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Steps toward thin film metal thermistors with microsecond time response for shock temperature measurements of polymers

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Abstract. Equations of state can be used to predict the relationship between pressure, volume and temperature. However, in shock physics, they are usually only constrained by experimental observations of pressure and volume. Direct observation of temperature in a shock is therefore valuable in constraining equations of state. Bloomquist and Sheffield (1980, 1981) and Rosenberg and Partom (1984) have attempted such observations in poly(methyl methacrylate) (PMMA). However, their results disagree strongly above 2 GPa shock pressure. Here we present an improved fabrication technique, to examine this outstanding issue. We make use of the fact that the electrical resistivity of most metals is a known function of both pressure and temperature. If the change in resistance of a thin metal thermistor gauge is measured during a shock experiment of known pressure, the temperature can be calculated directly. The time response is limited by the time taken for the gauge to reach thermal equilibrium with the medium in which it is embedded. Gold gauges of thickness up to 200 nm have been produced by thermal evaporation, and fully embedded in PMMA. These reach thermal equilibrium with the host material in under 1 µs, allowing temperature measurement within the duration of a plate impact experiment.

1. Introduction

The equation of state of a material provides a relationship between its thermodynamic state variables. In this paper, the variables of interest are pressure, density and temperature. In shock physics experiments, pressure and density are routinely measured or inferred from mass and momentum conservation [1–4]. Temperature, meanwhile, is usually inferred using a physically-based equation of state. Many such equations of state exist. However, they frequently disagree in their temperature predictions [5]. Direct temperature measurements are therefore valuable to constrain these equations of state. Polymers are of particular interest, as they are used as binders in polymer bonded explosives (PBXs). In this application heating of the binder during mechanical loading could cause reaction. Prediction of this heating is therefore useful for designing PBX systems. This requires a well-constrained, validated equation of state, and therefore direct temperature measurements.

Several techniques for shock temperature measurement exist. Pyrometry uses the radiation emitted by hot objects [6]. This requires knowledge of the emissivity of the sample, which usually changes during the experiment. It also requires that the sample emit enough radiation to study. For temperatures below 1000 K, black-body emission is quite dim. This imposes a long integration time on any pyrometric apparatus, which is incompatible with a shock experiment.

Raman spectroscopy can also be used for thermometry [7]. The ratio between the stokes and anti-stokes scattering intensities provides the temperature by a simple population-of-states argument. However, only materials with Raman peaks at shifts below 500 cm^{-1} provide sufficient anti-stokes intensity for this technique to be useful below 1000 K. The technique also requires that the material lack electronic transitions near the frequencies studied.

Two further, related techniques involve embedded gauges. Metals vary their resistance with temperature, and thermocouples produce a voltage dependent on their temperature. Both thin metallic foils [8–10] and thermocouples [11] have been embedded in polymers to measure shock heating. The time resolution of such gauges will be limited by heat conduction from the sample into the gauges [12]. Interestingly, Rosenberg and Partom [9] found different results for the temperature rise in poly(methyl methacrylate) (PMMA) shocked past 2 GPa to Bloomquist and Sheffield [10]. These inconsistent results make PMMA an interesting test material. The use of embedded resistive gauges was selected as the most promising means of investigating shock heating in PMMA.

2. Method

A gold measurement grid, 150 nm thick by 10 mm in diameter, was deposited on PMMA by thermal evaporation. The PMMA substrate was then thermally welded to another layer of PMMA, fully embedding the measurement grid. The grid resistance was then probed using a four-wire measurement during shock loading of the PMMA. Temperature was recovered using the temperature coefficient of resistance for gold, after applying a correction for the effect of pressure on resistance.

2.1. Gauge fabrication

PMMA plates, 70 by 70 mm square by 10 mm thick, were fully strain-relieved by thermal normalization. The process consisted of heating the plate above its glass transition temperature, then cooling it to room temperature slowly enough that no thermal stresses developed. Initial experiments found that, without this normalization, residual stresses from PMMA fabrication disrupted the gauge.

The normalized plate was covered with an acetate mask (figure 1) and coated with gold using an Edwards 306A thermal evaporator. Copper read-out legs, 4 mm wide by 25 µm thick, were attached to the square pads of the measurement grid using silver dag, a conductive paint. A second normalized PMMA plate was placed over the gauged plate, and the two were clamped between aluminium blocks with a force of 3 kN. The clamped plates were then thermally welded in a procedure similar to the thermal normalization process: the plates were heated above their glass transition temperature, then cooled to room temperature. The result was a 150 nm thick gold resistor, fully embedded in PMMA and ready to be tested. Figure 3 shows such a gauge.

Any air trapped between the gauge and the PMMA would render it effectively useless. The thermal conductivity of air is approximately a tenth that of PMMA. Since the limiting factor on the gauge's time resolution is the rate at which heat can enter the gauge from the surrounding PMMA, even a micron-thick layer would reduce the time resolution of the gauge by a factor of ten in a steady-state approximation. Worse, the high compressibility of air would result in a temperature rise of a few thousand kelvins in this layer. This temperature rise would obscure that in the PMMA, and may damage the gauge.

Happily, the low stiffness of the PMMA during the thermal welding process, together with the applied clamping force, made exclusion of air simple. Provided no dust grains were present on the gauge surface, no trapped air was observed on the gauge. Visual inspection of the gauge was sufficient to detect and reject gauges in which dust had allowed a bubble to form over the gauge region.

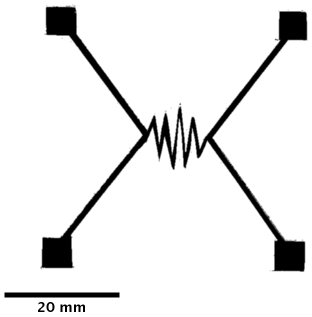

Figure 1. Shape of temperature-measurement grid. Dark areas received gold coating.

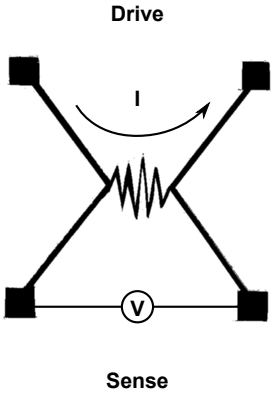

Figure 2. Circuit used to record resistance of measurement grid. V is the 1 MΩ input of an oscilloscope. I is provided by an LM334 constant-current source.

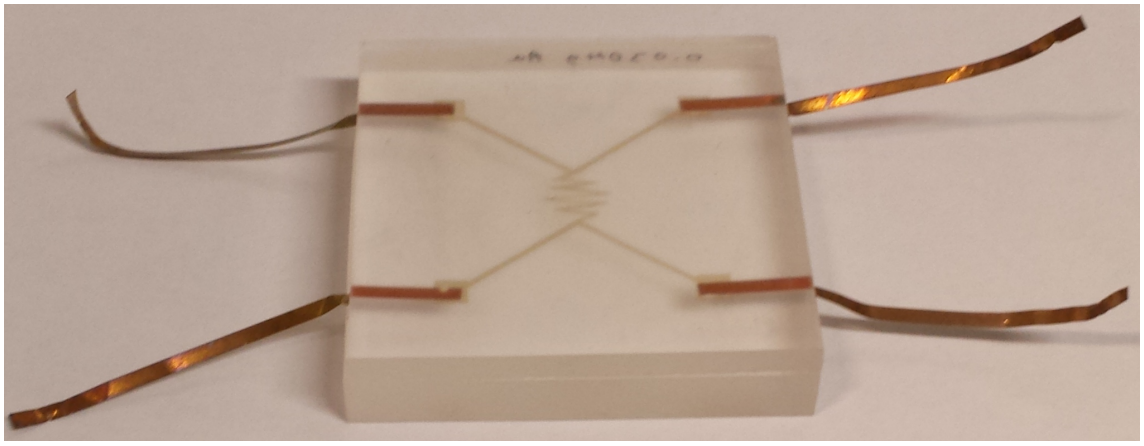

Figure 3. Example of a finished gauge. Side length of PMMA plate is 70 mm.

To assess the thickness and reproducibility of the resistor, several grids were studied immediately after coating with gold. An atomic force microscope was used to measure the thickness of the gold film. All were found to lie in the range 100 nm to 200 nm.

To calibrate the temperature coefficient of resistance, a finished gauge was placed in an oven along with a thermocouple. The resistance of the gauge was compared with the thermocouple output as the oven was heated from room temperature to 80 °C then allowed to cool, yielding a temperature coefficient of resistance of $3.42 \pm 0.01\,\mathrm{K}^{-1}$, which is within the range of values generally accepted for bulk gold.

The gauge thickness was chosen based on a theoretical treatment of heat conduction for the case of a one-dimensional plate in contact with two semi-infinite blocks of material [12]. According to this, a 150 nm thick gold foil embedded in PMMA will reach 85 % of the temperature in the PMMA within 700 ns.

2.2. Circuit design

The readout legs of the measurement grid were formed from the same 150 nm gold as the grid itself. Their resistance was therefore non-negligible. They also passed outside the region

of one-dimensional strain generated by the plate-impact experiment. Their resistance would therefore be expected to vary unpredictably throughout the experiment. To eliminate the variable resistance of the readout legs, a four-wire resistance measurement was performed on the grid.

A constant current was passed through the measurement grid using two of the four readout legs (the "drive" legs). The other two legs (the "sense" legs) were connected to a 1 MΩ terminated oscilloscope. The high input impedance of the oscilloscope ensured that negligible current flowed through the sense legs. The voltage recorded by the oscilloscope was therefore due solely to the voltage drop as the drive current passed through the measurement grid. Figure 2 shows the layout used. Current was supplied by a battery-powered LM334 constant-current source.

2.3. Plate impact

The gauge was shocked using the Cavendish plate impact facility [13]. This is a 50.8 mm light-gas gun. Aluminium plates, 10 mm thick by 48 mm diameter, at a velocity of $650\,\mathrm{m\,s^{-1}}$, were selected, which gave a stress of 2 GPa in the PMMA target. This stress was selected as the highest value for which Rosenberg and Partom [9] agreed with Bloomquist and Sheffield [10] on the shock heating of PMMA. It was therefore appropriate for testing this technique. A photonic Doppler velocimetry (PDV) system [4] was also used to monitor shock arrival at the gauge.

2.4. Data reduction

The temperature coefficient of resistance for the gauge at ambient pressure was established in section 2.1. However, pressure also affects the resistance of metals [14]. We make the approximation

$$\Delta R = \left.\frac{\partial R}{\partial P}\right|_{T=T_0} \Delta P + \left.\frac{\partial R}{\partial T}\right|_{P=0} \Delta T \ , \tag{1}$$

i.e. that the effects of temperature and pressure on the gauge resistance are independent [9]. The effect of pressure on resistance for gold has been measured in the case of hydrostatic loading [14]. To recover the pressure coefficient of resistance under the uniaxial strain imposed by a shock experiment, a correction must be applied. This correction is given by

$$\ln\left(\frac{R}{R_0}\right)_{\mathrm{1D}} = \ln\left(\frac{R}{R_0}\right)_{\mathrm{HS}} - \frac{2}{3}\ln\left(\frac{V}{V_0}\right)_{\mathrm{HS}} \ , \tag{2}$$

where the subscripts 1D and HS denote uniaxial and hydrostatic strain, respectively [2]. Values of $\frac{R}{R_0}$ and $\frac{V}{V_0}$ are tabulated [14].

After eliminating the effects of pressure in this way, the temperature may be recovered from the resistance using the known temperature coefficient of resistance, α:

$$\Delta T = \frac{1}{\alpha}\frac{\Delta R}{R_0} \ , \tag{3}$$

where R_0 is the resistance at elevated pressure and ambient temperature.

3. Results

Figure 4 shows a temperature-time trace for PMMA, shocked to 2.0 ± 0.1 GPa. Pressure correction has been applied to the entire trace. The shaded region indicates the uncertainty in the shock arrival time at the gauge[1]. A rapid disturbance in the recorded temperature near

[1] The rear face of the gauge package was opaque for this experiment. The PDV system was therefore only able to record shock arrival time at the rear surface of the sample. Shock arrival time at the gauge was recovered using the known dimensions of the sample and the Hugoniot of PMMA.

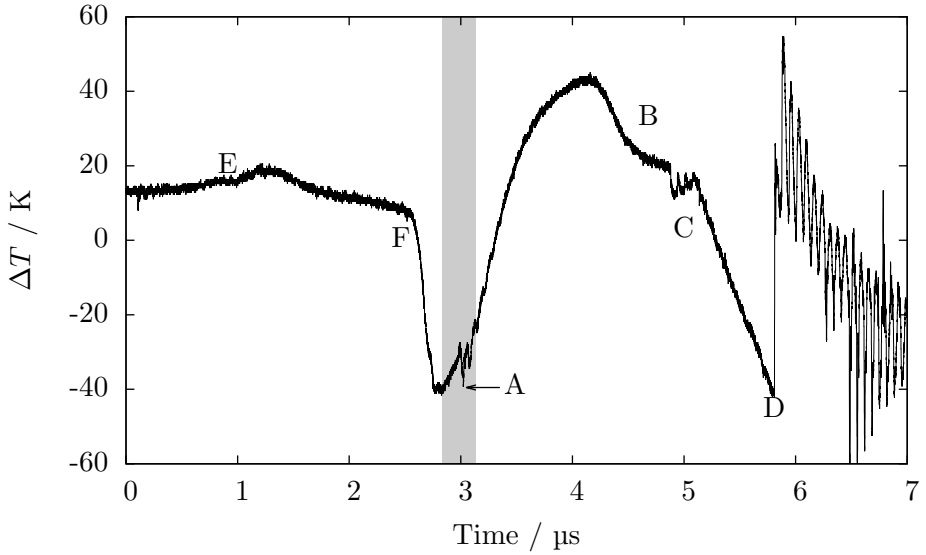

Figure 4. Temperature-time trace for PMMA shocked to 2.0 ± 0.1 GPa. Pressure correction applied to entire trace. Shaded region indicates uncertainty in shock arrival time at gauge, established by reference to shock breakout at back face of sample recorded by PDV. Time is relative to arrival of projectile at sample.

2.1 µs, labelled A, is very probably due to arrival of the shock at the gauge. The increased pressure would decrease the resistance of the gauge more rapidly than any thermal effects. Subsequent small oscillations show the response of the LM334 constant-current source to this disturbance, with stability regained within 500 ns.

Following this disturbance, the recorded temperature rises. The shape of the rise is broadly as expected for thermal equilibration. Following this, labelled B, there is an unexplained fall in reported temperature. Thereafter the release fan reaches the gauge, eventually causing its failure, at C and D respectively.

In addition to the unexplained fall at B, there are two features before the arrival of the shock wave. The first, labelled E, corresponds to the arrival of the projectile at the front face of the sample. It is possible that residual air in the sample chamber is trapped between the projectile and the sample. Compression of this air would produce a short-lived high-temperature region at the front of the sample. This may have coupled radiatively to the gauge, raising its temperature.

The second interesting pre-shock feature, labelled F, is a rapid drop in recorded temperature a few hundred nanoseconds before arrival of the shock at the gauge. The most likely explanation for this feature is that the shock wave is tilted due to misalignment of the target. A tilted shock wave would reach one of the gold/silver/copper junctions at the end of the readout legs before reaching the gauge. This in turn could result in a voltage developing in those junctions, disturbing the temperature measurement.

The reported 40 K temperature rise is in excellent agreement with Bloomquist and Sheffield's thermistor data [10] and lies on the line defined by Rosenberg and Partom's data [9].

4. Conclusions

We have demonstrated a methodology for producing very thin thermistors of pure metals which are fully embedded in PMMA host material without the need for an adhesive layer. Thermistors can measure the full range of temperatures of interest, ambient to 10^3 °C, with a practical level of uncertainty of 5 °C. The thermistors we have produced are sufficiently thin that we anticipate

they can reach thermal equilibrium with their host material within 1 µs. We have chosen to initially study PMMA as a host material because of the extant data in the literature, but it is self-evident that many other materials could be investigated using the same technique. To date our major success has been in the fabrication of mechanically sound target cells, and the initial data are encouraging. The next steps will be to verify the soundness of the electrical circuit in the face of an instantaneous change of electrical impedance, possibly with the incorporation of a differential amplifier physically close to the target cell. We anticipate that thin-film thermistors represent a useful tool for constraining the equation of state of a wide range of materials of interest.

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