DYNAMIC MEASUREMENTS OF THE GRÜNEISEN COEFFICIENTS OF TANTALUM, URANIUM, AND URANIUM - MOLYBDENUM ALLOYS

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The purpose of this work¹ was to develop a technique for making dynamic measurements of the Grüneisen coefficients of high atomic number materials and then use this technique to measure the Grüneisen coefficient of uranium, uranium alloyed with 1.5 w/o molybdenum, and uranium alloyed with 10 w/o molybdenum. Tantalum was used as the reference material to develop the procedures for data acquisition and interpretation.

The interest in uranium-molybdenum alloys was precipitated by their utility in the design of fast-burst pulse reactors. In fact, the Grüneisen parameter of the fuel is a most important physical parameter since a design limit of this type of reactor is dynamic fracture of the fuel.²

Pulsed electron beams with maximum electron energies between 2 and 4 Mev have been used to obtain dynamic measurements of the Grüneisen coefficients of low and intermediate atomic number materials.³⁻⁶ However, as the atomic number of the irradiated material increases, the electron range decreases and approaches the product of the time duration of the beam and the acoustic wave velocity of the material. Under these conditions, the material experiences an appreciable reduction in stress from that which would be attained if the energy deposition were instantaneous. Therefore, the main problem to be overcome in developing the measurement technique was to properly account for the time duration of the electron beam. Perry⁷ has overcome this problem for measurements on Ta by selecting 50 nanoseconds as the effective deposition time characteristic of the Febetron 705.

Under the conditions of one-dimensional strain, we have shown that the maximum compressive stress, σ , that would propagate to the interface of an irradiated isotropic, elastic material and a quartz gage is

$$\sigma = \frac{\gamma T \rho}{2CT} \int_0^{CT} e(x) dx ,$$

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where γ , ρ , and C are the Grüneisen coefficient, density, and longitudinal elastic wave velocity, respectively, of the irradiated material,

$$T \equiv \frac{2QV}{QV + \rho C} ,$$

where Q and V are the density and longitudinal elastic wave velocity of the quartz and $e(x)$ is the spatial dependence of the change in thermal energy per unit mass. It has been assumed that the thermal energy of the irradiated material is changing uniformly in time between 0 and T ; hence, T is defined as an effective energy deposition time. We have shown that (1) this expression for σ is valid for CT less than the thickness over which the thermal energy has been changed; (2) σ occurs at the time L/C where L is the thickness of the irradiated material; and (3) at the time $L/C + T/2$, the interface stress goes to zero. Therefore, quartz

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gage oscilloscope records of the time-dependent interface stress were used to provide measurements of both σ and τ . The spatial dependence $e(x)$ was measured at 1-mil and 3-mil intervals by attaching intrinsic thermocouples to 1 mil and 3 mil Ta and U calorimeter foils.

The Febetron 705 pulsed electron beam was used to irradiate the Ta, U, and U/Mo specimens in a partial vacuum. The pressure and specimen position in the test chamber were selected to provide a uniform energy deposition across the 1-inch diameter of the specimens. One-dimensional strain was maintained during the measuring time by using 0.1-inch-thick specimens and 0.3-inch-guard-ring-diameter quartz gauges. Since the lowest energy electron groups occur both early and late in the pulse, the time duration of the electron beam was varied by preceding the Ta specimens with Ta attenuators ranging up to 9 mils in thickness. Consequently, the average temperature change in the first mil of irradiated surface was varied from 248°C in the unattenuated Ta specimen to 28°C in the Ta specimen that was preceded by a 9-mil Ta attenuator. Under these conditions, the measured effective deposition time varied between 72.6 nanoseconds in the unattenuated beam and 41.9 nanoseconds in the 9-mil Ta attenuated case. These data are shown with the Grüneisen coefficients in Table I.

Table I. Summary of Tantalum Grüneisen Coefficient Measurements

<u>Ta attenuator thickness (mils)</u>	<u>Avg. temp. rise, front, 1 mil (°C)</u>	<u>Effective deposition time (nanoseconds)</u>	<u>Grüneisen coefficient of Ta</u>
0	248	72.6 ± 1.7	1.53 ± 0.11
2	170	65.3 ± 1.9	1.49 ± 0.14
4	114	57.8 ± 2.2	1.43 ± 0.09
6	58	49.0 ± 1.9	1.46 ± 0.13
9	28	41.9 ± 1.3	1.51 ± 0.13

The error bars shown for the effective deposition time represent one standard deviation in the measurements, whereas the error bars shown for the Grüneisen coefficients result from standard deviations of σ and τ measurements.

The consistency that was found in the Ta Grüneisen parameter measurements and their close agreement with the values computed from other thermodynamic parameters (specific heat at constant volume, linear thermal expansion coefficient, and isothermal compressibility of 1.6 ± 0.2 was interpreted as establishing this as an appropriate technique for treating the effective deposition time. This method of interpreting the effective deposition time was subsequently applied to the U and U/Mo alloys.

The uranium specimens were machined from rods which had been poured at the Los Alamos Scientific Laboratory at 1335°C into heated molds whose temperature varied between 750°C and 900°C. The depleted U, depleted U-1.5 w/o Mo, and depleted U-10 w/o Mo rods were removed from the molds after cooling overnight. One disk was then machined from each rod for the Grüneisen coefficient measurements. The depleted U-10 w/o Mo rod was then sealed in an evacuated quartz tube, heated to 950°C for 3 days, and water quenched. This heat treatment was made to ensure that the specimen consisted of only gamma phase. After the Grüneisen coefficient measurements, each of the four specimens were examined by x-ray radiography, x-ray diffraction, and optical metallography.

The 250-keV radiography showed that no voids were detectable within the 2-mil resolution of the equipment. Powder method x-ray diffraction showed that the depleted U and depleted U-1.5 w/o Mo specimens were at least 98-per cent alpha phase, whereas the two U-10 w/o Mo specimens were at least 98-per cent gamma phase. (The measuring equipment required a minimum of 2 per cent of a phase in order to detect it.) Polarized light metallography revealed that the depleted U and U-1.5 w/o Mo specimens had an anisotropic structure, but the 1.5 w/o Mo had reduced the grain size to about 0.05 of that in the depleted U. After cathodic etching the two U-10 w/o Mo specimens were observed optically. In both specimens, some uranium carbide and uranium nitride could be seen, but more clustering of these compounds was noted in the specimen which had undergone the additional heat treatment. There was also a limited amount of U_2Mo precipitation at the grain boundaries of both specimens. The additional heat treatment had doubled the grain size.⁸

The Grüneisen coefficient measurements for these materials are summarized in Table II and compared to values computed from other thermodynamic parameters.

Table II. U and U-Mo Grüneisen Coefficient Measurements

<u>Material</u>	<u>Effective deposition time (nanoseconds)</u>	<u>Grüneisen coefficient (this expt.)</u>	<u>Grüneisen coefficient (thermodynamic)</u>
Depleted U	113.0 ± 2.1	1.65 ± 0.16	1.70 ± 0.15
U-1.5 w/o Mo	89.7 ± 3.5	1.83 ± 0.18	1.97
U-10 w/o Mo	84.4 ± 2.3	1.41 ± 0.11	1.59
U-10 w/o Mo (heat treated)	80.3 ± 2.7	1.47 ± 0.18	1.59

In these experiments there was an increase in the effective deposition time as the atomic weight of the irradiated material increased. An increase from 72.6 nanoseconds in Ta to 89.7 nanoseconds in U-1.5 w/o Mo is consistent with the argument that, as the density of the irradiated material increases, the lower energy electrons are stopped closer to the front surface. Hence, they make a greater relative contribution to the peak compressive stress that propagates to the rear surface and, since these electrons are present early and late in the pulse, one would expect an increase in τ . However, this argument does not explain the large difference in deposition times observed between the depleted U and U-1.5 w/o Mo.

Comparison of the Grüneisen coefficients measured in these experiments to those computed from other thermodynamic data were especially good for the depleted U and within experimental error for the two alloys. It was observed that (1) the gamma phase has a slightly lower Grüneisen coefficient than the alpha phase, and (2) there was no significant difference in the Grüneisen coefficients of the two U-10 w/o Mo specimens, even though the heat-treated specimen had twice the grain size of the untreated specimen.

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