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Authors

Beier, R.A.
Pagni, P.J.

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R.A. Beier and P.J. Pagni

January 1982

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SOOT VOLUME FRACTION PROFILES IN
FORCED COMBUSTING BOUNDARY LAYERS

R.A. Beier¹ and P.J. Pagni

Lawrence Berkeley Laboratory

and

Mechanical Engineering Department

University of California

Berkeley, CA 94720

¹ Current address: Basic Research Department, Factory Mutual Research,
1151 Boston-Providence Turnpike, Norwood, Massachusetts 02062

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ABSTRACT

A multiwavelength laser transmission technique is used to determine soot volume fractions and approximate particle size distributions in a forced flow combusting boundary layer. Measurements are made in diffusion flames of polymethylmethacrylate (PMMA) and five liquid hydrocarbon fuels (n-heptane, iso-octane, cyclohexane, cyclohexene, and toluene) at different ambient oxygen mass fractions. Soot is observed in a region between the pyrolyzing fuel surface and the flame zone. Soot volume fraction increases monotonically with ambient oxygen mass fraction, $Y_{O\infty}$, e.g., n-heptane and PMMA are similar with soot volume fractions, f_v , ranging from $f_v \sim 5 \times 10^{-7}$ at $Y_{O\infty} = 0.23$ to $f_v \sim 5 \times 10^{-6}$ at $Y_{O\infty} = 0.50$. For an oxygen mass fraction the same as air, $Y_{O\infty} = 0.23$, soot volume fractions are approximately the same as values previously reported in pool fires and a free combusting boundary layer. For all fuels tested, a most probable radius is between 20 nm and 80 nm, and does not change significantly with oxygen mass fraction.

INTRODUCTION

Thermal radiation is the dominant mode of heat transfer in full scale fires [1], where soot emission dominates gas species emission. In a compartment fire, the composition of the gas in the enclosure changes, as the oxygen is consumed and products of combustion are formed. The effect of oxygen mass fraction on soot volume fraction is important for calculation of radiative heat transfer. Experimental studies of soot in well controlled flames provide insights to complex soot formation and oxidation processes. A forced flow, two dimensional, laminar combustng boundary layer is attractive for a study of flame soot, because the fire can be easily probed by optical techniques and predicted by mathematical models [2-4]. A schematic diagram of a laminar combustng boundary layer over a pyrolyzing fuel surface is shown in Fig. 1. A pyrolysis zone separates a flame zone from the fuel surface. Part of the carbon in the fuel is converted to carbon particulates, which are observed in a soot layer on the fuel side of the flame zone. Volume fractions of soot and approximate size distributions are determined by a multiwavelength laser transmission technique [5-7]. Previous measurements have determined soot volume fractions in pool fires [8-9] and a free flow combustng boundary layer [10]. In this study, the dependence of soot volume fraction on the ambient mass fraction of oxygen is determined in a forced combustng boundary layer.

Minchin [11] and Clarke, Hunter, and Garner [12] did early studies of the effect of fuel type on soot formation in laminar diffusion flames. They increased the height of a flame on a small circular burner by increasing the fuel flow rate until the flame

emitted soot at its tip. This height is called the smoke height or sooting height. When the sooting height is used as a relative measure of sooting tendency, a lower sooting height indicates a greater tendency to form soot. Jagdo, Prado, and Lahaye [13] measured local soot concentrations by probe and light scattering and absorption techniques in a candle-like diffusion flame. Related experiments for gaseous fuels have been reported by Kent, Jander, and Wagner [14], Haynes, Jander, and Wagner [15,16], and Chang and Penner [17].

Recently, Glassman and Yaccarino [18] measured sooting points to determine the effects of oxygen concentration on the sooting tendency of fuels. Santo and Taminini [19] used a modified Schmidt technique to measure the flame radiance of a pool fire against a variable background of black body radiation. These measurements allowed them to determine absorption-emission coefficients and trends in soot volume fraction for different ambient oxygen concentration. While these previous studies give qualitative trends, the present study reports quantitative data for soot volume fractions at different oxygen concentrations.

Sibulkin, Kulkarni, and Annamalai [20], Liu and Shih [21], Kinoshita, Pagni, and Beier [22-24] recently included radiation in models of a laminar combustng boundary layer. These models require a knowledge of soot volume fraction within the layer, which is provided by the present study.

EXTINCTION ANALYSIS

The multiwavelength laser transmission technique is described elsewhere [5-8,23], so only a brief outline of the assumptions and data analyses are given here. The transmitted intensity, I , of a

monochromatic beam through a polydisperse aerosol is related to the initial intensity, I_0 by

$$I(\lambda)/I_0(\lambda) = \exp(-\tau(\lambda)L), \quad (1)$$

where L is the beam pathlength. An aerosol spectral extinction coefficient, τ , is given by

$$\tau(\lambda, m, r) = \int_0^\infty N(r) Q(\lambda, m, r) \pi r^2 dr \quad (2)$$

where Q is the particle extinction efficiency from the Mie scattering theory [25] for spherical particles. Calculation of a spectral extinction coefficient requires knowledge of the optical properties of soot, $m=n(1-ik)$, and the size distribution, $N(r)$. The values of the optical properties which were derived by Lee and Tien [26] and listed by Bard and Pagni [8] are used. A Gamma size distribution [27] with $\sigma/r_m = 1/2$,

$$N(r)/N_0 = (27 r^3 / 2 r_{\max}^4) \exp(-3 r/r_{\max}) \quad (3)$$

is assumed, where $r_{\max}$ is the most probable radius, and N_0 is the total particle concentration. The volume fraction of soot is defined by

$$f_v \equiv \frac{4}{3}\pi \int_0^\infty N(r) r^3 dr \quad (4)$$

which, with equation (3), gives

$$f_v = \frac{54\pi}{3^8} \Gamma(7) N_0 r_{\max}^3 = 18.62 N_0 r_{\max}^3 \quad (5)$$

Measurements are made of the ratio of the transmitted and initial intensities, I/I_0 , at two different wavelengths, superimposed over the

same pathlength, L . Substituting these measured values into equation (1) gives two independent values of τ , one at each wavelength. These two τ values give two independent equations (2) to determine the two unknowns in the size distribution, N_0 and $r_{\max}$ [5,8]. The volume fraction of soot is obtained from equation (5).

Since soot particles may agglomerate into larger clusters, Lee and Tien [28] studied how different shapes affect the radiative characteristics of a medium. With long chains approximated as cylinders, they considered spherical and cylindrical particles as two limiting cases. From their calculations, particle shape is most important in the far infrared region. For the visible wavelengths of these transmission measurements and the near infrared region of most flame radiation, calculations based on spherical particles give satisfactory results.

EXPERIMENTAL METHOD

Multiwavelength laser transmission experiments are performed with the apparatus shown schematically in Fig. 2. The flames are in a wind tunnel designed to produce a uniform flow of an oxidizing mixture of nitrogen and oxygen above the fuel surface. The wind tunnel consists of a mixing chamber, contraction section, and test section. The mixing chamber is divided into sections by screens with one section packed with steel wool. A rapid contraction of two walls accelerates the flow and minimizes the thickness of the boundary layers on the walls at the entrance of the test section. With an area contraction ratio of 6, the shape of the contraction is similar to the one used by Brown and Roshko [29]. The horizontal test section has a rectangular cross section, 18 cm wide and 5 cm high. The sides of the tunnel and

the ceiling directly over the fuel surface are made of pyrex glass to provide visibility and access for optical measurements. The fuel wicks and solid fuel are placed flush in an inert floor, Marinite-XL (John-Manville Co.), 5 cm behind the exit of the contraction as shown in Fig. 3. The oxidizer flow is measured by a set of four critical flow nozzles for each of the component gases, oxygen and nitrogen. The nozzles are calibrated individually at operating conditions by a wet test meter. For each gas, the stagnant chamber pressure upstream of the critical flow nozzles is measured by a Bourdon tube gauge. A copper-constantan thermocouple upstream of the nozzles is used to measure the stagnant chamber temperature. The static pressure downstream of the nozzles is also monitored to insure choked operation of the nozzles.

The setup of the optical equipment for the laser transmission measurements is similar to that described by Pagni and Bard [7-9]. Two lasers are used: a Spectra-Physics Argon Ion tunable laser model 165, operating at either $\lambda = 0.4579 \mu\text{m}$, $0.4880 \mu\text{m}$, or $0.5145 \mu\text{m}$, and a Spectra-Physics Helium Neon laser model 125, emitting at $\lambda = 0.6328 \mu\text{m}$. After a cube beam splitter superimposes the two beams from the lasers, the beams pass through the same physical space in the fire. A first simple lens reduces the diameter of the beams in the fire. After a second lens collects the transmitted light, an equilateral prism separates the two beams. Each beam passes through another focusing lens and a narrow band pass filter, before it strikes a laser power meter (Newport Research Corp., model 820). The beam splitter provides a second beam from each laser, which is monitored as a reference intensity by the same prism-detector system. A PDP-11/34

microcomputer stores the data, after the output signals from the detectors pass through a d.c. amplifier and a (Digital Equipment Corp.) AR11 16-channel, 10 bit, A/D converter. A digital timer, with an adjustable period, triggers the A/D converter to read the output signals of the detectors.

Since the transmitted intensity of the laser beams varies greatly for different fuels and oxygen mass fraction, wicks of different widths are used as listed in Table 1. The wick is a ceramic fiber board, Fiberfrax Hot Board, Carborundum Co. With aluminum foil around the hidden surfaces of the wick, the inert wall does not absorb liquid fuel. The length of the wick is 12 cm except for toluene where the length is 8.4 cm. The samples of polymethylmethacrylate (PMMA) are usually 12 cm long, 12 cm wide, and 1.27 cm thick. The diameter of the laser beam limits the spatial resolution of the transmission measurements. For all pathlengths used the maximum beam width between points of $1/e^2$ intensity is less than 0.3 mm. Since variations in temperature and species change the index of refraction in the direction normal to the fuel surface, the fire deflects the laser beam. However, the deflection does not decrease the spatial resolution with the lense system shown in Fig. 2.

The entire wind tunnel is placed on a moveable table, so the distance between the fuel surface and the superimposed laser beams can be changed by moving the wind tunnel vertically. A complete scan of the fire is obtained by moving the wind tunnel horizontally between traverses. After the mass flow rates of oxygen and nitrogen are set, the fuel is ignited. For the liquid fuels, the supply of fuel in the wick gives a steady fire for a time period between 3 to 5 minutes,

depending on the oxygen mass fraction. This time period allows one vertical scan, with increments of 0.3 mm, through the boundary layer. At each position the outputs of the detectors are read 300 times by the data acquisition system within a 6 second interval. After each vertical scan the fire is extinguished, the horizontal position of the fuel is changed, and the wick is soaked with fuel, before the fire is reignited.

The pathlength, L , is measured after the fire is extinguished from the deposits of soot on the inert floor. On the side edges of the fire, the flame zone is in contact with the inert floor. Soot is deposited on the inert floor between the fuel surface and this line of flame contact. The width of the fire and the pathlength, L , are taken to be equal to the distance between the lines of flame contact on each side of the fuel surface. The pathlength is approximately 1 cm longer than the width of the wick. The measured values of I/I_0 for two different wavelengths and L are used in the extinction analysis to calculate $r_{\max}$ and f_v . Data for more than one wavelength pair are used to isolate the correct value of $r_{\max}$ [5,8,23].

For PMMA the fuel regression rate affects the beam distance from the fuel surface. The regression rate is measured before and after each vertical scan through the boundary layer by blocking the laser beams partially with the fuel surface. If the floor of the wind tunnel remains stationary, the surface of the pyrolyzing fuel slab moves and blocks a smaller part of the laser beams. The regression rate is the rate the wind tunnel floor is moved to maintain a constant reading of transmitted intensity for the partially blocked beam. A new sample of PMMA is burned for the transmission measurements at each

streamwise position.

RESULTS AND DISCUSSION

Profiles of volume fraction of soot are shown in Fig. 4 for five liquid fuels and PMMA at a free stream velocity of 1.5 m/s and ambient oxygen mass fraction, Y_{O_∞} of 0.25. The standard deviations in the data are about the same as the size of the symbols in the graph. When the data are compared with previous measurements [5,8,10,30], the ranking of fuels by soot volume fraction is preserved among small pool fires, and free and forced laminar boundary layers. Table 1 lists values of soot volume fractions, the most probable radius, and total particle concentrations at the peaks of these profiles. The five liquid hydrocarbon fuels have approximately the same thermal and physical properties, and adiabatic flame temperature. The large variation in soot volume fraction indicates the chemical structure of the fuel has a strong influence on soot formation. With wicks of 12 cm in length, measurements of burning time and mass of the wicks, before and after the burning, show the total pyrolysis rate at the fuel surface for toluene is 40% greater than that of n-heptane. The pyrolysis rates of the other liquid fuels are between these two extremes. For toluene, the largest soot volume fraction provides the largest radiative heat flux into the fuel surface, which causes the largest pyrolysis rate.

The soot volume fraction increases with downstream distance, x , for all fuels tested, but for fuels with a high soot volume fraction the variation with x is smaller. In Fig. 5, the profiles for cyclohexene indicate a larger variation in soot volume fraction with x than determined in the free flow case for $Y_{O_\infty} = 0.23$. The profiles

are sharply peaked at $x = 20$ and 40 mm due to the competing processes of formation and oxidation of soot. Convection of soot downstream tends to round the profile at $x = 100$ mm. The outer edge of the soot layer follows the flame zone away from the fuel surface with increasing downstream position, and the width of the layer increases.

The particle radii do not change significantly across the soot layer as shown in Fig. 6. The most probable radius is between 20 nm and 80 nm for all the fuels at $Y_{O_\infty} = 0.25$. Since the particle sizes do not vary with downstream position, the particles apparently do not have sufficient time to agglomerate into larger clusters. The peaks in the particle concentration profiles correspond to peak soot volume fractions as seen by comparing Figs. 5 and 7.

The soot volume fraction increases with increasing oxygen mass fraction for all the fuels. In Fig. 8, results for n-heptane show a small change in the oxygen mass fraction from $Y_{O_\infty} = 0.23$ to $Y_{O_\infty} = 0.25$ gives a 60% increase in the peak of the volume fraction profile at $x = 100$ mm. The results demonstrate the volume fraction is less sensitive at higher oxygen mass fractions, since a change from $Y_{O_\infty} = 0.35$ to $Y_{O_\infty} = 0.50$ gives only a 50% increase in the peak of the profile. As oxygen mass fraction increases, the outer edge of the soot layer moves with the flame toward the fuel surface. Thus, the width of the soot layer decreases. The increase in volume fraction of soot occurs for two reasons. Measurements of the burning time and the mass of the wick, before and after the burning, show the total pyrolysis rate at the fuel surface is doubled by increasing the oxygen mass fraction from $Y_{O_\infty} = 0.23$ to $Y_{O_\infty} = 0.50$ for a wick of 12 cm in length. Thus, more fuel feeds the soot formation process at the

higher oxygen concentrations. Also, an increase in the flame temperature increases the rates of formation and oxidation of soot as noted by Glassman and Yaccarino [18]. The large increase of soot volume fraction indicates the formation process prevails between these two competing processes.

For PMMA the width of the soot layer changes less with oxygen mass fraction than for the liquid fuels as shown in Fig. 8. The distance from the fuel surface to soot onset is less for PMMA, and this distance is approximately unchanged with increasing oxygen mass fraction. Again, a large increase in the peak of the volume fraction profile is found with an increase in the oxygen mass fraction. The particle radii do not change significantly with oxygen mass fraction as shown by the results in Table 1. For PMMA pool fires, the measurements of Santo and Taminini [19] indicate the absorption coefficient increases with oxygen mass fraction, but the Schmidt temperature is only weakly influenced. They concluded that soot volume fraction increases with an increase in oxygen mass fraction for Y_{O_∞} between 0.18 and 0.21.

By measuring sooting heights for ethene, Glassman and Yaccarino [18] found that increasing the oxygen mass fraction decreases the sooting tendency at lower oxygen mass fractions, but at higher mass fractions increasing the oxygen mass fraction increases the sooting tendency. In separate studies, Kadota, Hiroyasu, and Farazandehmehr [31], and Chakraborty and Long [32,33] measured the amount of soot leaving the combustion zone of a diffusion flame. In these previous studies, different fuels were used in diffusion flames with different geometries, so a direct comparison with this study is difficult.

The volume fraction of soot has a weaker dependence on the velocity of the free stream than oxygen mass fraction. In Fig. 10 the results for n-heptane are shown, where the free stream velocity, u_{∞} , has been changed by a factor of three. The width of the soot layer decreases with increasing velocity as the flame zone moves closer to the fuel surface. Although the soot volume fraction decreases with increasing velocity, the flux of soot at a given downstream position should be used to make a meaningful comparison. The downstream flux of soot at streamwise position, ℓ , gives a measure of the amount of soot formed but not oxidized between $x = 0$ and $x = \ell$. For a calculation of the flux of soot particles, detailed velocity profiles are needed.

CONCLUSIONS

For different ambient oxygen mass fractions, soot volume fractions and size distributions have been determined in a forced flow boundary layer. A soot region is observed on the fuel side of the flame zone. The ranking of fuels by soot volume fraction is preserved among small pool fires and laminar combustions boundary layers in free and forced flow. The chemical structure of the fuel has a large effect on soot formation. The soot volume fraction and the width of the soot layer increase with downstream distance. Soot volume fraction increases monotonically with mass fraction of oxygen, but volume fraction is less sensitive to changes in $Y_{O_{\infty}}$ at higher oxygen mass fractions. The particle size does not change significantly across the soot layer, or with oxygen mass fraction or fuel type. The combustions boundary layer is an attractive system for future work in soot formation modeling.

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NOMENCLATURE

f_v	particulate carbon volume/flame volume
I	radiant intensity
L	beam pathlength through fire
m	complex index of refraction
n	real index of refraction
nk	imaginary index of refraction
N_o	total particle concentration
$N(r)dr$	particle concentration in the size range dr about r
Q	extinction efficiency
r	particle radius
x	streamwise direction coordinate
y	transverse direction coordinate

Greek

λ	wavelength
σ	standard deviation
τ	extinction coefficient

Subscripts

i	first wavelength
j	second wavelength
m	mean
max	most probable, i.e. at maximum in $N(r)$
o	oxygen or incident
∞	ambient

Table 1. Summary of Sample Geometries and Peak Soot Volume Fractions and Size Distributions at $x = 4$ cm with $u_\infty = 1.5$ m/s.

Fuel	$Y_{ox,\infty}$	Sample Width (cm)	Wave-length Pair, i-j	Path-length L (cm)	Distance from Fuel Surface (mm)	$(I/I_o)_i$	$(I/I_o)_j$	$f_v \times 10^{-6}$	r_{max} (nm)	$N_o \times 10^{-9}$ (cm ⁻³)
Toluene (C ₇ H ₈)	0.25	1.7	2-4	2.6	2.7	0.0005	0.0011	22	79	2.4
Cyclohexene (C ₆ H ₁₀)	0.23	7.0	2-4	8.3	3.8	0.088	0.20	1.7	40	1.5
	0.25	3.5	1-4	4.7	3.6	0.075	0.25	3.1	31	5.4
	0.25	3.5	1-4	4.7	3.1	0.084	0.27	3.0	30	5.9
	0.25	3.5	2-4	4.5	2.8	0.16	0.27	2.4	48	1.2
	0.25	1.7	2-4	3.1	2.8	0.28	0.43	2.5	39	2.2
	0.35	3.5	1-4	4.7	3.0	0.0016	0.017	7.5	44	4.8
	0.35	3.5	2-4	4.5	3.2	0.010	0.038	6.1	48	2.9
Iso-octane (C ₈ H ₁₈)	0.25	7.0	2-4	8.3	3.2	0.20	0.32	1.1	49	0.52
Cyclohexane (C ₆ H ₁₂)	0.25	7.0	2-4	8.3	3.8	0.38	0.45	0.78	64	0.16
	0.23	12.0	2-4	13.8	4.0	0.62	0.74	0.21	36	0.24
n-Heptane (C ₇ H ₁₆)	0.25	7.0	2-4	8.4	3.7	0.61	0.68	0.37	56	0.11
	0.25	7.0	2-4	8.4	3.8	0.59	0.68	0.38	49	0.17
	0.35	7.0	1-4	8.7	3.5	0.053	0.20	1.9	32	3.2
	0.35	7.0	2-4	8.6	3.2	0.069	0.17	1.9	41	1.4
	0.50	7.0	2-4	8.1	2.9	0.015	0.048	3.1	50	1.3
	0.50	7.0	2-4	8.3	3.1	0.015	0.051	3.0	47	1.6
	0.25	12.0	2-4	12.0	2.6	0.48	0.58	0.37	49	0.17
Polymethyl-methacrylate (C ₅ H ₈ O ₂) _n	0.35	12.0	2-4	12.0	2.1	0.0096	0.043	3.0	42	1.6
	0.50	6.0	2-4	6.0	1.9	0.019	0.060	4.0	48	2.0

FIGURE CAPTIONS

Fig. 1 Schematic diagram of a steady, two dimensional, laminar, combusting, boundary layer on a pyrolyzing fuel slab.

Fig. 2 Schematic diagram of wind tunnel and optical equipment for simultaneous multiwavelength laser transmission measurements.

$L\lambda_i, L\lambda_j$	- Laser at λ_i or λ_j
M	- Mirror
B	- Beamsplitter
P	- Prism
FL1	- Focusing lens #1 ($f = 550$ mm)
FL2	- Focusing lens #2 ($f = 250$ mm)
FL3	- Focusing lens #3 ($f = 200$ mm)
FL4	- Focusing lens #4 ($f = 78$ mm)
F	- Bandpass filter (bandwidth = 30 Å)
D, D	- Detector for transmitted intensity
CFN	- Critical flow nozzle
TC	- Thermocouple

Fig. 3 Contraction and test sections of wind tunnel.

Fig. 4 Soot volume fraction, f_v , as a function of distance from fuel surface at $x = 4$ cm for five liquid, hydrocarbon fuels and PMMA.

Fig. 5 Soot volume fraction, f_v , as a function of distance from fuel surface at different downstream positions for cyclohexene. Wavelength pair numbers refer to λ_j . ($\lambda_1 = 0.4579$ μm , $\lambda_2 = 0.4880$ μm , $\lambda_3 = 0.5145$ μm , $\lambda_4 = 0.6328$ μm).

Symbol	x (mm)	Wavelength Pair
Δ	20	1-4
$\square$	20	2-4
∇	40	1-4
$\diamond$	40	2-4
$\blacktriangledown$	100	1-4
$\blacklozenge$	100	2-4
$\blacktriangle$	100	3-4

Fig. 6 Most probable particle radius, $r_{\max}$, as a function of distance from the fuel surface and downstream position for cyclohexene at $Y_{\infty} = 0.23$. The symbol definitions are those given in caption to Fig. 4.

Fig. 7 Total particle concentration, N_o , as a function of distance from the fuel surface and downstream position for cyclohexene in forced flow at $Y_{\infty} = 0.23$. The symbol definitions are those given in the caption to Fig. 4.

Fig. 8 Soot volume fraction, f_v , as a function of distance from the fuel surface at different oxygen mass fractions for n-heptane.

Symbol	Y_{O_∞}
■	0.23
△	0.25
◆	0.35
▽	0.50

Fig. 9 Soot volume fraction, f_v , as a function of distance from the fuel surface at different oxygen mass fractions for PMMA. The symbol definitions are those given in the caption to Fig. 7.

Fig. 10 Soot volume fraction, f_v , as a function of distance from the fuel surface at different free stream velocities for n-heptane at $x = 4$ cm and $Y_{O_\infty} = 0.35$.

Fig. 1

XBL 813-5397

Fig. 2

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Fig. 3

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Fig. 4

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Fig. 5

XBL 811-12814

Fig. 6

XBL 8111-12815

Fig. 7

XBL8III-12816

Fig. 8

XBL 8111-12817

Fig. 9

XBL8111-12819

Fig. 10

XBL8III-12820

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TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720