On Plasma Diagnostics

D.M. Benenson and H.S. Kwok
State University of New York at Buffalo
Amherst, New York U.S.A.

I. Introduction.

A variety of diagnostic techniques is useful, perhaps essential, in studies of plasmas - owing to (1) the wide range encountered in such variables as temperature, particle density, and velocity and (2) the desirability of independent verification of measurements. Of many methods, we note briefly below aspects concerning spectroscopy, laser velocimetry, and interferometry; laser induced fluorescence is described in more detail.

II. Spectroscopy.

1. General Remarks.

Classical spectroscopy (for example, through emission of line and continuum radiation) enables the determination of temperatures to as low as about 9000K. With the associated assumption of local thermal equilibrium, particle densities become determined; line broadening methods permit independent measurements of electron density. Where nonequilibrium exists or may be a factor, appropriate excitation temperatures may be determined through, for example, the multithermal equilibrium model developed in Ref. 1. At reduced temperatures the line and continuum radiation may become relatively weak. Rotational temperatures of molecular gases may be determined to the range of 6000K using individual rotational lines (which requires a spectrograph of high resolving power) or the integrated measurements of such lines concentrated about the band head (Refs. 2,3). [Below about 3500K thermocouple techniques may be employed (Ref. 4); the time response of such (invasive) probes restricts their range of applicability in dynamic arcs.]

Data acquisition techniques which rapidly scan or transverse the image of the plasma across the entrance slits of a spectrograph, together with appropriate electronic synchronization with respect to a base reference, permit the more or less instantaneous determination of temperatures within dynamic arcs (Refs. 5,6), such as are found in circuit breakers.

In most cases sufficient circular symmetry in cross-section exists so that Abel inversion of the experimentally obtained integrated intensity distribution is warranted. Procedures have been developed and applied to plasmas in which the cross-section is non circular (Refs. 7,8).

Plasma processing at high pressure, involving the interactions of the plasma and a flowing medium, frequently may encounter cases in which hydrodynamic turbulence plays an important role. Local temperatures within such plasmas, incorporating the effects of turbulence upon the radiation measurements, have been obtained in Ref. 9.

2. Laser Induced Fluorescence.

Here, a potentially useful diagnostics method for plasma chemistry is discussed - the application of laser-induced-fluorescence (LIF). LIF has been applied successfully to the study of combustion processes (Ref. 10), flames (Ref. 11), and low temperature plasmas (Ref. 12). In each of these cases conditions and requirements exist that are similar to those in plasma devices: moderately high temperature ($\gtrsim$ 2000K), nonequilibrium gas compositions, multitude of transient species, and a need for a probe that does not disturb the dynamics of the process to be studied. We shall describe some salient features of LIF as a diagnostic tool and then discuss some of its potential applications in plasma chemistry.

LIF originally was developed as a sensitive scheme for detecting minute amounts of short-lived chemical species in chemical dynamics studies (Ref. 13). It was recognized that, in addition to its sensitivity, this method can also yield information on state population distribution, kinetics of energy transfer, and lifetimes of the radical species. In particular, much attention has been devoted to the OH radical because of its important role played in the combustion process (Ref. 14). Basically, the LIF scheme consists of using a tunable laser to excite the electronic state of the species (atom or molecule) under investigation. For atoms, resonant fluorescence will be observed while the excited molecule redistributes its energy within the vibrational-rotational manifold before eventually coming down to the ground state. The transition to the ground state may be radiative or nonradiative depending on the quantum efficiency of the fluorescence process. For this reason most LIF was carried out with

atomic or diatomic species because of the high quantum yield (and simpler energy level structures). Observation of the emitted photons in the case of a radiation relaxation then constitutes a detection of the presence of that species. Because the excitation process is highly selective, LIF can easily detect a small amount of the desired radical within a very diluted mixture (Ref. 15).

In addition, if the laser is tuned through the excitation spectrum of the species, a population distribution among the various energy states can then be deduced. Assuming a Boltzmann type distribution one can then infer the internal temperature of the species, which should be the same as the LTE translational temperature. Moreov , because of the spatial coherence of the laser, it is possible to obtain very fine spatial resolution of this temperature - a very important piece of data in combustion studies. By pulsing or chopping the laser beam it is also possible to obtain temporally resolved temperature and concentration information.

Let us discuss here the advantages of LIF as applied to the study of plasma chemistry, in particular to plasma processing and to the diagnostics of high current interrupters. As compared to classical emission methods, LIF affords a much higher sensitivity and can be applied at much lower temperatures. For visible emission from a plasma temperatures greater than about 6000K, and reasonably high concentration of the species, are necessary. LIF has detectability limits in the $10^4 cm^{-3}$ concentration region and can identify a species in dilution as low as 1 part in 10^9. In addition, the initial plasma temperature is inconsequential to the LIF signal whereas, for emission measurement, a high temperature is required in order to have sufficient population in the excited states. The advantages of LIF in the diagnostics of curren interrupters during the current zero and recovery region are, therefore, obvious. Moreover, in many plasma processing applications, transient species need to be identified and the reaction kinetics need to be understood. In cases where temperatures are not high enough to produce sufficient visible fluorescence, LIF will be the only possible diagnostic tool. These advantages make LIF a very attractive tool for plasm diagnostics.

There are potential disadvantages and difficulties in LIF, however. First, with respect to classical methods, the technique requires a more elaborate set-up and considerable capital expenditure (involving lasers and data acquisition instrumentation). Aside from the cost factor, there is also the need for basic spectroscopic data for the species under study. While atomic data are abundant, electronic spectra and Einstein A coefficients of many diatomic radicals and molecules are not so well established (Ref.16). Considerable effort has been devoted to species of interest in combustion processes (e.g., OH), while species such as SF, important in SF_6 circuit breakers, are not too well known; further studies are needed in this area.

For SF_6 plasmas, it has been estimated (Ref. 17) that at 10,000K only S, F, F^-, are present. As the plasma cools to 2000 K (as is typical in actual situations), species such as SF, S_2 and F_2 begin to form, with SF being predominant. For SF the first electronic excited states are $A_1{}^2\Pi_{3/2}$ and $A_2{}^2\Pi_{1/2}$ lying $\sim$25,000cm^{-1} above the ground state. The spin-orbit splitting of the ground state is about 401cm^{-1} (Ref. 1 . The electronic excitation for S_2 and F_2 occur at higher frequencies, at 31,835cm^{-1} an 34,500cm^{-1}, respectively. These frequencies are all accessible by common dye lasers. This particular example of an SF_6 circuit breaker is particularly suited for study by LIF. In principle, the concentration of various species can be monitored by LIF as a function of time during the current interruption cycle.

III. Laser Velocimetry.

By making use of the coherence properties of the laser a precisely definable grid can be formed in space. The velocity of particles (or scattering centers) that cross this configuration is determined through (1) the frequency of the scattered light and (2) knowledge of the grid (fringe) spacing. Velocities to several kilometers per second (as occurs in such devices as gas blast circuit breakers) can be measured with this technique. Laser velocimetric methods have been applied to measurements of the flow field in the exhaust of plasma torches as well as within the column itself (Refs. 4,19,20), and of the turbulence intensity within a torch (Ref. 19).

Laser velocimetry is not without constraints: (1) the method is (moderately) invasive, requiring the presence of scattering centers or particles, (2) owing to drag forces, neither the velocity nor the trajectory of the particle generally will be the same as that of the surrounding fluid; appropriate modeling (Ref. 21) can account for these effects as well as the additional, dynamic changes in the size (and in the motion) of particles immersed in a high temperature environment (e.g., the plasma; Ref. 22), and (3) the presence of turbulence and of high acceleration (as in

nozzle flow) and high velocity fields can place severe restrictions upon the size of particles that are used to follow the local motions (Ref. 23).

IV. Interferometry.

Upon passing through a plasma or its heated environs (e.g., the thermal boundary layer surrounding column), light is primarily phase shifted (the attenuation and reflection being small) - the shift depending primarily upon the particle density (Ref. 24). The phase shift is associated with the local index of refraction (associated with particle density and temperature), expressible - in terms of parameters of the plasma and of the wavelength of the laser - through the dispersion relation. Through the coherence and relative monochromacity of the laser, laser interferometry has rapidly become accessible as a diagnostic. For example, temperatures in the range from room to plasma levels can be measured with this technique. Perhaps the simplest arrangement is that of differential interferometry employing the Wollaston prism (Ref. 25). Here, light incoming to the prism (and linearly polarized at 45°) is split into two components which emerge from the prism with a small angular separation and a phase difference. The interference fringes formed in the image plane are associated with gradients of the index of refractions (or of particle density or temperature) in the object plane (e.g., the plasma). With double exposure or real-time holographic interferometry (Ref. 26) the local value of the index of refraction can be obtained. Considering, for example, the double exposure method applied to the case of an arc immersed in a flowing medium, the first hologram would be obtained under the reference, cold flow conditions (i.e., in the absence of the plasma). With the plate as yet undeveloped, the second hologram would be recorded with hot flow operation. Following development of the plate and its illumination, the result would produce an interference pattern that describes the effect of the plasma upon the flow. In both cases Abel inversion of the resulting interferograms may be required to determine the distribution of temperature (Ref. 27). It should be noted that the presence of turbulence tends to obliterate the fringe pattern.

References.

1. T.L. Eddy, E. Pfender, and E.R.G. Eckert, IEEE Trans. on Pl.Sci. PS-1, 31-42, 1973.
2. T. Sakuta and Y. Kito, IEEE Trans. on Pl.Sci. PS-8, 29-32,1980.
3. P. Fauchais, K. Lapworth, and J.M. Baronnet, Interna.U.P.&A.Chem. 1980.
4. A Vardelle, J.M. Baronnet, M. Vardelle, and P. Fauchais, IEEE Trans.Pl.Sci 417-424,1980.
5. W. Hermann, U. Kogelschatz, L. Niemayer, K. Ragaller, and E. Schade, IEEE Trans. on Pl.Sci. PAS-95, 1165-1176, 1976.
6. J.E. Fitzgerald and D.M. Benenson, ASME J. of Heat Trans. 95, Series C, 535-440, 1975.
7. J.L. Wu and D.M. Benenson, IEEE Trans. on Pl.Sci, PS-4, 284-292, 1977.
8. Y. Yasultomo, K. Miyata, S.I. Himeno, T. Enoto, and Y. Ozawa, IEEE Trans. on Pl.Sci., PS-9, 18-21, 1981.
9. Y.K. Chien and D.M. Benenson, IEEE Trans on Pl. Sci., PS-8, 411-417, 1980.
10. J.D. Bradshaw, N. Omentto, G. Zizak, J.N. Bower and J.D. Winefordner, App.Op., 19 2709-2716, 1980.
11. J.H. Bechtel, App.Op., 18, 2100-2106, 1979.
12. J. Hildebrandt and H.J. Kunze, Phys.Rev. Lett. 45, 183-186, 1980.
13. R.N. Zare and P.J. Dagdigian, Sci., 185, 739-747, 1974.
14. C. Chan and J.W. Daily, App.Op., 19, 1963-1968, 1980.
15. J.W. Daily, App.Op.,17, 1610-1615, 1978.
16. G. Herzberg, IV Spectroscopic Data for Diatomic Molecules., D.Van Nostrand, 1978.
17. J. Kopiansky, Plenum Press, 329-354, 1978.
18. A Carrington and G.N. Carrie, J. Chem.Phys., 50, 2726-2732, 1970.
19. J. Lesinski, B. Mizera-Losinska, J. Jurewicz and M.I. Boulos, AIChE, 1980.
20. T.Y. Li, D.M. Benenson, and D.P. Malone, GEC Conf, 1979.
21. J. Lesinski, B. Mizera-Losinska, J.C. Fanton, and M.I. Boulos, AIChE.
22. M.I. Boulos, IEEE Trans. on Pl.Sci., PS-6, 93-106, 1978.
23. M. Lapp, C.M. Penney, and J.A. Asher, Aerospace Res.Lab. ARL-73-0045, 1973.
24. J. Thompson and M. Krintiansen, Editors, Texas Tech U., 1973.
25. U. Kogelschatz, App.Op. 13, 1749-1752, 1974.
26. W. Tiemann, Siemens, Forschung, Entwickl.Ber. 5, 163-166, 1976.
27. H.L. Walmsley, G.R. Jones, and M.R. Barrault, Uni.Liverpoor,ADRP-ULAP-756, 1978.