International Journal of Thermophysics, Vol. 17, No. 3, 1996

Measurement of Laser-Produced Plasma Impulses on Molten Metals for Thermophysical Property Determination[1]

Y. W. Kim[2, 3] and C. S. Park[2]

A high-power pulsed laser excitation of a material surface generates a well-separated sequence of plasma, fluid flow, and acoustic events. When the movement of the surface due to evaporation by laser heating is kept in pace with the thermal diffusion front, the ablative mass loss from a solid surface becomes strongly correlated with the thermal diffusivity of the target matter. The other thermophysical properties which figure in this correlation are the mass density, heat of formation, and molecular weight. The functional relationship, which is given in this text for the first time, can be exploited to measure the thermophysical properties. We have now extended such an approach to measurement of the thermal diffusivity of molten specimens by developing a new instrumentation for determining the ablative mass loss due to a single laser pulse. This has been accomplished by combining a facility for controlled generation of a molten specimen and a novel transducer for real-time measurement of the impulse imparted to the molten target by a laser-produced plasma plume. The transducer design, calibration, signal recovery, and method of extracting the mass loss per laser excitation are detailed by comparing the results for metallic specimens in the solid and molten state.

KEY WORDS: impulse transducer; laser heating; laser-produced plasma; molten metal; RF levitator-heater; thermal diffusivity.

1. INTRODUCTION

The dynamics of the interaction of a high-power laser pulse with a condensed-phase target presents many unusual opportunities to study the

[1] Paper presented at the Twelfth Symposium on Thermophysical Properties, June 19–24, 1994, Boulder, Colorado, U.S.A.

[2] Lewis Laboratory, Department of Physics, Lehigh University, Bethlehem, Pennsylvania 18015, U.S.A.

[3] To whom correspondence should be addressed.

0195-928X/96/0500-0713$09.50/0

thermophysical properties of the target specimen [1–4]. This is because the transient thermal states for the surface, vapors driven out of the surface, and the bulk are strongly influenced by the optical and thermophysical properties as well as the elemental composition of the target.

The key requirement for any useful application of the laser-produced plasma (LPP) plume resulting from pulsed laser heating of the condensed-phase target is that the plasma be of the same elemental composition as in the target. In a fundamental way, this requirement provides the basis for highly reproducible plasma production and thus facilitates quantitative measurement and calibration. Our investigation has established the criterion, in the form of a rule of thumb, for achieving the necessary and sufficient condition for satisfying the requirement [5, 6]. Namely, the criterion is that the velocity of the target surface movement due to laser ablation be approximately the same as that of the thermal diffusion front propagating into the bulk. This condition can be achieved by shaping the temporal profile of the pulsed laser power. It follows that for such a representative plasma there exists an explicit relationship between the thermal diffusivity and the amount of the target material ablated during a single laser exposure [3].

In this paper, we present a series of new studies which exploits the relationship for measurement of thermophysical properties of condensed-phase specimens. The key element of the experiment is to determine the total mass of the laser ablated target material through measurement of the impulse imparted on the target in the process. A new transducer has been developed for this purpose. For studies with molten metallic targets, the transducer has been incorporated into a new RF levitator-heater for momentary generation of molten metallic specimens. In the following, we first review the critical mechanisms involved in the production of the LPP plume, leading to the rule of thumb for controlling the elemental composition of the plume for a given bulk target. The design of the impulse transducer, its calibration, and the signal recovery are then described, toether with the process by which the ablated mass is arrived at. The experiments with the metallic alloy specimens in the solid as well as in the molten phase are compared.

2. REPRESENTATIVE LPP PLUME PRODUCTION

When elemental metallic targets are exposed to laser excitation at power densities in the range of 10^9 to 10^{11} $W \cdot cm^{-2}$, plasma plumes with an electron density in excess of 10^{21} cm^{-3} and a temperature of several hundred electron volts in the core at its peak are produced. Such properties

are determined from the second harmonic generation, measurement of ion acoustic wave frequency, plasma spectroscopy, and numerical simulation [3]. Streak photography shows two distinct stages of the plasma plume development: a "fast ion" plasma, with the radial expansion velocity reaching 2.5×10^7 cm $\cdot$ s^{-1}, and a plasma core, which evolves more slowly. A strong shock wave develops due to the expansion of the plasma core into the remnant gaseous atmosphere of the fast ion plasma (or the preexisting background gas). The shock speed may be as high as 5×10^6 cm $\cdot$ s^{-1}.

The onset of ionization, i.e., the birth of a weakly ionized plasma, initiates direct heating of the plume by the inverse Bremsstrahlung process. For sufficiently large laser powers, the plasma can attain the critical electron density state above which the plasma frequency exceeds the laser frequency, thus preventing the laser from heating the target surface.

Production of an LPP plume, whose composition is the same as that of a multielement target, is necessary for both materials analysis and processing applications. If the velocity of the surface movement is small compared with the thermal diffusion velocity, the evaporation takes place preferentially for those species of high evaporation rates. The result would be an LPP plume of elemental composition which is different from that of the bulk. As the surface heating rate is raised and the movement of the bulk surface speeded up, the elemental dependence of the evaporation rate basically disappears. If the laser heating rate is further increased, the laser beam becomes soon disconnected from the surface due to its direct coupling to the plasma as mentioned above. The plasma emission spectrum becomes dominated by the continuum emission due to the plasma processes, in contrast to the atomic contributions.

The criterion is then to choose the laser pulse power profile in such a way that the movement due to evaporation of the bulk surface is kept in pace with the thermal diffusion front advancing into the interior of the target. The LPP plume thus becomes representative of the bulk in elemental composition. It also becomes spectroscopically productive and robust.

Given that the conditions of the rule of thumb are met, the rate of mass removal scales as the thermal diffusivity of the target matter, subject to the energy required for evaporation and the plasma plume's ability to disperse away from the target surface so that the evaporation process can continue. This means that the thickness of the surface ablation grows inversely with the heat of formation and decreases with increasing molecular weight. The scaling behavior of the measured thickness of ablation according to the elemental properties, as noted above, was demonstrated for four different solid targets, aluminum, copper, manganese, and lead [3].

Using the data in Ref. 3, we have formulated the functional relationship of the thickness, θ, of ablation by LPP excitation to the thermal

diffusivity, D_T, molar weight, M, and heat of formation, H_f, as shown in Eq. (1):

$$\theta = CD_T^{\alpha} M^{\beta} H_f^{\gamma} \tag{1}$$

where $C = 11.07 \pm 0.45$, $\alpha = 0.91 \pm 0.01$, $\beta = -\alpha$, and $\gamma = -1$, when D_T is expressed in units of $cm^2 \cdot s^{-1}$, H_f in $J \cdot g^{-1}$, and θ is given in cm.

It is now possible to extend this as a general method of thermal diffusivity measurement for arbitrary materials. The specimen may be an alloy composed of many elemental species. Here its elemental composition is determined by time- and space-resolved spectroscopy of the LPP plume. On the other hand, the ablative mass loss can be determined by measuring the impulse imparted to the target by an LPP plume, and the thickness of the ablated mass layer can be found from the known mass density of the target specimen. The analysis is not restricted to metallic substances. For transparent materials the window of application can be defined in a different part of the parameter space.

3. MOLTEN MATERIAL SOURCE AND IMPULSE TRANSDUCER

We have extended the above LPP method of thermophysical property measurement to materials in molten states. For this purpose, we have developed a levitation-assisted molten metal source [3], which is powered by a 15-kW RF power generator, operating at frequencies from 70 to 200 kHz. The RF coil has the basic levitator-heater design with three turns and a fourth turn in reverse at the top. The diameter of the levitator-heater coil is about 3.0 cm at its maximum. A metallic rod specimen 6.4 mm in diameter and 5.0 cm in length is placed vertically just below the minimum-B point. The top end is heated to melting while being supported by the levitation field; this prevents the molten metal from spilling over.

The impulse on the target specimen due to LPP ablation is given by the momentum with which all of the ablated species are leaving the surface. Denoting the particle velocity distribution function of the ith species by $f_i(\mathbf{r}, \mathbf{v}, t)$, the mass by M_i, and the component of the velocity normal to the target surface by $v_\perp$, the impulse can be expressed in the form of an ensemble integral summed over all species and integrated over the duration of the laser pulse [Eq. (2)]:

$$\text{Impulse} = \sum_{i=\text{species}} \int_0^{\infty} dv_\perp \, M_i(t) \, f_i(\mathbf{r}, \mathbf{v}, t) \, A v_\perp^2 \tag{2}$$

$$= \sum_{i=\text{species}} \tfrac{1}{2} A k_B \int_0^{\infty} dt \, n_i(t) \, T_i(t) \tag{3}$$

where A denotes the area of the surface exposed to the laser excitation and k_B is the Boltzmann constant. $n_i(t)$ and $T_i(t)$ denote the number density and the temperature of the ith species in the LPP plume as a function of time, respectively. The second step [Eq. (3)] is obtained by invoking the equipartition theorem. The collision time within the plume is of the oder of 1 fs, and the relevant dynamics in the system develops in the nanosecond time regime, meaning that the plasma remains in thermal equilibrium throughout its evolution.

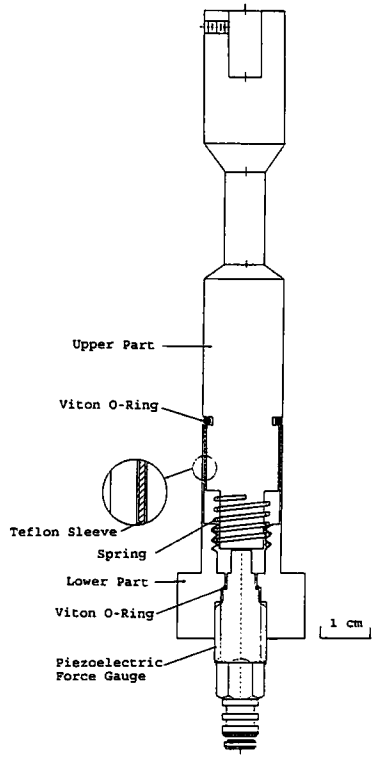

Fig. 1. A schematic diagram of the impulse transducer as installed in the handle of the specimen holder. The upper part where the specimen is mounted is held pressed against the piezoelectric force gauge by a stretched spring. The spring is anchored at the bottom of the upper part of the specimen holder and screwed into a female thread in the lower part.

Notice that the impulse is determined entirely by the number density and temperature of the individual species, not by their velocities. This therefore suggests that in an implementation of the method, the ablative mass loss can be deduced from the measured impulse by calibrating it against an independently measured mass loss from a reference target. All subsequent impulse measurements are then determined relative to the reference target. The thermal diffusivity can also be determined relative to that of the reference target material.

The impulse transducer capability is built into the base of the specimen holder, as shown in Fig. 1. The entire upper part of the specimen holder is spring-loated onto a piezoelectric force gauge (Kistler Model 9702). The impulse from the LPP plume drives the upper part of the specimen holder, including the specimen, into motion, and this displacement is detected by the piezoelectric force gauge after a characteristic time delay. The transducer is then calibrated, as assembled, by dropping a steel bearing ball from a known distance above. The time of flight over a fixed distance interval is measured before, and after, each impact so that the coefficient of restitution can be determined and the impulse calculated. The distance of fall and the size of the ball are varied in order to change the impulse. Maintaining a constant-pressure contact of the upper part of the specimen holder with the force gauge is crucial to the accuracy and reproducibility. This is accomplished by using a coarse spring. The spring is anchored at the bottom of the upper part of the specimen holder and is screwed into a female thread machined into the lower part; before each run the spring tension is reinitialized by twisting the upper part of the specimen holder against the lower part. A calibration curve is generated for each alloy specimen by measuring the maximum value of the transducer output.

4. SIGNAL RECOVERY AND ANALYSIS

A sequence of four successive LPP runs on a solid specimen (SS304) is shown in Fig. 2. First, notice the remarkable regularity with which the response is detected. Second, it is sensitive enough to discriminate the succeeding LPP runs; this is consistent with the spectroscopic observation of the LPP plume in that the solid specimen surface is covered with oxides and other contaminants and requires several LPP exposures to remove them.

Remember that the molten metal is produced in an RF levitator-heater. The movement of the specimen tip due to the LPP impulse induces an emf signal, which is in turn coupled into, and detected through, the piezoelectric transducer electronics. The question arose as to how to recover the small transducer signal. A series of measurements for a solid specimen

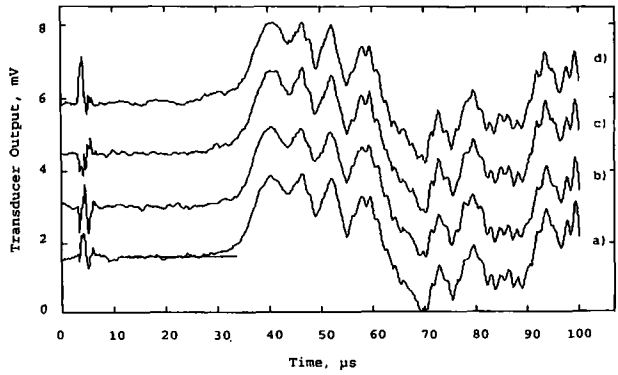

Fig. 2. Impulse transducer outputs from the first four consecutive LPP excitations of a solid specimen (SS304) in the order a to d.

with, and without, the RF field provided the clue. Figure 3 shows four traces taken under different conditions. Trace b is obtained by subtracting trace c, which was taken with the RF field turned off, from trace a with the RF field. It indeed displays the induced emf signal at the RF frequency. Using the short segment of the signal preceding the arrival of the impulse at the piezoelectric force gauge as a template, we can reconstruct the induced emf signal of proper amplitude and phase. This is then subtracted from the trace taken in the presence of the RF field to find the true impulse signal.

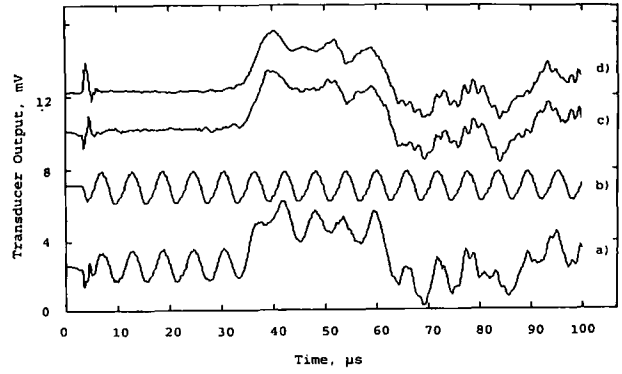

Fig. 3. Impulse transducer output for a solid specimen (SS316) with RF power turned on (a), impulse transducer output with RF power turned off (c), induced emf signal as obtained by subtracting trace c from trace a (b), and impulse signal from another LPP excitation as recovered by subtracting the reconstructed induced emf signal (d). See text for the reconstruction procedure.

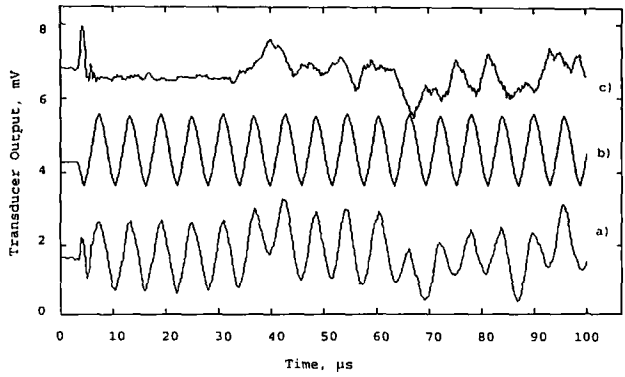

Fig. 4. Impulse transducer output for a molten metal specimen (SS304) as detected with RF power on (a), reconstructed induced emf signal (see text) (b), and impulse signal as recovered by subtracting trace b from trace a (c).

In Fig. 3, trace d shows a similarly recovered result from another LPP run, in excellent agreement with trace c.

The impulse transducer output from a molten metal specimen is similarly processed for signal recovery. Since there are some uncertainties due to fluctuations in the induced emf signal both in frequency and amplitude, we optimize the frequency and amplitude of the reconstructed induced emf signal so as to minimize the rms scatter in the subtracted signal. Figure 4 shows the transducer output (a), reconstructed induced emf signal (b), and recovered impulse signal (c).

We have thus far accomplished $\pm 2\%$ uncertainty in maximum impulse measurement for solid specimens and $\pm 4\%$ for molten metal specimens. By means of the calibration curves, the result can be converted into the impulse at the surface due to LPP ablation. The total mass loss per laser pulse can be found once the elemental composition of the LPP plume and their mean velocity normal to the target surface are determined. The composition is obtained from the time-resolved spectroscopy of the LPP plume by the method we have already developed [6]. The mean velocity of the escaping plume species is measured from the Doppler shifts of the elemental emission lines at the peak of the plasma temperature and density. The reduction of the thermal diffusivity of the molten metal follows from the definitive relationship of Eq. (1) between the product of the mass loss and heat of formation and the thermal diffusivity per unit molar weight.

5. CONCLUSION

The necessary and sufficient conditions for generation of an LPP target-representative plume have been defined through experimental and numerical simulation studies. This makes it possible to measure certain thermophysical properties of materials at high temperatures such as molten metals, which are otherwise difficult to investigate by the conventional means. We have developed a new method employing LPP plumes for determination of the thermal diffusivity of metallic alloys in solid or liquid state. A novel impulse transducer has been designed and implemented for real-time measurement of the mass loss by laser ablation. In a slightly different experimental arrangement, the LPP plumes can be exploited for measurement of other thermophysical properties such as the specific heat, viscosity, and surface tension.

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Department of Energy, American Iron and Steel Institute, CTU 5-2 Consortium of metals producers, and Lehigh University.

REFERENCES

1. J. L. Bobin, *Phys. Rep.* **122**:173 (1985).
2. L. J. Radziemski and D. A. Cremers, eds., *Laser-Induced Plasmas and Applications* (Marcell Dekker, New York, 1989).
3. Y. W. Kim, *Int. J. Thermophys.* **14**:397 (1993).
4. Y. W. Kim, in *Shock Wwaves*, K. Takayama, ed. (Springer Verlag, Heidelberg, 1992), pp. 941–946.
5. Y. W. Kim, *High Temp. Sci.* **26**:57 (1990).
6. Y. W. Kim, in *Advanced Sensing, Modelling and Control of Materials Processing*, E. F. Mathys and B. Kushner, eds. (Minerals, Metal and Materials Society, Wrrendale, PA, 1992), pp. 45–57.