

EVAPORATION AS A DIAGNOSTIC TEST FOR HYDRODYNAMIC
COOLING OF LASER-ABLATED CLUSTERS

Cornelius E. Klots
Chemical Physics Section
Oak Ridge National Laboratory
Oak Ridge, TN 37831

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

I. Introduction

The properties of materials laser-ablated from a surface are of considerable interest. The interrogation of these properties inevitably occurs at a point some distance from the surface. One might then ask what processes have occurred in the intervening path length. Immediately, for example, one wonders whether the material was released as such from the surface or was formed as a result of collisions at a distant point. Similarly, one might ask if an observed "temperature" of the material is characteristic of the ablation process or of subsequent events. We will indicate here how measurements of metastable evaporation rates can provide clues which are pertinent to these questions.

II. The Kinetics of Evaporation

Consider an aggregate of material bound together by van der Waals forces. If it is sufficiently hot, it may undergo evaporation. Any such event will cool the residual material, which will then undergo further evaporation at a lower rate. As a first approximation, the aggregate will have cooled to the point where the rate constant for a subsequent emission is equal to the reciprocal of the time since the original "big bang".

A more careful analysis [1] indicates that such an evaporating population, isolated and normalized to unity at time t_0 , will be given at a later time by:

$$P(t) = 1 - (C/\gamma^2) \ln \{t/[t_0 + (t - t_0) \exp(-\gamma^2/C)]\} \quad (1)$$

where C is the heat capacity of the aggregate in units of k_B , and γ is given by

$$\gamma = \gamma_0^2/[1 - (\gamma_0^2/C)^2/12 \dots] \quad (2)$$

where γ_0 is the Gspann constant. On a microsecond time scale, and in the absence of discontinuities in the heat of evaporation, a "best value" for γ_0 equal to 23.5 (± 1.0) has been suggested [2].

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

*The submitted manuscript has been
authored by a contractor of the U.S.
Government under contract No. DE-
AC05-84OR21400. Accordingly, the U.S.
Government retains a nonexclusive
royalty-free license to publish or reproduce
the published form of this contribution or
allow others to do so for U.S. Government
purposes.

Other cooling mechanisms exist. Thermionic emission has now been identified in several materials [3,4], but is at best unlikely to occur more than once in a given aggregate. Infrared emission will also occur but only on time scales much longer than envisaged here.

III. Pertinence to Ablation

Equation (1) prescribes what is in effect an upper limit to how hot an aggregate can be. Thus, if it is even hotter at the moment of ablation, or if it is generated later via massive condensation, then it will have undergone subsequent evaporation. On the other hand, if the ablation is more gentle, or if a subsequently condensing aggregate is cooled by an adiabatic expansion, before the cessation of collisions, then it is likely to be less prone to evaporation than equation (1) would prescribe.

IV. Two Experiments

Radi et al. [5] have ablated carbon clusters from a graphite rod and have studied their evaporation probabilities. They are compatible with equation (1) above [6]. When the same carbon clusters were generated by ablation from a polyimide surface [7] using generally lower laser fluences, the measured decay rates are instead about an order of magnitude lower.

This difference cannot be attributed to a laser fluence effect, under the supposition that a hotter laser produces hotter clusters. Campbell et al. [7] found that as they increased the fluence, their clusters became cooler, i.e. less prone to evaporation. These authors suggested an increased role for hydrodynamic cooling when ablation was more massive.

V. Discussion

Ablation of a polyimide necessarily releases non-carbonaceous material into the vapor phase. *If it does not condense, then this material would be cooled by any hydrodynamic flow effects and would in turn be available to cool the condensing clusters. This may explain the differing resultant "temperatures".*

A second difference exists between the two experiments. Radi et al. [5] carefully rotated their carbon rod during their experiments so as constantly to expose a new surface; Campbell et al. [7] apparently did not. Deep pitting of a surface might tend to accentuate any hydrodynamic flow effects.

Experiments to distinguish between these two possibilities are not hard to envisage. The immediate point is that both possibilities can be avoided, i.e. that clusters can be made which are as hot as they can be, and that a diagnostic test for this condition exists.

Acknowledgment

Clarifying discussions with E. E. B. Campbell are gratefully acknowledged. This research was sponsored by the Office of Health and Environmental Research, U. S. Department of Energy under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

References

- [1] C. E. Klots, J. Phys. Chem. 92, 5864 (1988).
- [2] C. E. Klots, Int. J. Mass Spectrom. Ion Phys. 100, 457 (1990).
- [3] A. Amrein, R. Simpson, and P. Hackett, J. Chem. Phys. 94, 4663 (1991).
- [4] E. E. B. Campbell, G. Ulmer, and I. V. Hertel (Unpublished Manuscript).
- [5] P. P. Radi, M. T. Hsu, J. Brodbelt-Lustig, M. Rincon, and M. T. Bowers, J. Chem. Phys. 92, 4817 (1990).
- [6] C. E. Klots, Z. Phys. D. (Submitted for Publication).
- [7] E. E. B. Campbell, G. Ulmer, and H. G. Busmann, and I. V. Hertel, Chem. Phys. Lett. 175, 505 (1990).