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SWIRL-STABILIZED COMBUSTOR(U) AIR FORCE ENGINEERING AND
SERVICES CENTER TYDALL AFB FL ENGIN.. R W RICE ET AL.
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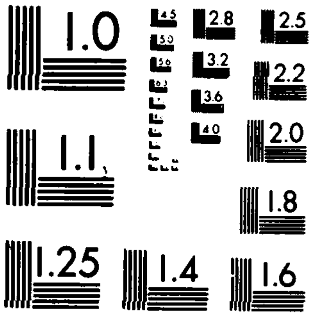

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A STUDY OF FACTORS AFFECTING SOOT FORMATION IN A SWIRL-STABILIZED COMBUSTOR

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APRIL 1983

FINAL REPORT
22 JANUARY 1980 - 30 SEPTEMBER 1982

APPROVED FOR PUBLIC RELEASE: DISTRIBUTION UNLIMITED

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REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER
ESL-TR-82-49

2. GOVT ACCESSION NO.
AD-A128 255

3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle)
A STUDY OF FACTORS AFFECTING SOOT FORMATION IN A SWIRL-STABILIZED COMBUSTOR

5. TYPE OF REPORT & PERIOD COVERED
FINAL REPORT
22 JAN 80 - 30 SEP 82

6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s)
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8. CONTRACT OR GRANT NUMBER(s)

9. PERFORMING ORGANIZATION NAME AND ADDRESS
HQ AFESC/RDVS
TYNDALL AFB FL 32403

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
JON: 01008002

11. CONTROLLING OFFICE NAME AND ADDRESS
AIR FORCE ENGINEERING AND SERVICES CENTER
TYNDALL AFB, FLORIDA 32403

12. REPORT DATE
1 April 1983

13. NUMBER OF PAGES
33

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)

15. SECURITY CLASS. (of this report)
UNCLASSIFIED

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; Distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

Availability of this report is specified on reverse of front cover.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)
Aerosol
Combustion
Combustor
Particle
Soot

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The effects of a variety of combustion and sampling variables on soot concentration and particle size distribution were studied using a swirl-stabilized laboratory combustor and an electrical aerosol analyzer (EAA). Tests were conducted over a range of reference velocities (2.5 to 10m/s) and equivalence ratios (0.05 to 0.3) for five gaseous fuels: propane, n-butane, ethylene, propylene, and 1-butene. Soot concentration and size were found to decrease with increases in either gas reference velocity or equivalence ratio. Alkenes produced more and larger soot particles than alkanes of corresponding carbon

Cont'd

number. *Sooting* tendency increased with increased carbon chain length. Probe placement and sample line length affected soot concentration measurements. Smoke number readings were obtained for comparison with EAA data.

Recommendations were made for further work on the topics lightly surveyed in this study and for extensions which could possibly answer important questions raised here concerning soot growth/oxidation phenomena.

PREFACE

This report was prepared at the Department of Chemical Engineering, Clemson University, Clemson, South Carolina and the Air Force Engineering and Services Center, Engineering and Services Laboratory (AFESC/RD), Tyndall Air Force Base, Florida 32403, under Job Order Number 01008002. This research was performed by a Summer Faculty Research Program Professor and AFESC personnel. This report covers work performed between January 1980 and September 1982. Capt Daniel D. Berlinrut was the AFESC (RDVS) project officer.

This report has been reviewed by the Public Affairs Office (PA) and is releasable to the National Technical Information Service (NTIS), where it will be available to the general public, including foreign nationals.

This technical report has been reviewed and is approved for publication.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

SECTION I

INTRODUCTION

The U. S. Air Force has a multifold interest in investigating the factors that affect combustion and soot formation in gas turbines. First, from a tactical standpoint, it is desirable to reduce the visibility of the exhaust plume from jet engines. Second, incandescent soot particles have high emissivity and transfer radiant energy from the flame to the jet engine combustor liner. This influences both the liner lifetime and the cooling air requirement affecting engine performance. The final concern is with the environmental impact of jet engine emissions, particularly during static engine tests. This last topic is important for both aesthetics and health reasons because most soot particles are of the proper size (0.005 to 0.1 microns) to be ingested deep into the lungs (Reference 1). It is also believed that the polynuclear aromatic hydrocarbons comprising or adsorbed on soot (Reference 2) are carcinogenic (Reference 3).

Several investigations of jet engine emissions concluded that combustor design modifications are the most effective method of controlling emissions (References 4 and 5), but the use of soot-suppressing fuel additives has recently received interest (References 6 and 7). The chemical properties of aviation fuels have also been shown to have a large influence on sooting tendency. In particular, hydrogen-to-carbon (H/C) ratio and weight percent hydrogen have been used to correlate feed properties and soot-related measurements such as flame radiation and opacity (References 8 and 9). The polycyclic aromatic content of a fuel has also been cited for its impact on soot formation (Reference 10). In addition to fuel properties, combustion conditions, e.g., temperature, fuel-to-air ratio, velocity, etc., have a major effect on the quantity and nature of soot produced. Increasing temperature has been shown to reduce soot formation in premixed flames. This is believed to be because, as temperature increases, the OH radical concentration in the flame increases more rapidly than that of soot precursors and these hydroxyl radicals are especially effective in the oxidation of soot precursors (Reference 11). However, an opposite effect of temperature has been reported in tests utilizing other combustor configurations (Reference 12). Fuel pyrolysis is believed to be the dominant process in soot formation in diffusion flames and the ultimate soot concentration in the exhaust is dependent on the relative global reaction rates and characteristic times for the competing pyrolysis and oxidation reactions. When gas reference velocity is increased, flame speed reduces the time available for pyrolysis and increases the probability of oxidizing soot precursors. In laminar diffusion flames, where the time for pyrolysis is long, molecular structure has been found to be more important than H/C ratio (Reference 12). For highly turbulent flames, where pyrolysis is rate - limited, the opposite is true, thus, it is questionable whether results from laminar flame studies should be used to predict turbine engine behavior. Only modest effects on sooting have been found (Reference 10) for fuel-to-air (F/A) ratio for lean flames ($F/A \simeq 0.01$).

Most studies of soot formation have involved relatively complex molecules or mixtures, e.g., actual jet fuel, and indirect methods of soot characterization, e.g., flame emission, opacity, and Smoke Number measurements (References 10, 13, and 14). While valuable, such studies do not provide: (1) information concerning the soot particle size distribution, (2) accurate measures of soot concentration, and (3) information on the sooting behavior of specific compounds. Such data are needed if a fundamental understanding of the roles played by combustor parameters and molecular structure is to be obtained. The investigation described here, hopefully, represents a first step in this direction.

SECTION II

OBJECTIVES

Objectives for this project existed on three levels. The first objective was to gain confidence in performing combustion aerosol measurements. The second objective was to gain an understanding of the soot formation mechanism in jet turbines by relating laboratory combustor results to operating variables. The final objective was to determine the effect of specific system parameters such as fuel structure, reference velocity, equivalence ratio, sampling system design, etc., on soot concentration and average particle size.

SECTION III

EXPERIMENTAL PROCEDURES

The experiments performed this summer involved a variety of equipment. A brief description of each of the major items will be given below, followed by a discussion of how they were used together in a typical case.

A. LABORATORY COMBUSTOR

All experiments were made using a swirl-stabilized laboratory combustor scaled down and modified by G.S. Samuelsen and his co-workers at the University of California, Irvine, from an original design used at the Air Force Aero Propulsion Lab. A detailed description of that combustor [illegible] been given elsewhere (Reference 15), thus only the most pertinent features [illegible] the scaled-down version will be described here. The combustor consisted o[illegible] 100 cm length of 5.7 cm outside diameter, 5.2 cm inside diameter, glass [illegible]bing attached by gasketed flanges to a stainless steel main housing which co[illegible]ned a 3 cm outside diameter centerbody that extended 6 cm axisymmetric[illegible] away from the flange. Fuel gas was injected into the glass tube throu[illegible] 0.13 cm inside diameter jet in the centerbody. Air was introduced through an annular drilled distributor plate surrounding the centerbody. A swirl was imparted to the air flow by a stainless steel swirl vane encircling the centerbody and equipped with blades slanted at 30°, 45°, or 60° (45° unless otherwise noted). The swirl vane was fitted snugly inside the glass tube 3 cm from the centerbody face. This geometry resulted in rather complex aerodynamics, including a turbulent annular "sheath" of air surrounding the combustion gases and a strong reverse flow near the radial center of the tube.

Pitot tube measurements taken at the mouth of the glass tube during cold flow tests confirmed the presence of a reverse flow at the center of the tube. This effect was also observed during studies at the University of California, Irvine (Reference 16). However, they found that the radial velocity profiles were considerably different during combustion runs. At a reference velocity of 7.5 m/s and an equivalence ratio of 0.05 the centerline velocity near the exit plane was negative during cold flow run, but positive during combustion runs. At a reference velocity of 15 m/s and an equivalence ratio of 0.05 a negative centerline velocity was still observed during combustion. Supporting equipment for the combustor included a rotameter and associated valves for metering gaseous fuel and a calibrated orifice/water manometer system for metering dry compressed air.

B. AEROSOL ANALYZER

A Thermo Systems Inc., Model 3030 Electrical Aerosol Analyzer (EAA) interfaced with an Apple II microcomputer and connected to an Integral Data Systems Model 445 Printer provided measurements for both soot concentration and average particle diameter, as well as bar graphs showing particle size distribution. Three calculational bases were used: (1) number of particles/cm^3, (2) aerosol surface area in μ^2/cm^3, and (3) aerosol volume in μ^3/cm^3. These will, hereafter, be represented by the symbols N, S, and V, respectively. The method of data reduction used by this system is Kapadia's (Reference 17) application of a statistical technique originated by Twomey (Reference 18). Details of the data reduction and operation of the EAA can be found elsewhere (Reference 19). Flow of air and aerosol to the EAA, induced by a vacuum pump, was measured using a mass flow indicator and adjusted to specifications using metering valves. The aerosol sample (4 +/-0.2 SLPM) passed through a diffusion drier and an aerosol charge neutralizer (Kr-85 source). The EAA was operated in the nine-channel mode in which it detected particles in the 0.01 to 1.0 μm size range. This is one of the few means of accurately measuring aerosol size distribution over this particle size range.

C. GENERAL OPERATING PROCEDURE

Typically, after the room exhaust fan had been turned on, fuel flow was established at the desired value and air flow was initially set at a very low value to facilitate ignition. Next, a small propane torch, was lit and its flame was directed toward the combustor tube mouth. Following ignition, air flow was rapidly raised to the desired value to prevent "burn-back" onto the centerbody. In most instances the sample probe used was a 0.32 cm inside diameter stainless steel nozzle (bent into the shape of a hook), which would ordinarily be used with a Hill In-Stack Air Sampler/Cascade Impactor. The other sample probe used is a water-cooled probe with inlets for NO_2 dilution. Unless otherwise stated, the sample line used was 5 feet of heated (150°F) 1/4-inch Teflon® tubing. On several occasions, Smoke Number measurements were made using a Roseco Model 473A Engine Smoke Emission Sampler and a Welch Densichron reflectance meter. The procedure followed was that set forth in the Aerospace Recommended Practice Manual (ARP 1179).

SECTION IV

RESULTS AND DISCUSSION

Given the relatively short research period available and the large number of potentially important parameters, it was decided that the optimum approach would be to first determine the effect of extraneous, e.g., sampling-related, factors; then to study two or three combustion parameters in detail. Specifically, the work was divided into two main areas, each with several subtopics as shown below.

1. Equipment Testing and Sensitivity Studies.
    a. Sample probe type and location
    b. Sample line length and temperature
    c. Sample dilution
    d. Fuel jet (orifice) size
    e. Swirl vane angle
2. Combustion Parameter Studies
    a. Reference velocity, ν
    b. Equivalence ratio, ϕ
    c. Chemical nature of the fuel
    d. Fuel additives

A. SAMPLE PROBE TYPE AND LOCATION

As mentioned earlier, crude pitot tube measurements of gas velocity profile showed a strong reverse flow near the tube center and a strong forward flow near the tube wall. Numerous tests were made to determine the extent to which this flow pattern affected soot measurements. For a relatively low velocity case, Figure 1 shows that at the tube mouth (axial distance = 0 inches) the aerosol concentration was only about one-third of that near the wall, while only 2 inches outside the mouth of the tube (axial distance =-2) the concentration, N, was nearly independent of radial position. Table 1 contains additional data which show that probe location mainly affected concentration, with S and V being more sensitive than N. Average particle size was only slightly dependent on probe position, with somewhat larger particles prevalent near the wall. These data also illustrate why the probe was generally located at the radial center and 3.5 inches away from the tube, i.e., (-3.5, 0), because at that axial location, EAA results were not sensitive to radial location.

Table 2 shows that differences in EAA measurements when using a 0.63 cm inside diameter impactor probe, rather than the standard 0.32 cm inside diameter probe, were relatively small and no consistent pattern was observed. The

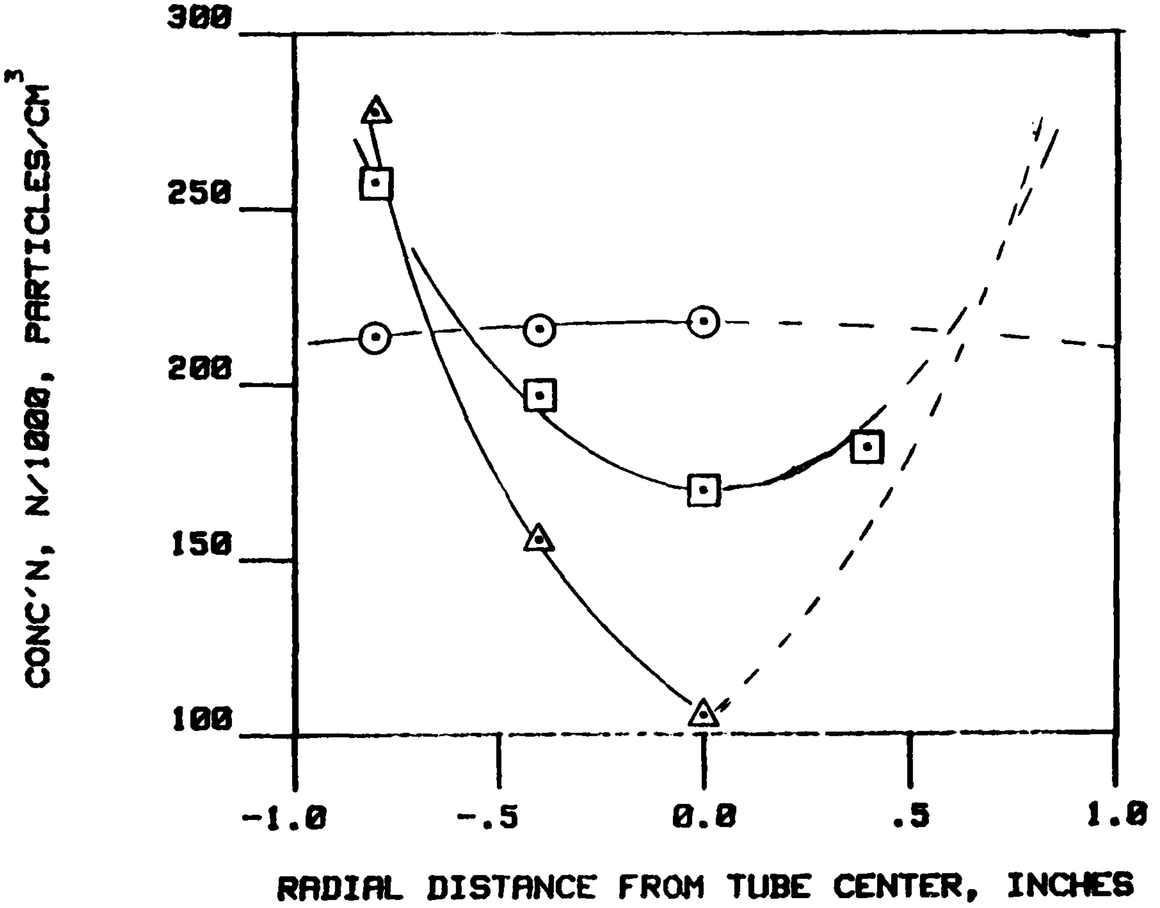

Figure 1. The Variation of Soot Particle Concentration with Probe Location. Propane at Ø = 0.3, V = 3.5 m/s; △, ⊡, and ⊙ correspond to axial distances of 0, 1, and 2 inches, respectively, away from tube end.

TABLE 1. EFFECT OF PROBE LOCATION ON PARTICLE CONCENTRATION AND AVERAGE PARTICLE DIAMETER

CONDITIONS AND PROBE LOCATION	PARTICLE CONCENTRATION			AVERAGE DIAMETER, um		
	$N \times 10^{-3}$	S	V	$d_N \times 10^2$	$d_S \times 10^2$	$d_V \times 10^2$
PROPANE, ϕ=0.3, V=3.5m/s:	**Parti-cles/cm^3**	μ^2/cm^3	μ^3/cm^3			
(0,0)	08	174	0.8	2.0	2.6	2.9
(0,-0.4)	158	295	1.5	2.1	2.7	3.2
(0,-0.8)	280	737	4.4	2.5	3.3	3.9
(-2,0)	220	450	2.3	2.2	2.9	3.3
(-2,-0.4)	218	464	2.4	2.3	3.0	3.4
(-2,-0.8)	216	438	2.2	2.2	2.9	3.3
ETHYLENE, ϕ=0.15 V=.5 m/s:						
(0,0) (1)	82	976	21	3.3	10.2	16.2
(0,-1) (1)	349	6599	146	5.3	11.0	16.4
(-3.5,0) (2)	348	4569	90	4.0	9.7	14.5
(-3.5,-1) (2)	433	6481	131	4.5	10.0	15.0

(1) at 85% dilution

(2) at 75% dilution

TABLE 2. EFFECT OF PROBE DIAMETER AND SAMPLE LINE LENGTH ON SOOT CHARACTERISTICS

SAMPLE LINE LENGTH, FEET	$N\times10^{-3}$ Particle/ cm^3	S μ^2/cm^3	V μ^3/cm^3	$d_N\times10^2$ um	$d_s\times10^2$ um	$d_v\times10^2$ um
5 (1)	516	2090	17.6	3.3	4.6	5.6
10 (1)	390	1709	14.0	3.2	4.5	5.4
15 (1)	282	1001	7.4	2.8	4.1	4.9
PROBE INSIDE DIAMETER, INCHES						
1/8 (1)	426	1240	8.1	2.6	3.6	4.3
1/4 (1)	497	1136	6.5	2.3	3.2	3.8
1/8 (2)	475	3090	34	3.6	5.8	7.3
1/4 (2)	572	4271	47	4.0	6.0	7.4

(1) Propane at $\phi=0.3$, $V=3.5m/s$, Probe at (-2,0)

(2) Propane at $\phi=0.3$, $V=2.5m/s$, Probe at (-2,0)

0.32 cm probe was chosen for general use because calculations indicated that it should most closely approximate isokinetic sampling for the gas velocity range used.

B. SAMPLE LINE LENGTH AND TEMPERATURE

Table 2 also illustrates that tripling the length of the sample line essentially halved the concentration of the sample reaching the EAA. Losses due to impaction on the sample line walls were modest, as might be expected for such small particles. Size distribution shifted slightly to lower diameter as line length increased. This presumably reflected the higher probability that large particles would be captured by the walls. In making these measurements it was necessary to adjust the EAA Aerosol Flow metering valve to offset the increased flow resistance of the longer sample lines. Tests showed no appreciable effect of sample line temperature until above 200°F, where the EAA Analyzer Current was typically 45 percent higher at 250°F than at 150°F. However, significant transients were observed when sample line temperature was increased, possibly indicating a desorption effect.

C. SAMPLE DILUTION

Once the aerosol concentration exceeded roughly 10^6 particles/cm^3, the EAA would generate an error message indicating failure to obtain a satisfactory iterative fit to the data. This condition was encountered for almost all samples from combustion of alkenes. In such cases it was necessary to dilute the "true" aerosol sample by introducing a known flow of N_2 through a tubing tee located near the probe. Several tests were made to determine: (1) which of the three concentration measures (N, S, or V) was best correlated with percentage of exhaust gas in the sample (=100 percent - percent dilution) and (2) whether dilution caused a major shift in size distribution due to condensation or other effects.

First, because of the sensitivity of EAA data to the aerosol gas sample flow rate (nominally 4 slpm) and the day-to-day shift in this quantity, it was necessary to periodically determine the dilution gas flow rate corresponding to virtually complete dilution. To do this, EAA Analyzer Current was measured at increasing dilution gas flow rates until it approached the "background" value, typically 0.01 to 0.04. Then a curve was fitted to the data and extrapolated to this nearly zero value. Figure 2 shows two such plots, with values of 3.7 and 4.5 slpm for 100 percent dilution. Figure 3 shows the variation of normalized concentration (diluted gas value/undiluted gas value) on all three bases versus percentage of exhaust gas in the sample for n-butane at =0.1 and v =7.5 m/s. A nearly identical plot was also obtained for propane at = 0.3, v =3.5 m/s, so Figure 3 is representative of dilution behavior over a wide range of variables. The most obvious feature of this figure is that S appears to vary in an almost perfect linear manner with percent dilution. The other two quantities, N and V, show definite nonlinear behavior; thus S is the best basis for use in comparisons where extrapolation of data from one dilution level to another is necessary. A firm explanation for this effect of dilution would require more investigation and it may be a function of dilutor geometry, dilution gas temperature, etc. However, one possible reason why S was the most linear in its behavior may be that the size distribution curve, dS/d log D versus D, was often nearly Gaussian, whereas those for N and V were generally

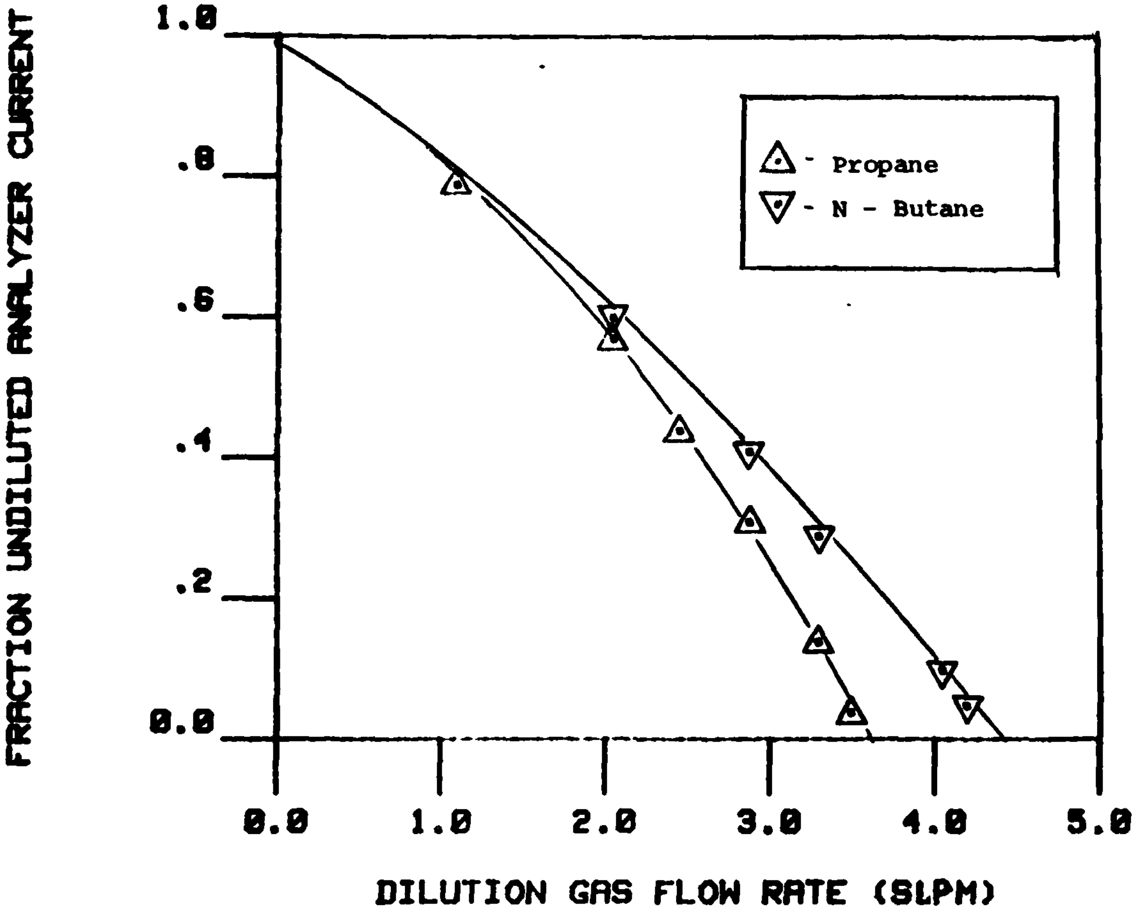

Figure 2. Aerosol Dilution Study; Determination of Hypothetical 100% Dilution Gas Flow Rate.

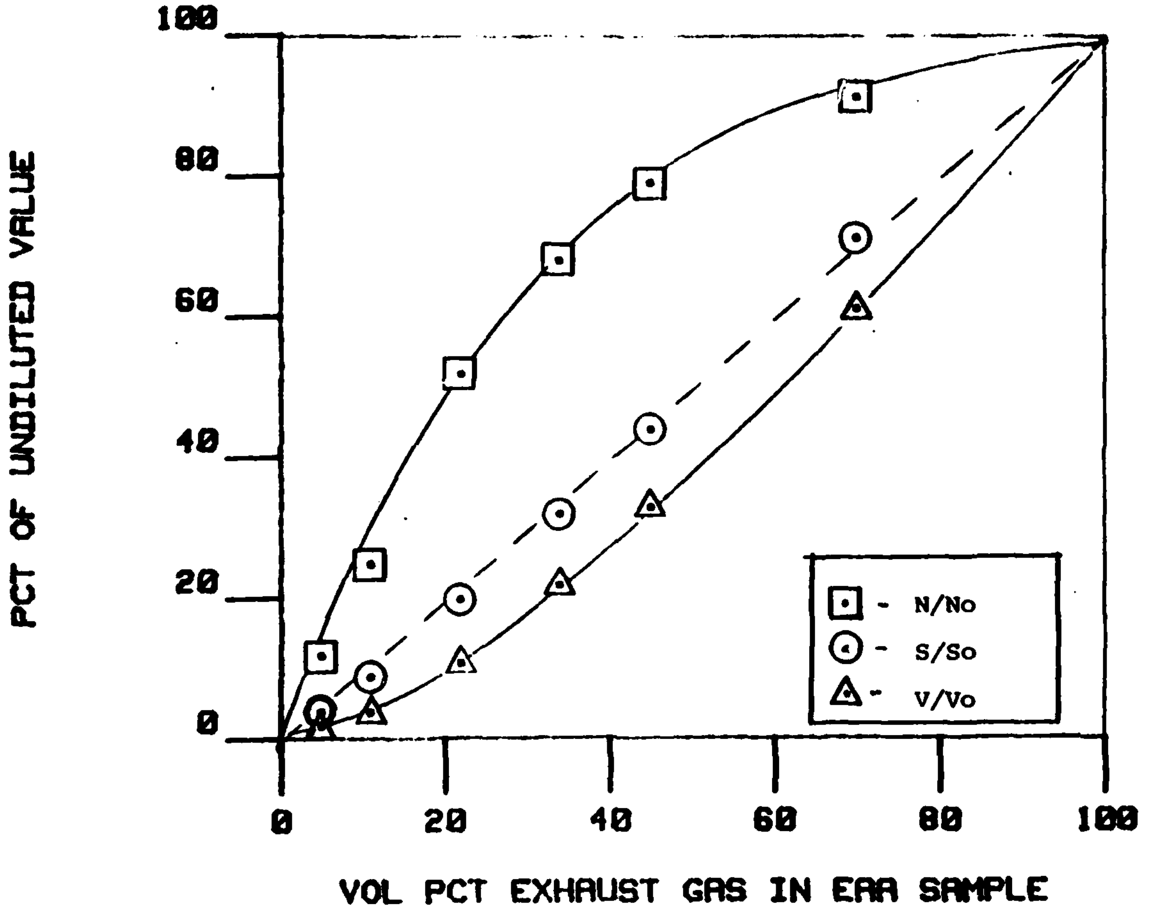

Figure 3. Dilution Study; The Variation of Various Measures of Soot Concentration with Volume Percent Exhaust Gas. N - Butane at ϕ = 0.1, V = 7.5 m/s.

more skewed, and thus more affected by dilution - induced changes at the extremities of the distribution. Some of these features can be seen in Figure 5, which shows bar graphs for two cases of dilution representing nearly a factor of 3, difference in percentage of exhaust gas. Figure 4 also illustrates that S varies linearly with dilution for a variety of cases. The dashed line on this plot illustrates that, at high percent dilution, it is important to get an accurate value for the hypothetical 100-percent dilution flow rate mentioned earlier. Ideally, important comparisons of sooting behavior should be made at identical dilution conditions.

D. MISCELLANEOUS EQUIPMENT STUDIES

A small number of runs were directed at qualitatively checking the effect of fuel orifice size and swirl angle. No appreciable effect on soot characteristics was found for propane when a 0.07 cm inside diameter fuel jet was substituted for the standard 0.13 cm one. However, the range of and for stable flame operation differed. Similarly, the major effect of using a 60° swirl rather than the usual 45° appeared to be a more restrictive operating range.

E. COMBUSTION PARAMETER EFFECTS

Velocity - Table 3 shows data for two cases which illustrate the effect of nominal (cold flow reference) gas velocity on both soot concentration and size. Case (a) for propylene at $\phi=0.1$ shows that increasing velocity over the range, 5.5 to 10 m/s, caused an apparent increase in N, a small decrease in S, and a somewhat larger decline in V. For all three calculational bases, the average particle size definitely decreased as velocity increased. The geometric standard deviation of the number distribution, σ_G, also decreased as velocity increased, indicating a narrower size distribution. In Case (b) for propane at $\phi=0.3$, all measures of concentration decreased significantly as velocity was increased over the range 2.5 to 3.5 m/s. The effect here was much more pronounced than in Case (a), possibly indicating that fuel-rich flames, e.g., Case (b), are more sensitive to velocity variations. Again, there was a significant decrease in average particle size as velocity increased. Several potential explanations may exist, but one contributing factor is the shorter residence time for all regions of the combustor flame, i.e., pyrolysis, carbon/hydrogen oxidation, etc., at higher velocity. A shorter time for pyrolysis would result in "nucleation" of fewer soot particles and less growth of any particles formed. Although probably not as important as the pyrolysis time effect, higher velocity would also reduce the time available for particle growth downstream of the visible flame. This could be checked by using combustion tubes of different lengths at a fixed gas velocity. Finally, at higher velocity, there is increased turbulence, and therefore better mixing of fuel, soot precursors, and oxidizer species (such as O_2, $O\cdot$, $OH\cdot$). One might expect lower particle counts and sizes due to improved mass transport resulting in an increase in the global reaction rate for oxidation.

1. Equivalence Ratio

Table 4 presents data for two cases similar to those used in Table 3, but here velocity was held constant and equivalence ratio, ϕ , (defined as actual fuel-to-air ratio divided by the stoichiometric fuel-to-air ratio) was varied.

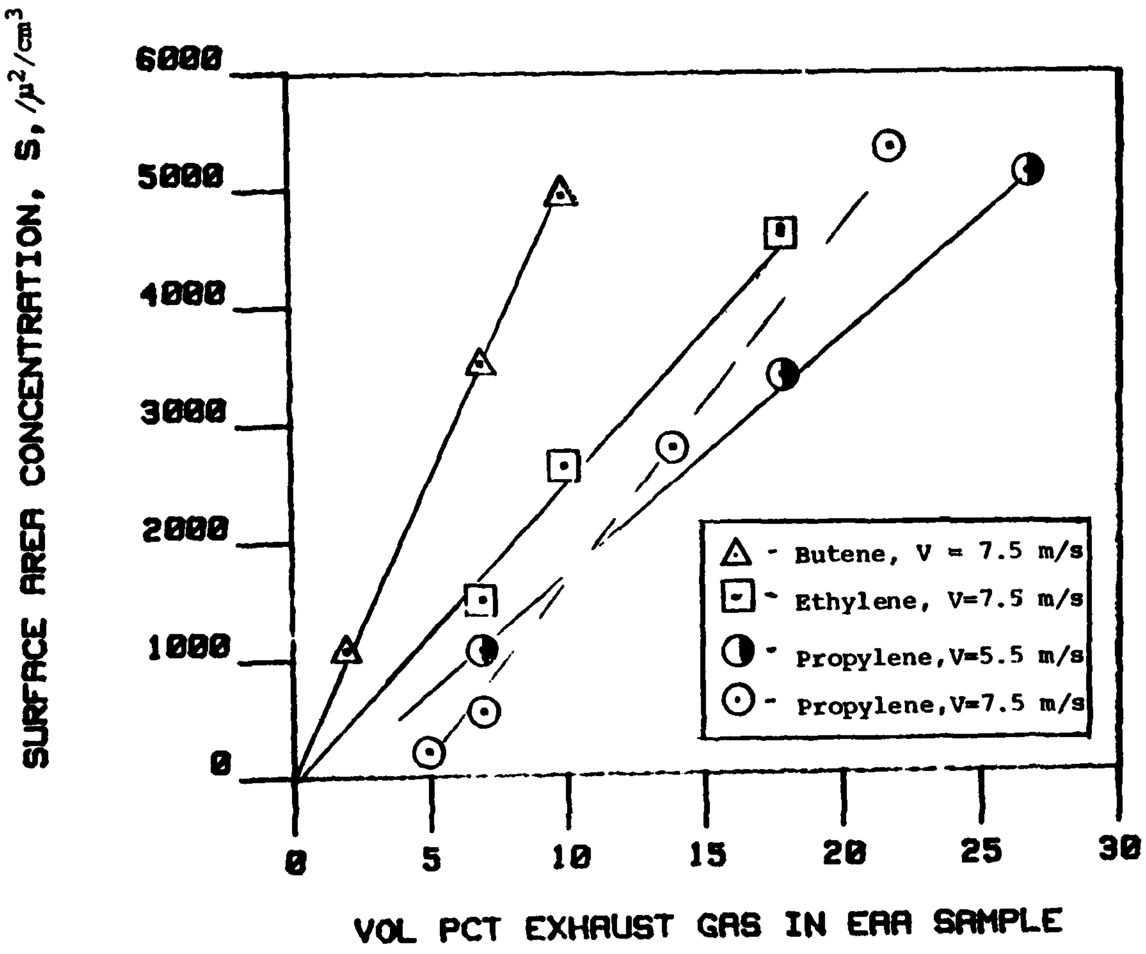

Figure 4. Variation of Aerosol Concentration (S, in μ^2/cm^3) with Volume Percent Exhaust Gas in the EAA Sample.

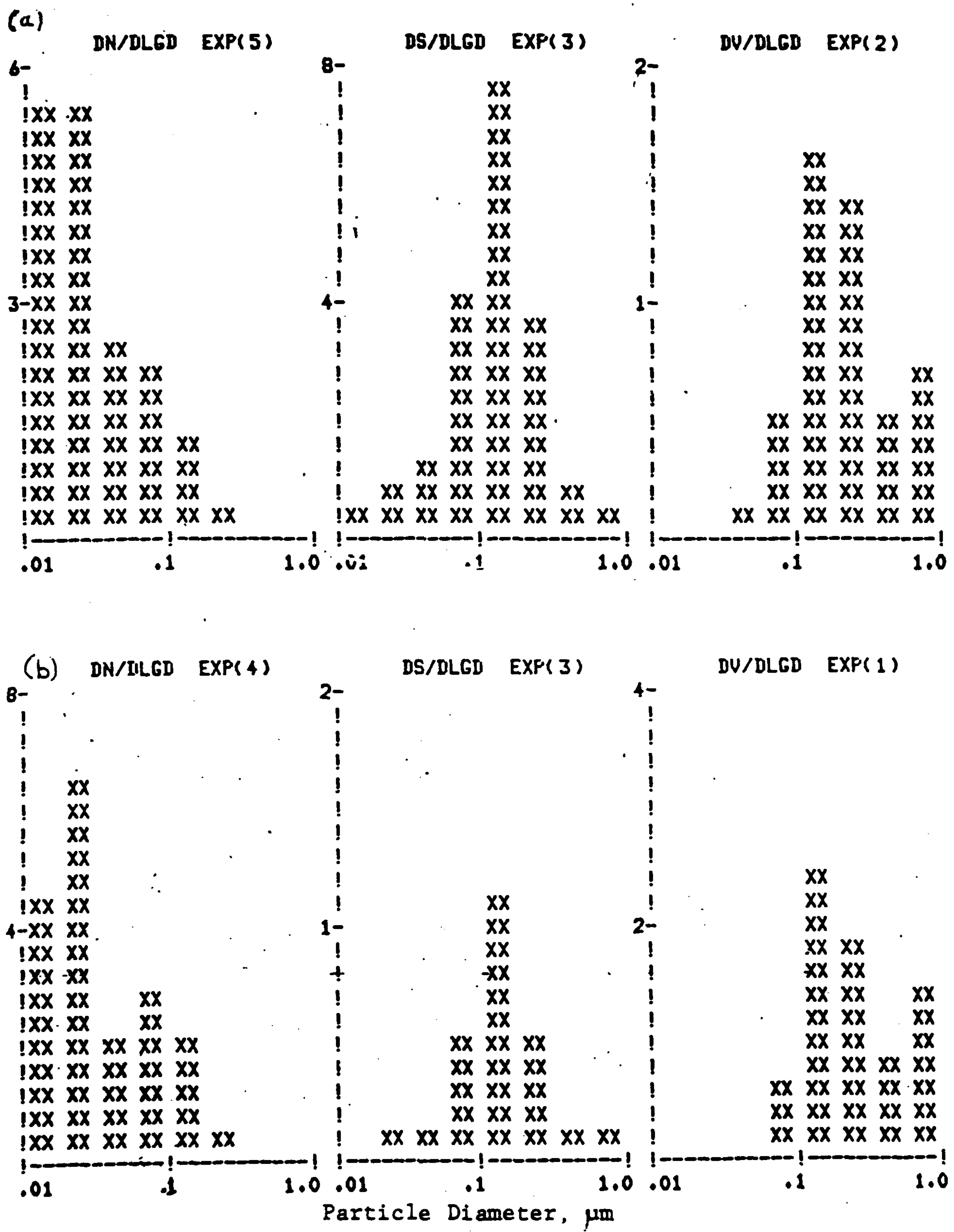

Figure 5. EAA Particle Size Distribution Data, Propylene at Ø = 0.1, V = 7.5 m/s: (a) at 82% dilution, (b) at 93% dilution.

TABLE 3. EFFECT OF GAS VELOCITY ON SOOT CHARACTERISTICS

a. Propylene, ϕ =0.1, Probe at (−3.5,0), 73% dilution

Velocity m/s	Analyzer Current	N/1000	S	V
5.5	4.08	261	5129	158
7.5	4.55	348	4998	140
10.0	5.10	461	4768	115

Velocity m/s	$100d_N$ μm	$100d_S$ μm	$100d_V$ μm	σ_G
5.5	3.5	14.7	23.2	2.53
7.5	3.2	12.9	22.1	2.31
10.0	3.0	10.8	19.6	2.11

b. Propane, ϕ =0.3, Probe at (−2,0)

Velocity m/s	Analyzer Current	N/1000	S	V
2.5	6.23	427	5141	76
3.0	5.62	434	4214	55
3.5	3.28	363	1840	17

Velocity m/s	$100d_N$ μm	$100d_S$ μm	$100d_V$ μm	σ_G
2.5	4.9	7.8	10.0	1.82
3.0	4.5	6.9	8.6	1.58
3.5	3.3	5.0	6.1	1.64

TABLE 4. EFFECT OF EQUIVALENCE RATIO ON SOOT CHARACTERISTICS

I. Propylene, V=7.5m/s, Probe at (-3.5,0) 73% dilution

EQUIVALENCE RATIO, ϕ	ANALYZER CURRENT	$N \times 10^{-3}$ Particles/cm^3	S μ^2/cm^3	V μ^3/cm^3
0.05	4.06	397	3953	102
0.10	4.55	348	4998	140
0.15	3.78	229	4076	106

EQUIVALENCE RATIO, ϕ	$d_n \times 10^2$ μm	$d_s \times 10^2$ μm	$d_v \times 10^2$ μm	σ_G
0.05	2.7	11.6	20.5	2.16
0.10	3.2	12.9	22.1	2.31
0.15	4.7	12.1	20.5	1.97

II. Propane, V=3.5m/s, Probe at (-2,0)

EQUIVALENCE RATIO, ϕ	ANALYZER CURRENT	$N \times 10^{-3}$ Particles/cm^3	S μ^2/cm^3	V μ^3/cm^3
0.11	6.55	433	5433	80
0.20	4.27	364	2916	34
0.30	2.68	309	1473	13

EQUIVALENCE RATIO, ϕ	$d_n \times 10^2$ μm	$d_s \times 10^2$ μm	$d_v \times 10^2$ μm	σ_G
0.11	5.0	8.0	9.9	1.65
0.20	4.0	6.3	7.7	1.63
0.30	3.1	4.8	5.9	1.59

For Case (I) as ϕ was increased from 0.05 to 0.15 for propylene, N decreased and d_N increased. Both S and V, as well as d_S and d_V, appeared to reach maximum values around $\phi = 0.1$. Case (II) shows the behavior most often observed when ϕ was increased; namely, all measures of concentration decreased. In addition, all measures of particle size also decreased substantially in this instance. At least two factors are affected when ϕ is increased. First and most obvious, is the fact that the fuel-to-oxidizer ratio increases. With everything else equal this might be expected to increase soot formation because more hydrocarbon per unit volume is introduced into the pyrolysis zone and soot precursors would be more likely to encounter and grow by capture of fragmented hydrocarbon species and less likely to encounter oxidizer species. Indeed, as ϕ was increased, the physical appearance of the flame changed, with the flame becoming longer and brighter yellow. This indication of increased incandescence would normally be assumed to mean increased soot production. Because this trend was not generally observed, one is forced to consider that the dominant effect of increasing ϕ, as it was actually done experimentally, was to increase the fuel velocity at fixed air velocity. As discussed in explanation of the effect of velocity, this would decrease the residence time of the fuel in the pyrolysis zone decreasing both soot nucleation and growth. However, this explanation is not supported by the small number of results obtained when using the smaller (0.07 cm) fuel orifice. No major differences in soot characteristics were observed, although the velocity of the fuel emerging from the jet would have differed substantially. This is an oversimplification of the complex fluid mechanics that undoubtedly occur just beyond the centerbody and within the visible flame. The observed behavior cannot be explained and further experiments are needed.

2. Chemical Nature of the Fuel

Of the five fuels studied two were alkanes (propane and n- butane) and three were alkenes (ethylene, propylene and 1-butene). These choices enabled two principal comparisons: (1) the effect of degree of unsaturation (related to H/C ratio) at fixed carbon chain length, e.g., propane versus propylene; and (2) the effect of chain length for a given molecular type or homologous series. Table 5 presents the results of this study summarized in the form of a "relative sooting tendency" based on n-butane as the reference. As an example, the relative sooting factor of 4.5 for 1-butene based on N means that, at the specified conditions, 1-butene produced soot with 4.5 times the particle concentration produced by n-butane. Several observations can be made. First, for a given molecular type, sooting tendency increased with increasing carbon chain length, with the effect on V being the most dramatic. Considering the soot-making mechanism involves, among other things, formation of polynuclear aromatics. This trend is understandable since the formation of multiring compounds requires the combination of carbon chains. The probability of ring closure is greater for longer chains because these have more potential contact points (carbon atoms) for incorporation into a growing network. A second observation is that, for a given chain length, alkenes produce more soot than alkanes. This is consistent with the picture of soot precursor formation occurring via successive elimination of hydrogen to form highly unsaturated polymeric species. Alkenes, being originally less saturated, might be thought of as already being "partially pyrolyzed." More importantly, the double bond of the alkenes make them more reactive for pyrolysis, ring formation, and/or attachment as an adduct to a growing carbonaceous residue. Interestingly, there is a much larger difference between 1-butene and propylene than between

TABLE 5. COMPARISON OF RELATIVE SOOTING TENDENCY FOR VARIOUS GASES BASED ON A NORMALIZED VALUE OF 1.0 FOR N-BUTANE*

I. ϕ=0.1, V=7.5m/s, Probe at (-3.5,0)

	RELATIVE SOOTING FACTOR BASED ON			
GAS	ANALYZER CURRENT	NUMBER CONCENTRATION	SURFACE AREA	VOLUME
PROPANE	0.8	0.9	0.7	0.7
N-BUTANE	1.0	1.0	1.0	1.0
ETHYLENE	7	3.0	18	70
PROPYLENE	6	2.5	19	100
1-BUTENE	11.5	4.5	33	150

*-Propane/butane comparison at 0% dilution.

*-Ethylene/propylene/butene/butane comparisons at 90% dilution. All data except for propylene taken on same day.

II. ϕ=0.3, V=3.5m/s, Probe at (-3.5,0)

	RELATIVE SOOTING FACTOR BASED ON			
GAS	ANALYZER CURRENT	NUMBER CONCENTRATION	SURFACE AREA	VOLUME
N-BUTANE	1.0	1.0	1.0	1.0
PROPYLENE	5.3	1.3	21	80
1-BUTENE	12.3	4.7	43	160

propylene and ethylene, although the chain length ratio is less (4/3 versus 3/2). In any case, the effect of molecular type is much greater than that of chain length. Given the large differences in combustor conditions for the two cases shown in Table 5, the agreement of relative sooting tendency values is surprisingly good.

Table 6 compares particle size data for the same cases presented in Table 5. Consistent with the previous discussion, an alkene produces much larger soot particles than its corresponding alkane. The effect of chain length is less obvious here than in Table 5, with 1-butene seemingly producing a slightly smaller average particle than propylene. In this particular case, the differences among these two alkenes are nearly within expected experimental error, and more testing would be needed to obtain statistically significant comparisons. In making such comparisons, particularly where dilution is involved, vital care must be taken to ensure that all sampling parameters, including EAA aerosol flow and probe location, be held constant.

F. COMPARISON OF EAA AND SMOKE NUMBER RESULTS

Table 7 presents EAA results (S and d_s) and Smoke Number values for several cases involving propane, ethylene, and propylene. S was chosen for the EAA data because, as a measure of surface area, it was most closely related to the factors affecting the light reflectance of a soot deposit on filter paper, which is also surface-related. Furthermore, since the EAA results for the alkenes involved diluted samples while those for Smoke Number did not, S was the obvious choice. As demonstrated earlier, only S can be safely extrapolated to the 0-percent dilution basis needed here. Given the narrow range of the results, all that can be said is that there is a rough correlation with a given unit of Smoke Number corresponding to an S increment of 600-900 μ^2/cm^3 for the alkenes, but only 350-400 μ^2/cm^3 for propane.

G. ADDITIONAL COMMENTS

Two other items deserve mention. One is that gas temperature measurements were occasionally made in the sampling region near the end of the combustor tube using a chromel-alumel thermocouple and a temperature gauge. Some of these results are presented in Table 8 to illustrate: (1) that steep axial and radial temperature gradients existed and (2) that equivalence ratio had a dramatic effect on exhaust gas temperature. Given the large differences in gas temperature near the tube exit, soot characteristics depended less on probe placement than might have been expected if particle growth by condensation was an important factor in this temperature range. Actually, any such effects could have been masked by the effect of the 150°F sample line. The use of a physically nonintrusive measurement technique such as laser-optical scattering might reveal differences that were obscured in this study.

Finally, an attempt was made to obtain soot concentration measurements, using a cascade impactor, for comparison with EAA results, but this proved unsuccessful. The glass fiber filter elements in the impactor adhered too strongly to the metal support plates, making neat removal impractical and impossible to accurately determine deposited soot in milligrams. This problem could probably have been solved but there was not enough time available.

TABLE 6. COMPARISON OF AVERAGE PARTICLE DIAMETER FOR SEVERAL FUEL GASES AT COMMON CONDITIONS

I. ϕ =0.1, V=7.5m/s, Probe at (-3.5,0)

GAS	AVERAGE PARTICLE DIAMETER			
	d_Nx10^2 μm	d_sx10^2 μm	d_vx10^2 μm	σ_G
PROPANE (1)	2.1	3.2	6.8	1.45
N-BUTANE (1)	2.4	3.6	6.7	1.47
N-BUTANE (2)	1.8	2.7	4.5	1.42
ETHYLENE (2)	3.0	9.0	16.3	2.02
PROPYLENE (2)	3.1	12.2	21.4	2.17
1-BUTENE (2)	3.3	10.7	19.1	1.96

(1) at 0% dilution

(2) at 90% dilution

II. ϕ =0.3, V=3.5m/s, Probe at (-3.5, 0)

GAS	AVERAGE PARTICLE DIAMETER (3)			
	d_Nx10^2 μm	d_sx10^2 μm	d_vx10^2 μm	σ_G
N-BUTANE	1.8	2.6	6.0	1.40
PROPYLENE	4.4	14.1	21.8	2.38
1-BUTENE	2.7	13.2	21.9	2.28

(3) at 75% dilution

TABLE 7. COMPARISON OF SMOKE NUMBER AND EAA RESULTS

(Smoke Number Samples were undiluted, EAA samples were diluted as indicated)

			EAA DATA		
RUN NO.	CONDITIONS	SMOKE NUMBER	ACTUAL S MEASURED μ^2/cm^3	S, EXTRAPOLATED TO 0% DILUTION μ^2/cm^3	$d_s x10^2$ μ
1	Propane, ϕ=0.11, V=3.5m/s, 60° Swirl, Probe at (-2,0), 15 ft. Sample Line	14	5119	5119	8.1
2	Ethylene, ϕ=0.15, V=7.5m/s, Probe at (0,0), 80% dilution	15	---------NOT MEASURED---------		
3	Same as Run 2, but Probe at (-3.5,0)	29	3542	17710	9.5
4	Same as Run 3, but V=5.5m/s	28	4569	22845	9.7
5	Propylene at ϕ=0.1, V=7.5m/s, Probe at (-3.5,0), 5 ft. Sample Line, 78% dilution	28	5389	24495	12.2
6	Same as Run 5, but ϕ=0.3, V=3.5m/s, 73% dilution	28	6201	24804	14.2
7	Same as run 6, but with 15 ft. Sample Line	30	---------NOT MEASURED----------		

TABLE 8. THE VARIATION OF GAS TEMPERATURE WITH PROBE LOCATION AND OTHER COMBUSTOR VARIABLES

CONDITIONS	PROBE LOCATION (inches, inches)	APPROXIMATE GAS TEMPERATURE, °F
Propane at ϕ=0.3, V-3.5m/s	(0,0)	1250
	(0,-1)	600
	(-1,-1)	400
	(-1,0)	850
Propylene at ϕ=0.3 V=3.5m/s	(0,0)	1250
	(-3.5,0)	500
Propylene at ϕ=0.1 V=7.5m/s	(0,0)	550
	(-3.5,0)	150
Propylene at ϕ=0.15 V=7.5m/s	(-3.5,0)	275

SECTION V

CONCLUSIONS

The following conclusions were reached.

1. Sample probe location and sample line length have a definite effect on EAA measurement of soot concentration, but a weaker effect on particle size distribution.

2. For a given gas and equivalence ratio, increasing the reference velocity decreases both soot concentration and average particle size, probably due to decreased residence time for soot nucleation and growth in the pyrolysis zone.

3. Soot concentration and particle size appeared to decrease with increasing equivalence ratio at constant air velocity. This observation runs counter to expectations based on a simplistic consideration of the probable effect of increased fuel concentration on soot formation. It may be related to the effect of fuel velocity cited above, but no convincing explanation is available at this time. More experimentation is needed on this topic to "decouple" the fuel velocity and equivalence ratio effects.

4. Sooting tendency and average particle size increase with the degree of unsaturation of the fuel molecule and, to a lesser extent, with carbon chain length. These trends are consistent with the mechanism of soot formation via growth of a hydrogen-deficient polymer.

5. EAA measurements of soot concentration on a surface area basis, i.e., S, correlate linearly with percent of dilution, while N and V do not. For this reason, results for diluted samples are best compared using S.

SECTION VI

RECOMMENDATIONS

The following are recommendations for future work.

1. The current study was a rapid survey of a multivariable research area; therefore, many topics only lightly touched here need further study. Additional runs could determine the importance of extraneous parameters such as: (1) probe type (isokinetic, water-cooled versus simple impactor), (2) combustor tube length, (3) swirl angle and placement on the centerbody, and (4) fuel jet size. The effect of tube length might be twofold. First, it affects the time available for condensation downstream of the visible flame. Second, because of the strong recirculation caused by the swirl, the proximity of the tube mouth to the flame might be important.

2. The effect of equivalence ratio on soot characteristics should be further studied to remove some existing ambiguities. Comparison with similar results using premixed flames could be useful.

3. To continue the work begun on relating sooting tendency to molecular structure, tests should be extended to include such gaseous compounds as isobutane, butadiene, and 1-butyne (ethyl acetylene). The isocompounds would help separate the effects of carbon number and continuous chain length. Tests of unsaturates such as dienes and alkynes could provide valuable insight into the soot growth phenomenon. Both are prime soot precursors.

4. Tests should be extended to liquid fuels. Soot from single compounds, mixtures of 3 or 4 components, and actual aviation fuel might be compared, using the EAA to extend the study of molecular structure on sooting.

5. The effect of various concentrations of additives such as ferrocene, metal naphthenates, and solutions of metal salts such as $Ba(NO_3)_2$ and $BaCl_2$, should be studied by conducting sooting experiments with these aspirated (atomized) into gaseous fuel or dissolved in a liquid fuel.

6. The Laser - Doppler Velocimeter/Particle Sizing System soon to be received at Tyndall AFB should be used to unobtrusively observe the critical soot-making regions in and around the flame. This should better elucidate the detailed interaction between convective fluid mechanics, molecular transport, and reaction kinetics. Spectroscopic measurement of the relative concentration of important highly reactive (very low concentration) species such as OH radicals and other chemical intermediates would prove interesting.

7. Finally, characterization by chemical means of soot deposited under extremely different conditions might be attempted. Even something as simple as determining the extent to which soot dissolves in a series of solvents could be revealing.

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