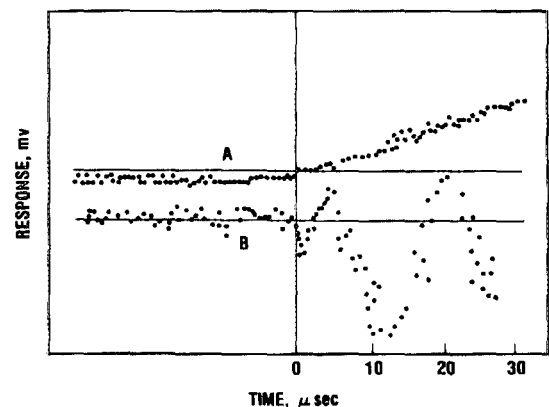

FIG. 5. Infrared emission (a) and accelerometer response (b) of PETN impacted at 5.5 m/s velocity.
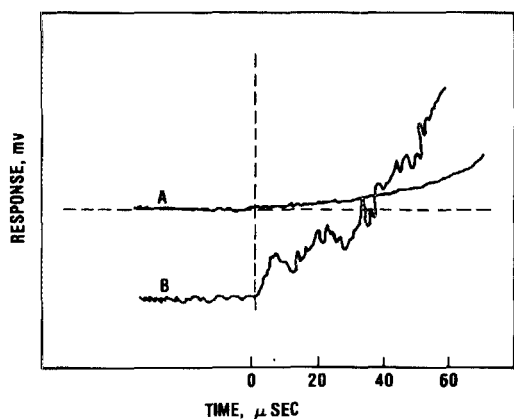
FIG. 6. Infrared emission (a) and accelerometer response (b) of Teflon impacted with 5.5 m/s velocity. (Note that infrared emission occurs within 1 μs of impact.)

are used in our system. For RDX, Fig. 4, the change in velocity of the drop weight in the first 15 μs after impact (194.5 μs) is approximately $\Delta V = 2 \times 10^{-3}$ m/s. The change in kinetic energy of the falling drop weight is

$$\Delta U = \tfrac{1}{2} M \left[V_0^2 - (V_0 - \Delta v)^2 \right] \approx M V_0 \Delta V.$$

For the case at hand $V_0 = 5.54$ m/s and $M = 0.5$ kg, so that $\Delta U = 5.54 \times 10^{-3}$ J.

If all of the above energy were to be distributed over the bulk of the crystal, the temperature increase would only be $\Delta T = \Delta U / mc_v = 0.2$ °C, where $c_v = 0.3$ cal/g °C and $m = 21$ mg. This temperature increase is far below that can reasonably be detected by our infrared sensors. Further, as shown in Figs. 2 and 3, the typical two-color blackbody temperature is of the order of 200 °C and this is far above what the conventional calculation would predict for the heating at this early time. This result indicates that the energy of deformation is being localized. Just to bring the local energy density up to a level where the temperatures begin to exceed the threshold level of the detectors requires that this local volume occupy approximately 1/150 at the impacted crystal. To achieve energy densities and temperatures approaching the apparent blackbody temperature requires that the local volume be approximately 1/1000 of the volume of the impact crystal.

III. SUMMARY OF RESULTS

The results of this study clearly indicate that the use of time-resolved infrared radiometry is a viable diagnostic tool for the investigation of rapid deformation effects on inert and energetic materials. The data presented here demonstrates a definite correlation between the pressure-time profile and the infrared emission time profile of impacted materials. This technique can be used to probe the origins of heat development in materials under deformation and can be used in the elucidation of the mechanisms involved in predetonation and deformation phenomena.

The results reveal that infrared emissions occur in at least several different crystalline and polymeric materials under impact in a drop weight apparatus to within a few microseconds after the impact begins. The observed time resolution is limited only by the timing devices of the apparatus and the amplitude of the signals. The results of this study also shows that by using a model for blackbody radiation, two-color temperature-time profiles may be obtained from the experimental data. Clearly, the results here appear to be in contradiction to current conventional models that describe the energy buildup in materials under deformation prior to chemical reaction. Further, it is possible to determine, with the model, some knowledge about the fraction of sample emitting heat during deformation.

The determination of temperature and of fraction of sample heated depends on the blackbody assumption. A series of experiments are planned for the near future to verify this assumption by using an infrared array detector to measure the spectrum of the emitted radiation, and to measure the spatial evolution of heat from the sample.

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