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NO. 582

SOOT PRODUCTION BY DIFFUSION FLAMES

**PART 2. EFFECT OF VARIOUS DILUENTS ON THE SMOKE POINT
OF LAMINAR ETHYLENE DIFFUSION FLAMES**

by

I. McLINTOCK and D. J. RASBASH

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Summary

The smoke points of laminar ethylene diffusion flames have been measured under various conditions of dilution. Nitrogen, argon, helium and carbon dioxide have been introduced as diluents to both the oxygen and fuel streams of the burners. In the former case measurements have been made wherever possible over the oxygen index range 0.12 to 0.45 (12% to 45% oxygen). Smoke points have also been determined during the addition of water vapour to a nitrogen-oxygen stream, over the same range of oxygen index values.

The shape of the smoke point against oxygen index curves is surprisingly complex. Dilution also caused temperature and structural changes in the flame. In the light of these results the factors believed to be of relevance to soot production are discussed.

The smoke point values were compared with previously recorded values, and appear to depend on variables such as burner design.

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Introduction

In Part 1 of this Report⁽¹⁾ the processes of soot production in diffusion flames were reviewed. It was indicated that quantitative information on rates of soot production was meagre; in the field of fire research it would be useful to have further quantitative information concerning the effects of additives and diluents. Part 2 of the Report now presents measurements of smoke points of ethylene diffusion flames to which various diluents have been added. Further work will be concerned with quantitative measurements of sooting rates at fuel flows beyond the smoke points.

Diffusion flames of gaseous fuels were selected as being most likely to lead to reproducibility of results. In one important respect, however, such a system differs from the more practical case of a burning solid fuel: the fuel flow is artificially controlled and does not depend on heat transfer processes from the flame. Addition of diluent to the flame will lower the flame temperature; in the case of solid or liquid fuels this will lead in turn to a decrease in fuel flow. It would be logical, therefore, to extend the investigations to liquid and/or solid fuels once the simpler system has been adequately studied. A further parameter, the dependence of fuel flow on heat transfer from the flame to the liquid or solid, will then be introduced.

Experimental

The flames were burned on a concentric-tube burner, shown schematically in Figure 1. The burner tubes were of stainless steel and provision was made for altering independently the diameter of both inner and outer tubes. The flames were enclosed in a 6 inch diameter cylindrical Perspex tube; the length of this enclosure could be varied to suit various sizes of flame. The aluminium plate which covered the top of the enclosure (Fig. 1) was fitted with a $1\frac{1}{2}$ inch diameter exhaust hole. Soot left the flames as a cylindrical column which finally passed through a $\frac{7}{8}$ inch diameter glass tube of length 2 inches. This glass tube was supported centrally in a position $\frac{1}{4}$ inch below the exhaust hole (Fig. 1). Such an exhaust arrangement was necessary for the subsequent experiments on sooting rates and was therefore adopted in the smoke point measurements. The two aluminium plates (Fig. 1) were fitted with grooves and asbestos washers; the Perspex enclosure fitted into these and was clamped tightly between them.

In preliminary experiments on soot collection it was found that the use of an enclosure of smaller diameter led to loss of soot by deposition on the containing walls. Further experiments showed that some recirculation of combustion products was inevitable when the 6 inch diameter enclosure was fitted, but that this could be minimised and standardised by suitably choosing the length of the Perspex tube. In all the smoke point experiments presented here the length of the Perspex enclosure was 12 inches.

The burner could be removed from the enclosure before ignition of the flame, in order to prevent the risk of a premixed explosion in the enclosure. Two burner sizes were used in the experiments, with different inner-tube diameters but

the same outer tube. The dimensions were as follows:

<u>Burner A.</u>	Inner tube, internal diam.	5.75 mm.
	external diam.	7.93 mm.
	Outer tube, internal diam.	19.90 mm.
<u>Burner B.</u>	Inner tube, internal diam.	8.09 mm.
	external diam.	10.10 mm.
	Outer tube, internal diam.	19.90 mm.

Ethylene was selected as a suitable fuel for initial study, partly because it is readily available in reasonably pure form and partly because of its unsaturation.

Ethylene, air, oxygen, nitrogen, argon, carbon dioxide and helium, compressed in cylinders were obtained from proprietary sources. All gases were used without purification other than filtration (cotton wool) and drying (silica gel). According to specification the ethylene was at least 98.2% pure, the major impurity being carbon dioxide (0.27%).

Gas flows were metered to a precision of $\pm 1\%$ by capillary flowmeters, calibrated by the soap bubble method. The flow lines were fitted with conventional reducing valves, manometers and needle valves. Dibutyl phthalate was used in all flowmeters and manometers. In appropriate cases gases were metered separately and passed through a mixing vessel before being supplied to the burner. All flows quoted refer to atmospheric pressure and $22 \pm 2^\circ\text{C}$.

In some experiments the metered gases were saturated with water vapour before entering the outer tube of the burner. This was achieved by passing them through a column of granular pumice-stone soaked in distilled water at a known temperature; the pumice stone column was drip-fed with distilled water to ensure that it remained continuously saturated.

In the present work gas flows were such as to produce laminar flames with heights of the order of 10cm. Approximate flame heights were measured by a scale attached to the Perspex enclosure.

Smoke points were determined visually by observing the exhaust gases against a white background, and corresponded to the fuel flow at which soot was just visible in these gases. Reproducibility was reasonable and identical results were obtained by different observers.

The maximum colour temperatures of the yellow luminous zone were measured by optical pyrometer in an attempt to find the relative effect of different diluents on flame temperature.

Results

(a) Addition of diluent to oxidant.

Metered oxygen flows were mixed with separately metered flows of nitrogen, argon, helium and carbon dioxide respectively. Smoke points were determined for oxygen indices over the range 0.12 - 0.45; oxygen index = oxygen flow / (oxygen + diluent) flow. The value of the oxygen index could be varied in three ways depending on which of the three variables, oxygen flow, diluent flow, or (oxygen + diluent) flow, was kept constant. In the case of nitrogen-oxygen mixtures all three methods were employed but in the case of the other three diluents only the oxygen flow was kept constant. The data from these experiments are given in Figs 2-7. The curves relating smoke point to oxygen

index are complicated in that they all exhibit either inflexion points or minima. Some aspects of the shape of these curves will be discussed later. It should be noted however that with the exception of oxygen helium mixtures (Fig. 5) a reduction of oxygen index below 0.21 increased the smoke point in every case.

For carbon dioxide-oxygen mixtures the flames did not smoke for any fuel flow or oxygen index combination corresponding to the range covered for the other diluents. In this case the flame was extinguished if the oxygen index fell below about 0.17. The effect of adding carbon dioxide as diluent to air is shown in Fig. 3. Smoke points were also determined for a range of oxygen-nitrogen mixture which had been saturated with water vapour at a temperature of 23°C. This is equivalent to the addition of 2.8% water vapour to the nitrogen oxygen mixture. These results are included in Fig. 2 and 3. Fig. 3 clearly shows that both carbon dioxide and water vapour had a much greater effect on the smoke point than did nitrogen.

The size and shape of the flame varied widely according to the conditions. This is illustrated by Figs. 8 and 9 which show flames with oxygen index 0.21 and nitrogen and carbon dioxide respectively as diluents; the changes in flame structure resulting from the change of diluent is evident. For a given diluent the flame size and structure also altered with change in oxygen index. In order to describe these alterations it is first necessary to describe in greater detail the smoking laminar flames as they occurred in the experiments; this is now carried out with reference to Fig. 10.

The narrow blue reaction zone (A) represents the region in which exothermic combustion of fuel (or degraded fuel) with oxygen occurs. On the model of Burke and Schumann⁽²⁾ this could represent the locus of points at which fuel and oxygen have interdiffused to give a combustible mixture. The top sections of this zone are not visible. It is possible that the zone does not close over the flame axis, but on the other hand the top sections might be masked by the yellow luminous zone (C); in this connection it is interesting to note that in Fig. 9 the blue zone encloses almost the entire flame.

Region B is a dark space, conical in shape. It is considered⁽¹⁾⁽³⁾⁻⁽⁵⁾ that the processes of carbon formation take place here, in the virtual absence of oxygen but in the presence of inerts and combustion products. Carbon combustion occurs in the yellow luminous zone (C) and possibly in the red luminous zone (D), from the tip of which a cylindrical column of soot emerges.

Dilution of oxidising atmosphere with nitrogen, argon or helium under the conditions corresponding to Figs. 2, 4 and 5 led to the following changes in the flame:

- (1) The reaction zone (A) thickened.
- (2) The dark zone (B) lengthened gradually from 1.5 cm at an oxygen index of 0.21 to approximately 3 cm at an oxygen index of 0.13.
- (3) The overall flame height, as measured to the top of zone (D) increased by about 60% over the oxygen index range 0.21 to 0.13.
- (4) Region C became less luminous but the red luminosity of region D changed to a dull yellow colour. The general effect appeared to be, that of decreasing overall luminosity while at the same time spreading it more homogeneously over the flame.
- (5) The fine structure (feathers of flame) of region D disappeared.
- (6) The flame widened and became smoother in outline.

- (7) At sufficiently low values of the oxygen index (approx. 0.13) the flame lifted off the burner and continued to burn at a distance of about 1 cm above it.
- (8) At oxygen index values of about 0.11-0.12 the flame became very weak, flickering and close to extinction.

The gradual change in flame structure with increasing dilution (at constant oxygen flow) is illustrated in Figs. 11 and 12. The corresponding flame for oxygen index 0.21 is shown in Fig. 8.

The maximum temperature (optical pyrometer) of the yellow luminous zone (C), measured at the smoke point, decreased gradually from 1190°C to about 1100°C over the oxygen index range 0.21 to 0.13. No significant differences were noted with change of diluent, with the exception of carbon dioxide; in this case the temperature at oxygen index 0.21 was 1000°C, decreasing to 920°C at an oxygen index value of 0.17. The measured temperature decreases will result both from the increasing dilution and from the increase in fuel flow corresponding to higher smoke points. The significance of the measurement is dubious because of the temperature variation throughout the flame.

At oxygen index values greater than 0.21 the flame structure altered gradually to that shown in Figs. 13 and 14. The blue reaction zone became shorter and thinner. There was a marked decrease in the length of the dark zone (B) and of the yellow luminous zone (C). The luminosity of region C increased and was fairly homogeneously spread over the zone. The red luminous region of Fig. 10 was replaced by a thin reddish-black cylinder enclosed in a thin sheath of yellow luminosity. A pencil of soot emerged from the top of this cylinder.

(b) Addition of diluent to fuel

Nitrogen, argon, helium and carbon dioxide also increased the smoke point of the ethylene diffusion flames when they were added to the fuel stream, burning in a constant air flow. Results are presented in Figs. 15 and 16. The addition of diluent to the ethylene also caused structural changes in the flame. The dark zone (B) lengthened and appeared to change in colour from black to reddish-yellow, although this latter effect might have been due to a mantle of luminous carbon particles in the lower edges of the flame. Increasing dilution was also accompanied by flame-flickering. The temperature (optical pyrometer) of the yellow luminous zone decreased by some 100°C over the range of dilutions shown in Figs. 15 and 16.

Discussion

From the results presented in Figs. 2-5 it is evident that the effect of a diluent on smoke point is not straightforward. It is clear that changes in flame temperature resulting from changes in dilution are insufficient to explain the smoke point curves. In particular the curves (Figs. 4 and 5) for argon and helium dilution are quite different although these gases have equal heat capacities and might be expected to alter flame temperature by equal amounts.

A major difference between helium molecules and those of the other diluents is that of size. The small size of the helium molecule will result in a higher diffusion velocity and probably a higher concentration of helium molecules, relative to the other diluents, within the flame. This leads to the suggestion that diffusional processes involving diluent molecules are of importance in the soot formation process. This suggestion is substantiated by the fact that the smoke point curves are influenced by the dimensions of the burner; diffusion should be influenced by changes in burner dimension. It is noteworthy that the effect of changing fuel-tube diameter is most pronounced in the case of helium.

It was suggested in Part 1⁽¹⁾ that inert diluents might suppress carbon formation in region B (Fig. 10) of the flame by acting as third bodies in the removal of free radicals involved in the carbon formation process. In this respect the effectiveness of a diluent will depend on its molecular size and on its concentration within the flame. As mentioned previously, molecular size will itself determine concentration since the diluent molecules reach the flame by diffusion.

Some information is available⁽⁶⁾⁽⁷⁾ on the relative efficiencies of these molecules in halogen atom recombination reactions. For example, in iodine atom recombination, the relative efficiencies are approximately He : A : N₂ : CO₂ : H₂O = 1 : 2 : 2.5 : 8.0 : 29 according to Russell and Simons⁽⁷⁾. If these values are assumed to be approximately relevant to the present case helium should suppress soot formation to a smaller extent than the other diluents in the absence of other factors. If diffusion rates are considered, on the simple basis of being inversely proportional to the square root of the molecular weight, the overall relative efficiencies as third bodies should be of the order: He : A : N₂ : CO₂ : H₂O = 1 : 0.6 : 0.9 : 2.4 : 14.

Another mechanism of smoke suppression which could operate for carbon dioxide and water vapour but not for the other diluents is that the smoke particles are oxidised in regions C and D by the diluent. If this effect is not controlled by diffusion assuming instantaneous reaction in the flame then it might be expected that carbon dioxide would be as effective as water vapour; if diffusion rate into the flame were the controlling factor then carbon dioxide would be expected to be 0.7 times as effective as water vapour. It is assumed that in oxidising the smoke particles the carbon dioxide and water vapour are reduced to carbon monoxide and hydrogen.

An indication of the mechanisms which actually operates in the flame may be obtained by comparing the predicted relative effectiveness of the diluents according to the different mechanisms, with that actually obtained in the experiments. Some predicted relative effects of the three diluents nitrogen, carbon dioxide and water vapour are summarised in Table 1.

Table 1
Predicted relative effects of nitrogen
water vapour and carbon dioxide

Mechanism, or sum of mechanisms	Predicted relative effect of N ₂ : CO ₂ : H ₂ O
(a) Chain breaking in region B	1 : 3.3 : 12
(b) Temperature reduction in all regions (proportional to atomicity)	1 : 1.5 : 1.5
(c) Chain breaking in region B	diffusion controlled {
(d) Temperature reduction	
(e) Oxidation in region C and D	1 : 1.2 : 1.8
(f) Oxidation in region C and D (diffusion controlled)	0 : 1 : 1
(g) Mechanism (a) + (e)	0 : 0.7 : 1
(h) Mechanism (a) + (e)	1 : (3.3 + n) : (12 + n)
(i) Mechanism (b) + (e)	1 : (1.5 + n) : (1.5 + n)
(j) Mechanism (d) + (f)	1 : (1.2 + 0.7n) : (1.8 + n)
(k) Mechanism (d) + (e)	1 : (1.2 + n) : (1.8 + n)

(N.B. - n is an arbitrary number dependent on the relative extent to which the two mechanisms are involved).

The experimental ratio obtained from the increase in the smoke point by the addition of the diluents to air as shown in Fig. 3 with 2.8% added diluent is 1 : 45 : 38. This result fits best mechanisms (h) and (k) in table 1, which would predict ratios of 1 : 45 : 45 and 1 : 45 : 46 respectively. The value of n calculated for these two mechanisms are 4.35 and 4.4 respectively which suggests that the oxidation effect of both water vapour and carbon dioxide is much more important than either the chain breaking effect or the temperature effect of any of the diluents tested.

Addition of diluent to the fuel will result in different contours of diluent concentration within the flame and the diluent molecules will not have been heated by passage through the reaction zone. It is interesting that a saturation effect is observed under these circumstances. One possible explanation of the smaller effect of helium as compared to that of argon or nitrogen (Figs. 15 and 16) would be that it diffused outwards from the flame much more readily. Similar considerations would explain the larger effects observed (Figs. 15 and 16) when diluents are added to the fuel stream of Burner B as compared to that of Burner A (Burner B has the larger fuel-tube diameter and therefore a greater residence time for diluents within the flame). A full investigation would require concentration and temperature measurements at points throughout the flame for different conditions of dilution.

The temperature factor, third body factor and the oxidation factor have been mentioned above and in Part 1(1). Other possible factors are outlined below:

(1) The effect of altering oxygen index by varying the oxygen flow is illustrated in Figs. 6 and 7. It would be premature to discuss these figures in any detail but the minima in the curves show the effect to be a composite one. A possible explanation of the minima might be that at higher flow rates of oxygen the soot formed is more easily oxidised in the flame as the oxygen flow is increased but at low flow rates of oxygen, the effect of the diluent in preventing soot formation becomes more important as the oxygen flow is decreased.

(2) The length of the carbon formation zone (B) increases with increase in dilution. This will be a factor tending to promote soot formation because of the increased residence time of the fuel molecules in the zone. The corresponding, but relatively smaller, percentage increase in total flame length will counteract this by tending to increase the amount of carbon combustion.

(3) In Figs. 2, 4 and 5 the total flow in the outer tube of the burner increases by a factor of more than three over the oxygen index range 0.40 to 0.13. In Fig. 5 the same flow decreases by less than 50% over the same range. Such changes in flow may well influence soot production in the flame and must be considered as a possible additional factor.

(4) It is likely that the carbon formation zone (B) of the flame is heated largely by inward diffusion of hot molecules of diluent and combustion products. The thermal conductivities of these molecules will therefore determine the temperature of this zone and this could be yet another way by which the molecular size of the diluent will influence soot formation. There is an obvious relation between this and the temperature and diffusion-rate factors.

In summary, the following factors are considered to be of possible relevance to soot production in the laminar diffusion flames studied:

(1) Flame temperature. The heat capacity and thermal conductivity of the diluent are relevant properties here.

- (2) Molecular size of diluent, a property which will determine the concentration of the diluent within the flame, and also its efficiency as a third body in radical recombinations.
- (3) Oxygen content of diluent.
- (4) Oxygen flow.
- (5) Flame length and structure.
- (6) The total flow of gas from the burner.
- (7) The burner dimensions, in so far as these will influence some of the factors enumerated above.

Mention should be made of the actual values of the ethylene flows at the smoke point. If S is defined as:

$$S = \frac{\text{fuel flow at smoke point}/\text{O}_2 \text{ flow at smoke point}}{(\text{fuel flow}/\text{O}_2 \text{ flow})_{\text{stoichiometric}}}$$

The smoke point for oxygen index 0.21 and oxygen flow 210 cm³/min occurs at an S value of 1.97 for Burner A and 2.04 for Burner B. This is considerably higher than Schalla and McDonald's⁽⁸⁾ value (computed from Fig. 6 of their paper) of about 0.4 for an ethylene diffusion flame i.e. considerably lower than stoichiometric. According to Norrish, Porter and Thrush⁽⁹⁾ the corresponding S value for a premixed ethylene-oxygen explosion is 3.0 and this value would probably increase on the addition of nitrogen. It is possible that the burner of the present experiments allowed more efficient mixing of fuel and oxygen, and less dilution of the flame with recirculated combustion products, than did that of Schalla and McDonald⁽⁸⁾. Barr⁽¹⁰⁾ used a burner apparently similar to that of the present experiments and his S value for n-butane was approximately 0.65 which does not compare favourably with Schalla and McDonald's value of about 0.25 for the same fuel. Again their value is the lower one. It seems as if the value of S depends on the design of the burner.

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Acknowledgment

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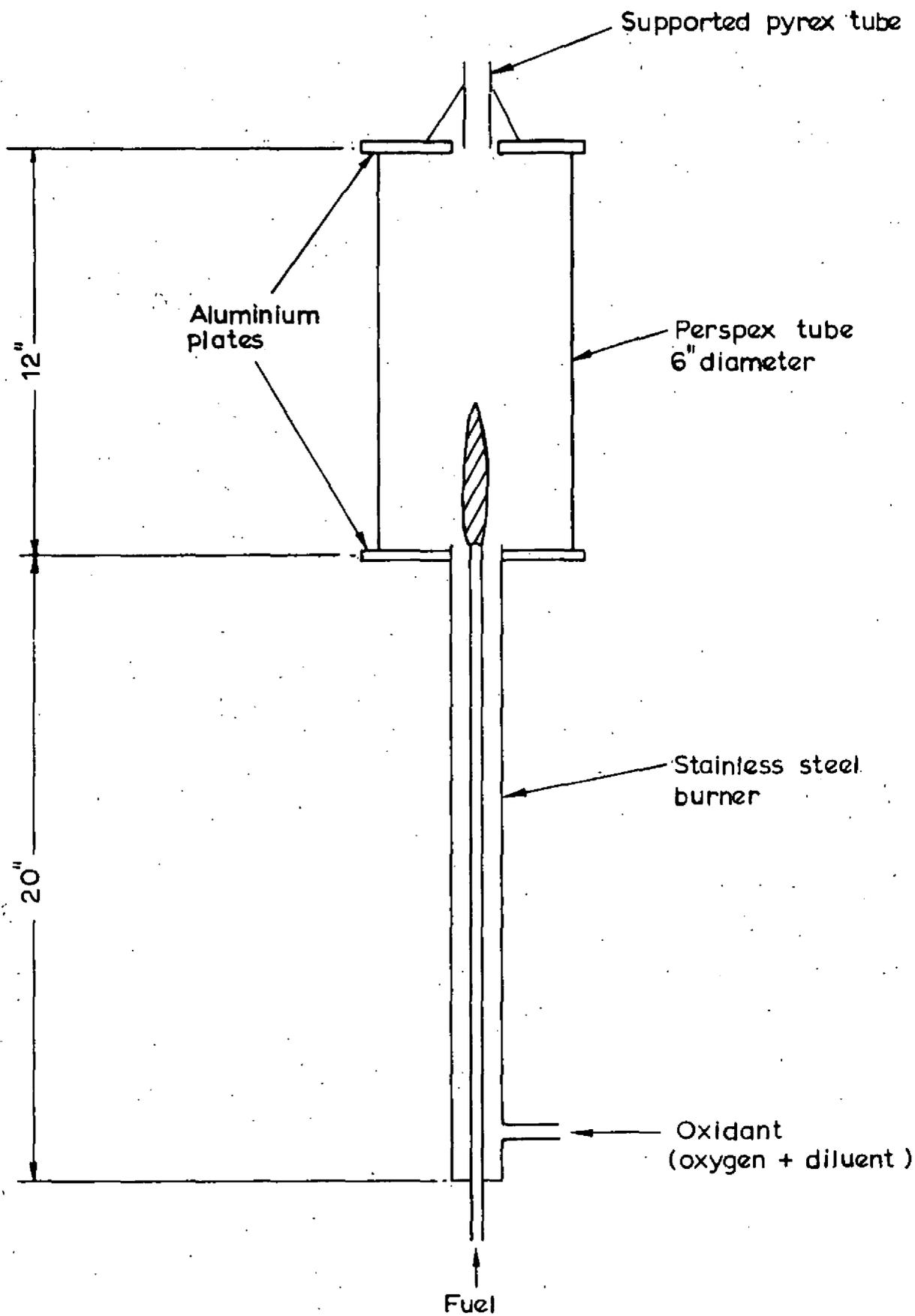
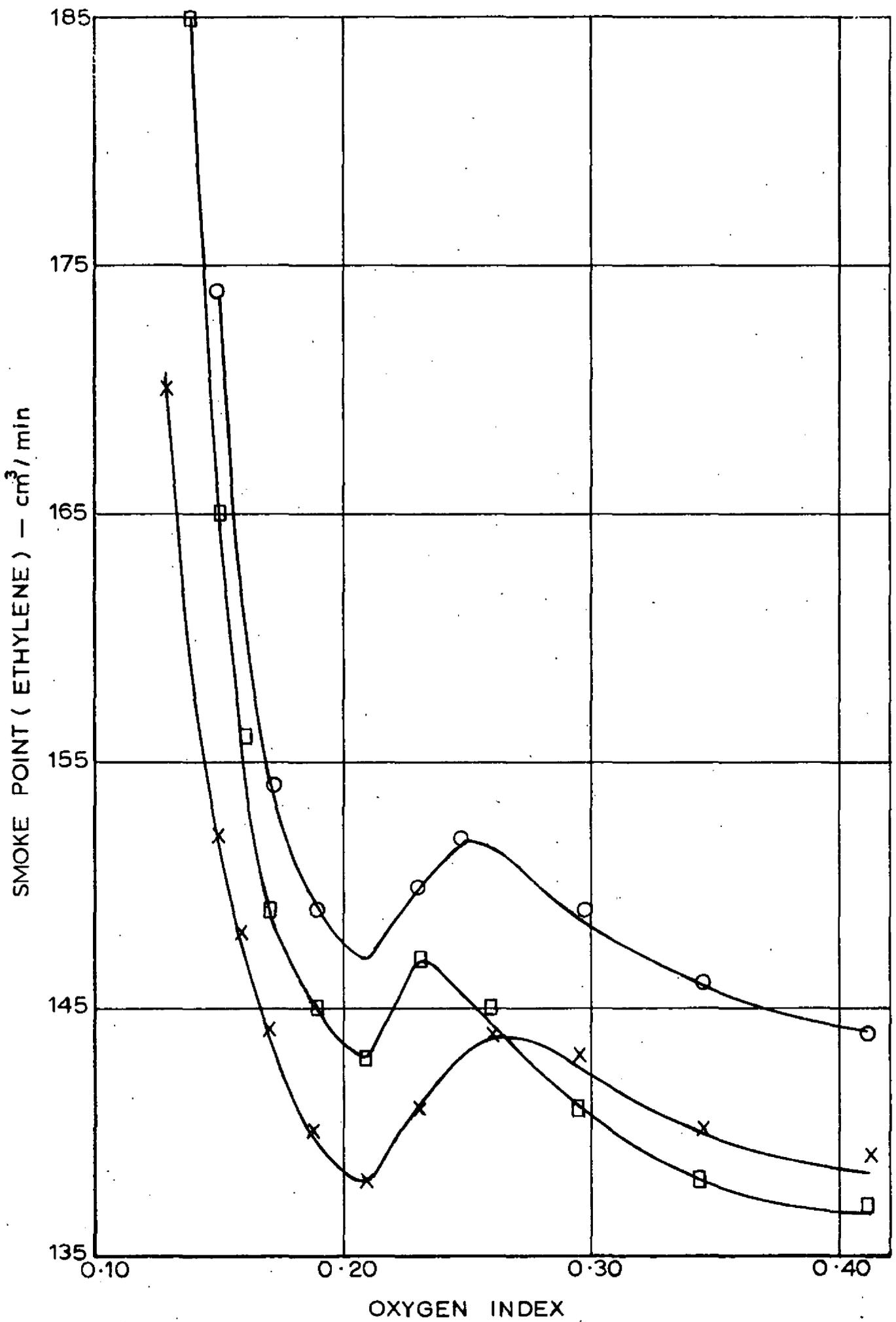


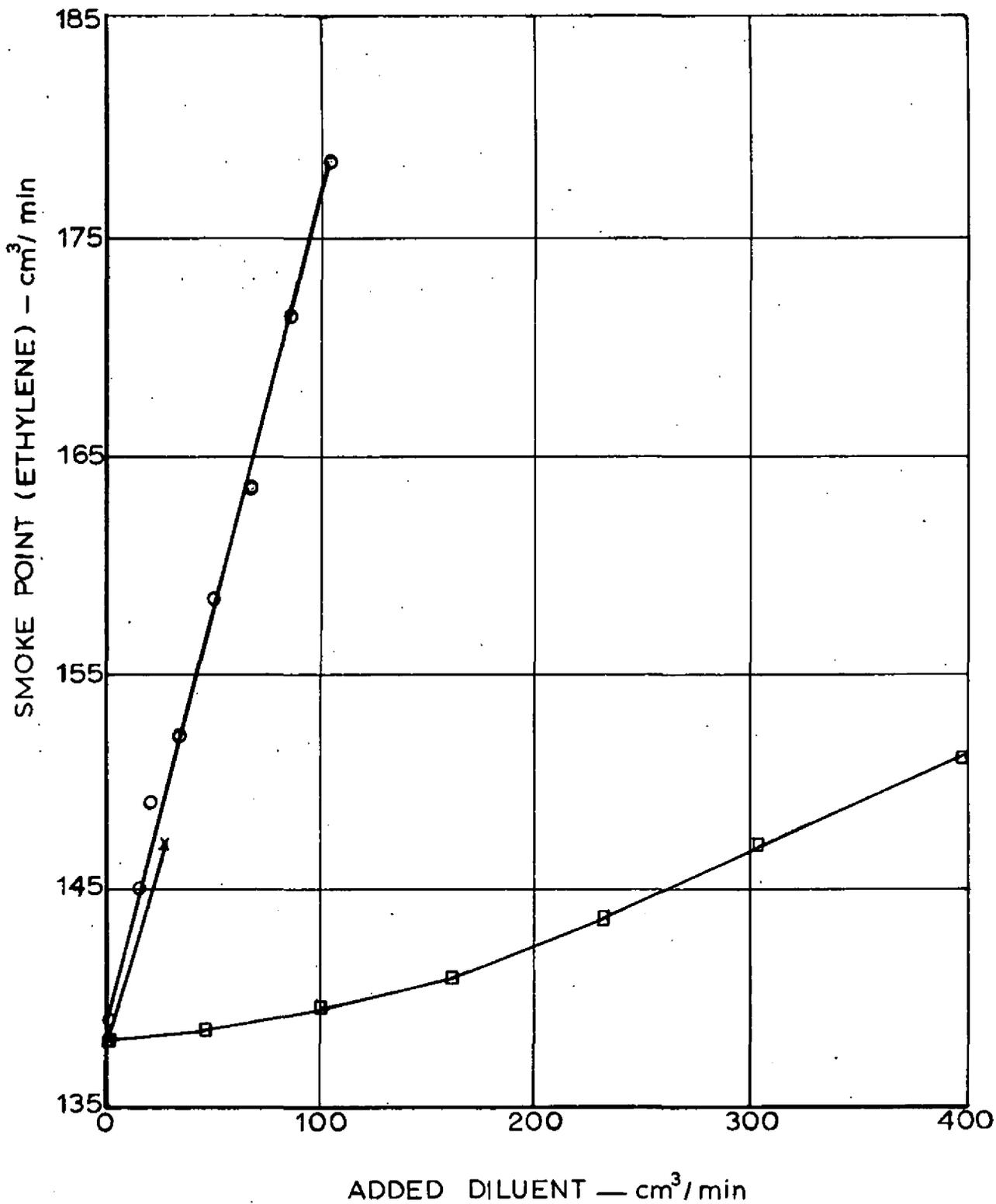
FIG.1. DIFFUSION FLAME BURNER APPARATUS



O Burner A 2.8% water vapour added
 X Burner A
 □ Burner B
 Oxygen flow constant at $210 \text{ cm}^3/\text{min}$
 Nitrogen flow varying

FIG.2. INFLUENCE OF OXYGEN INDEX ON SMOKE POINT

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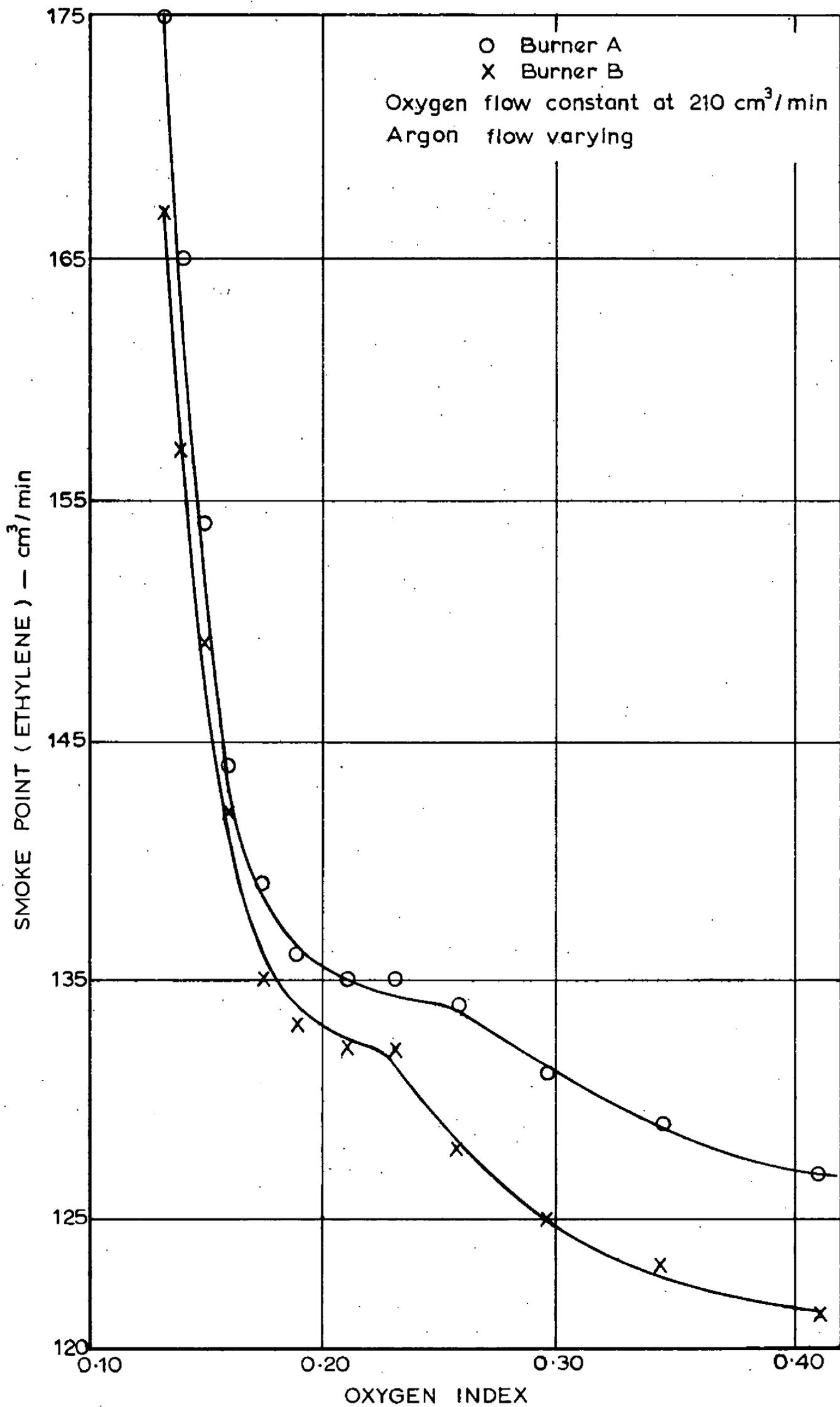
Burner A

- o Carbon dioxide
- x Water vapour
- Nitrogen

RATE OF FLOW OF AIR*—1000 cm³/min

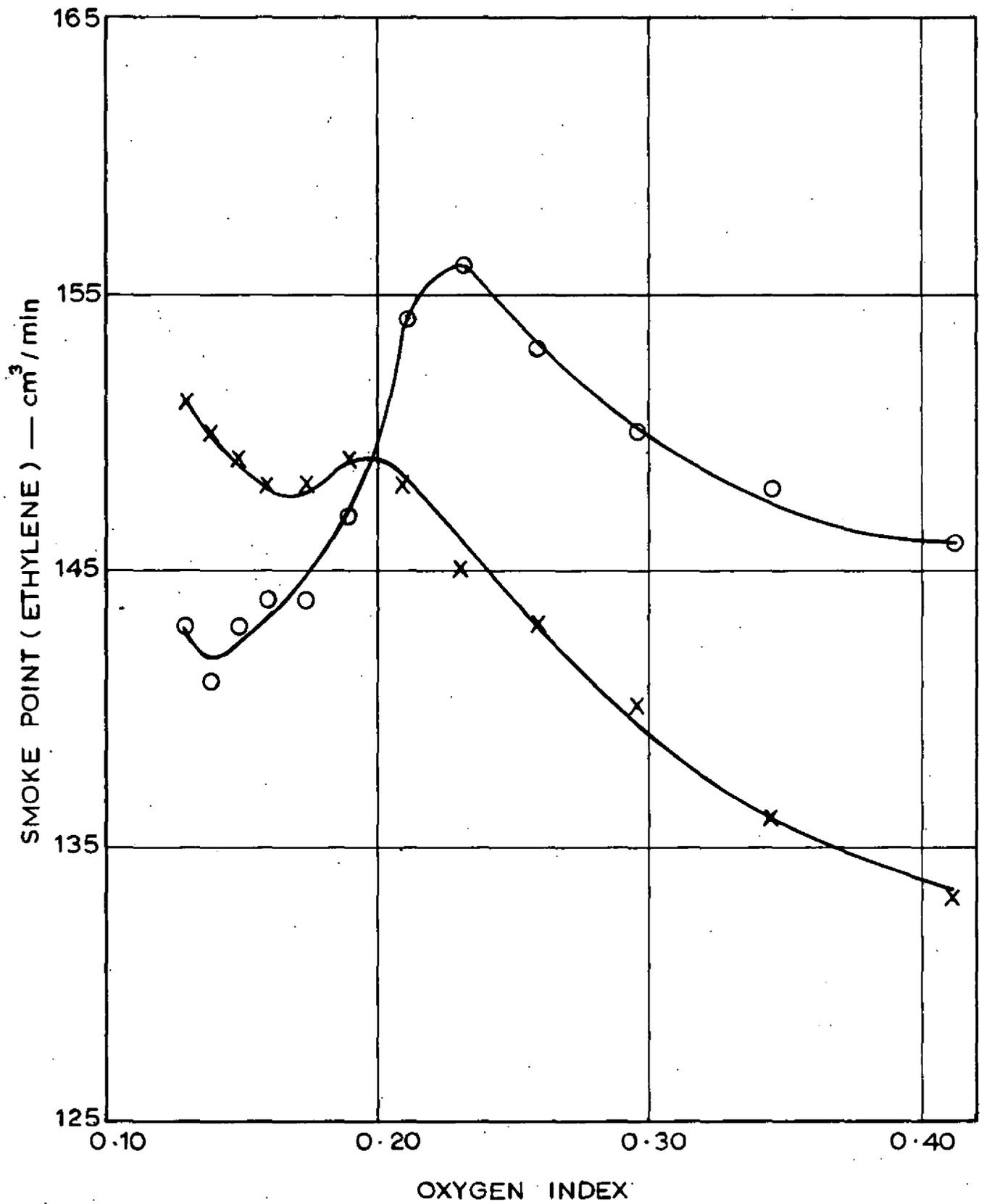
*210 cm³/min oxygen 790 cm³/min nitrogen
for water vapour and nitrogen curves

FIG.3. EFFECT OF ADDED CARBON DIOXIDE, WATER VAPOUR AND NITROGEN ON THE SMOKE POINT OF ETHYLENE-AIR FLAMES



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FIG.4. INFLUENCE OF OXYGEN INDEX ON SMOKE POINT



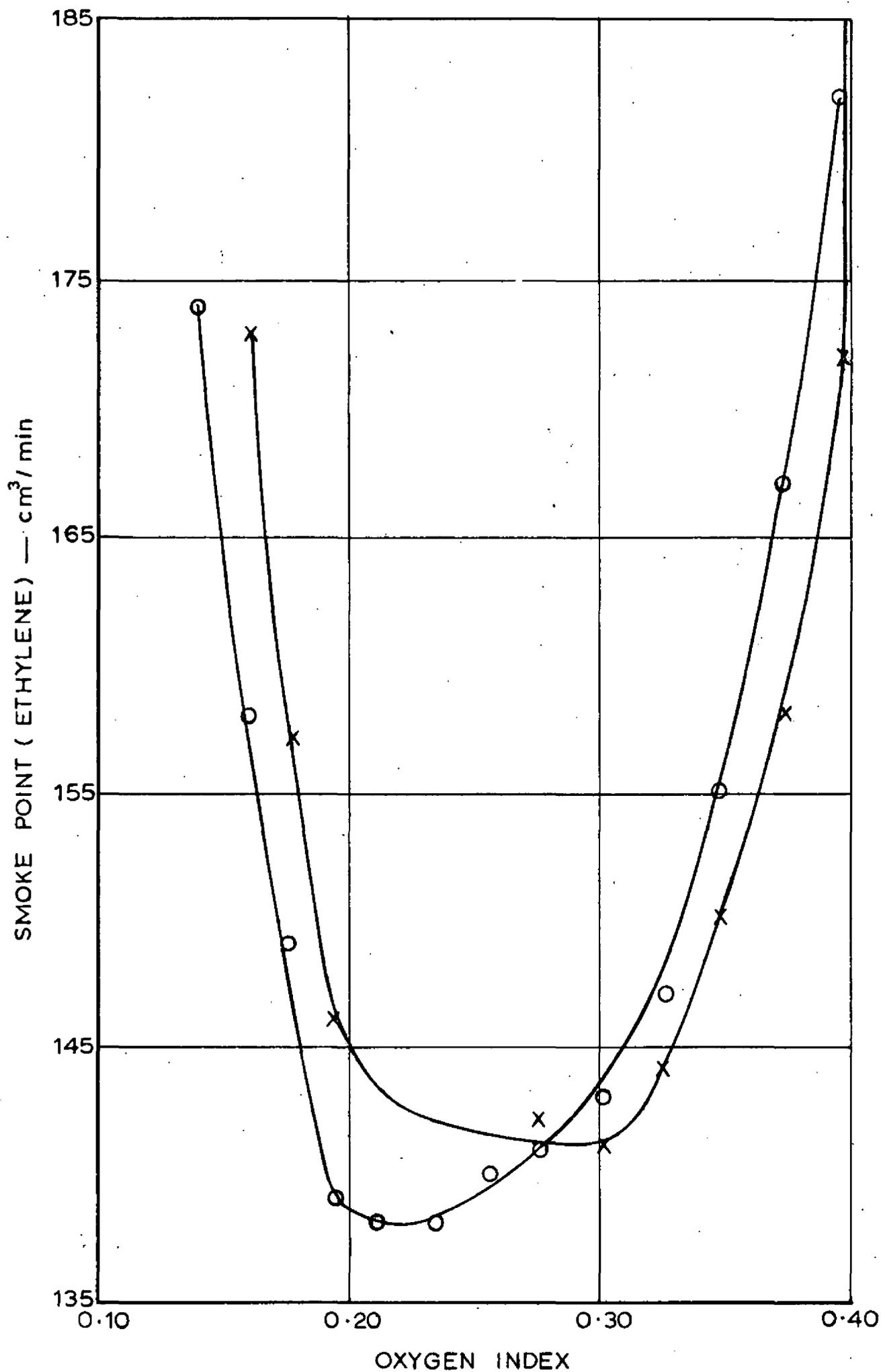
O Burner A

X Burner B

Oxygen flow constant at $210 \text{ cm}^3/\text{min}$

Helium flow varying

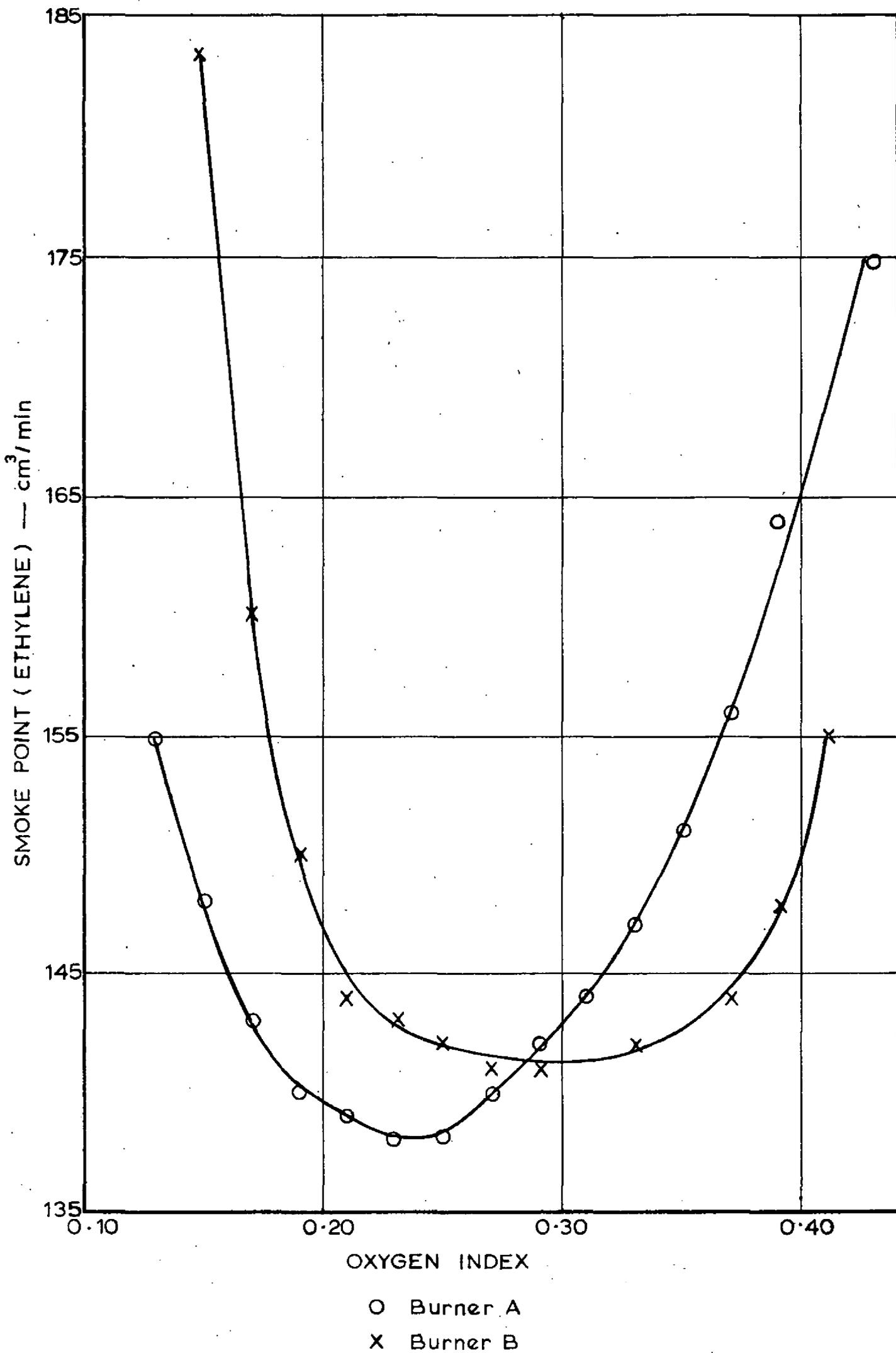
FIG.5. INFLUENCE OF OXYGEN INDEX ON SMOKE POINT



O Burner A
X Burner B

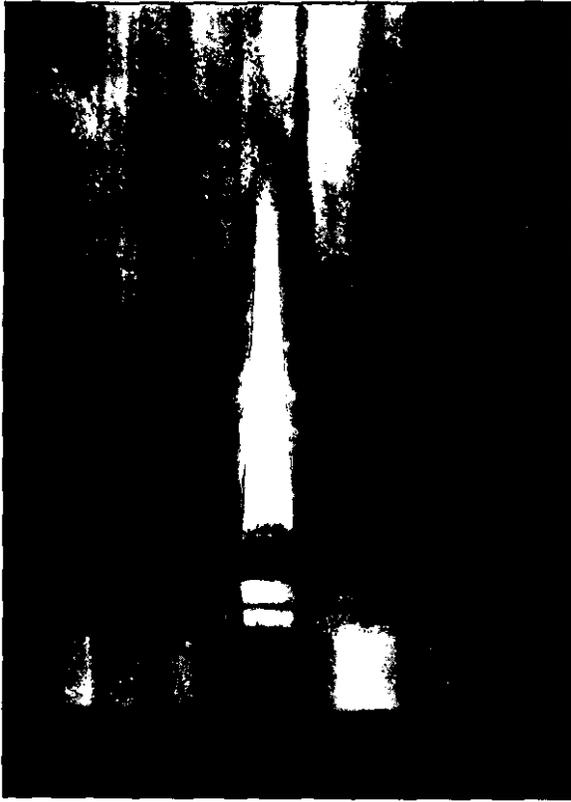
Nitrogen flow constant at $790 \text{ cm}^3/\text{min}$
Oxygen flow varying

FIG.6. INFLUENCE OF OXYGEN INDEX ON SMOKE POINT



Total (nitrogen + oxygen) flow constant at 1000 cm³/min

FIG.7. INFLUENCE OF OXYGEN INDEX ON SMOKE POINT



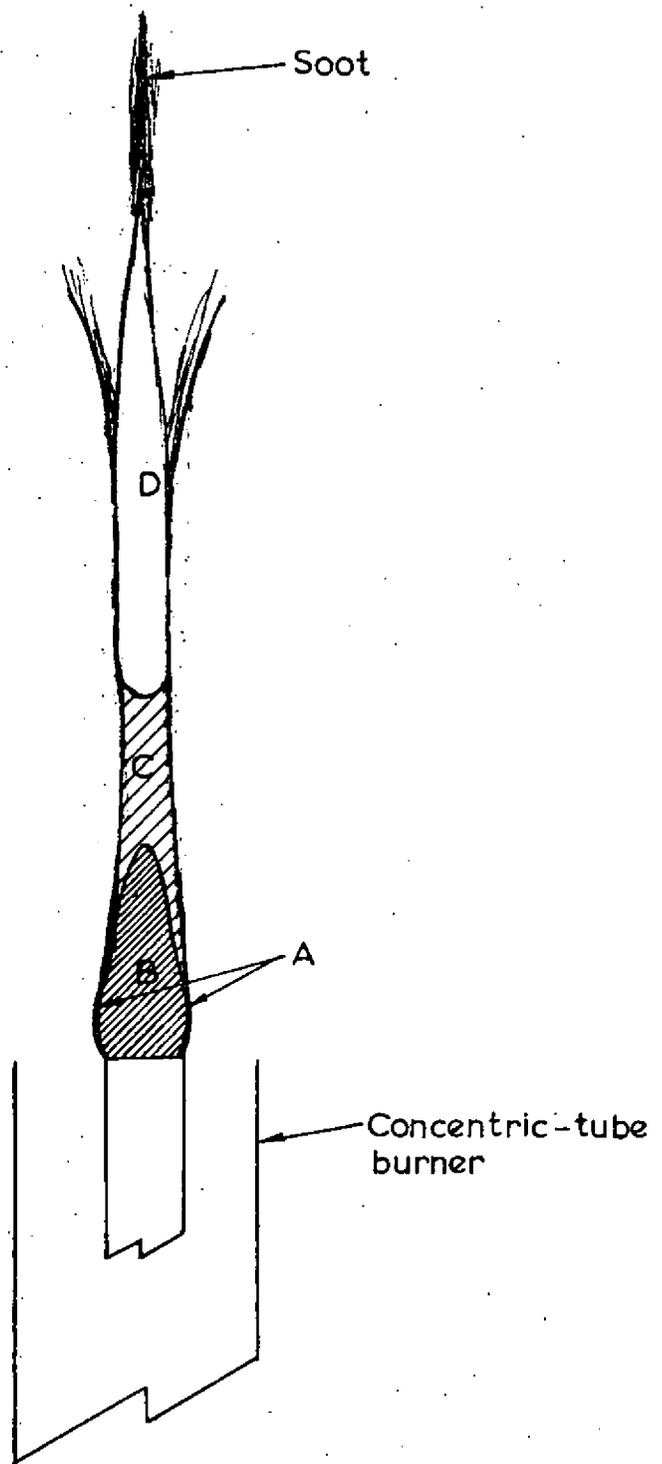
Oxygen index 0.21
Oxygen flow $210 \text{ cm}^3/\text{min}$
Ethylene flow $138 \text{ cm}^3/\text{min}$

FIG. 8. ETHYLENE DIFFUSION FLAME AT THE SMOKE POINT.
NITROGEN AS DILUENT



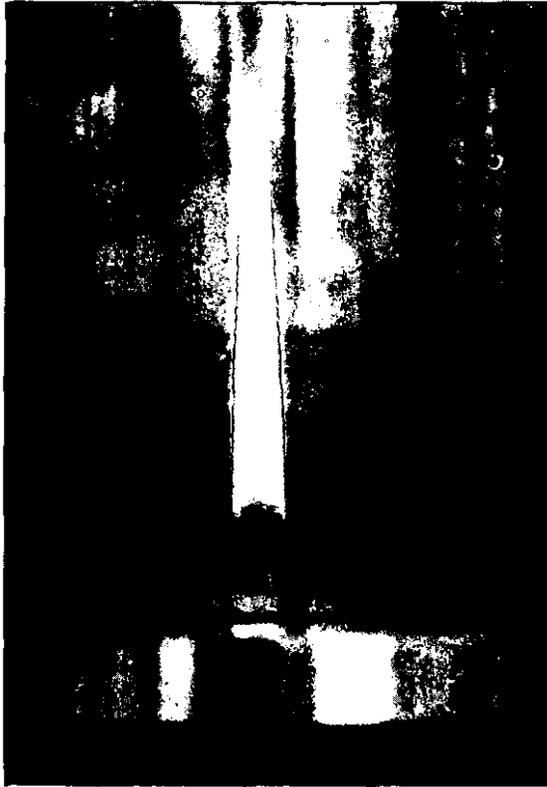
Oxygen index 0.21
Oxygen flow $210 \text{ cm}^3/\text{min}$
Ethylene flow $140 \text{ cm}^3/\text{min}$

FIG. 9. ETHYLENE DIFFUSION FLAME.
CARBON DIOXIDE AS DILUENT

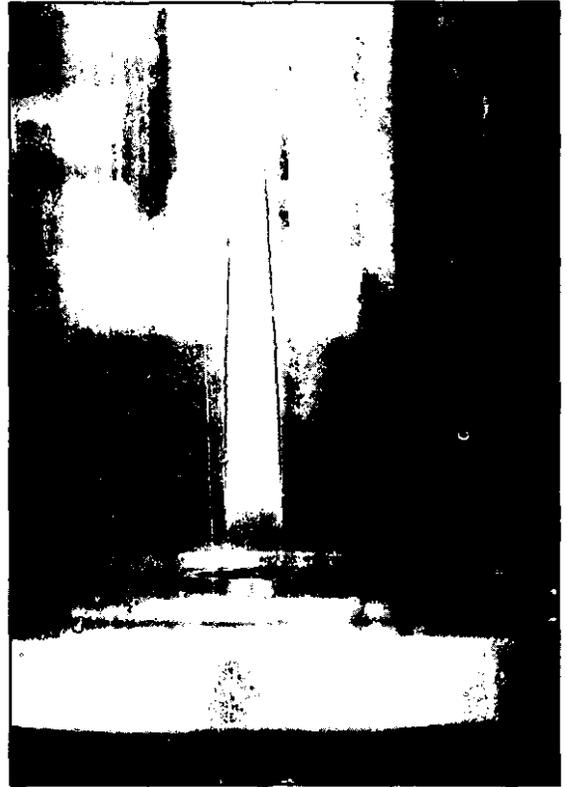


- A Blue reaction zone
- B Dark zone
- C Yellow luminous zone
- D Red luminous zone

FIG.10. A LAMINAR ETHYLENE DIFFUSION FLAME AT THE SMOKE POINT



(a) Oxygen index 0.17
Ethylene flow $144 \text{ cm}^3/\text{min}$



(b) Oxygen index 0.15
Ethylene flow $152 \text{ cm}^3/\text{min}$

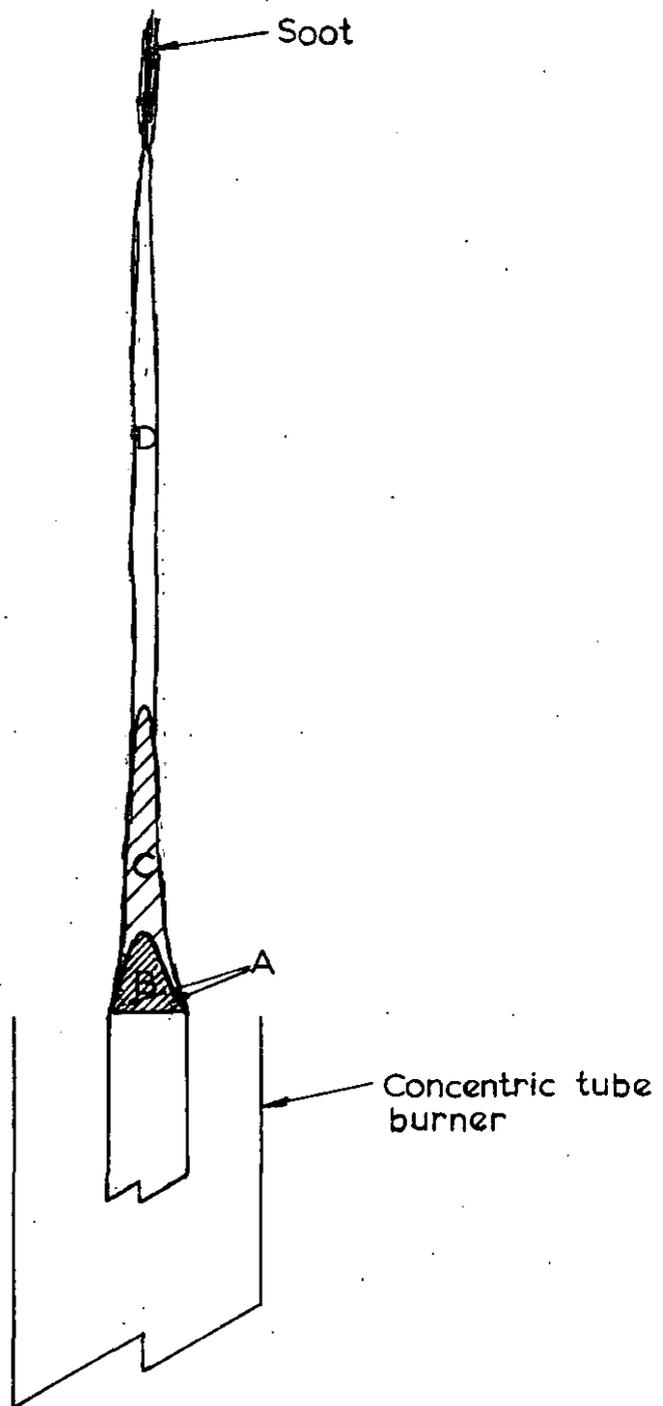


(c) Oxygen index 0.13
Ethylene flow $170 \text{ cm}^3/\text{min}$

FIG. 11. ETHYLENE DIFFUSION FLAMES AT THE SMOKE POINT.
EFFECT OF DILUTION WITH NITROGEN, AT A CONSTANT
OXYGEN FLOW OF $210 \text{ cm}^3/\text{min}$

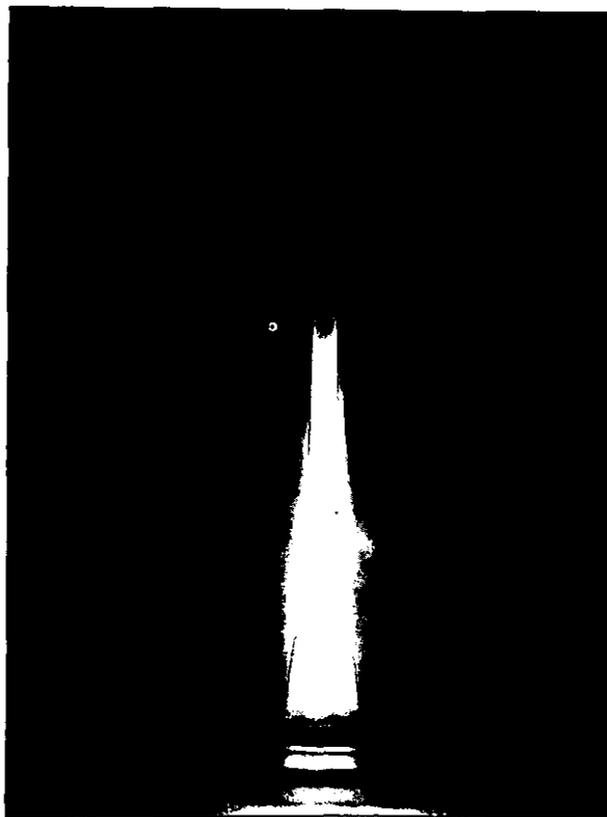


FIG. 12. LIFTED ETHYLENE DIFFUSION FLAME; NITROGEN AS DILUENT, OXYGEN INDEX 0.13



- A Blue reaction zone
- B Dark zone
- C Yellow luminous zone
- D Red or dark zone with luminous boundary

FIG.13. A LAMINAR ETHYLENE DIFFUSION FLAME AT THE SMOKE POINT
OXYGEN INDEX GREATER THAN 0.21



**FIG. 14. ETHYLENE DIFFUSION FLAME AT THE SMOKE POINT.
TYPICAL FLAME STRUCTURE FOR OXYGEN INDEX
VALUES GREATER THAN 0.21**

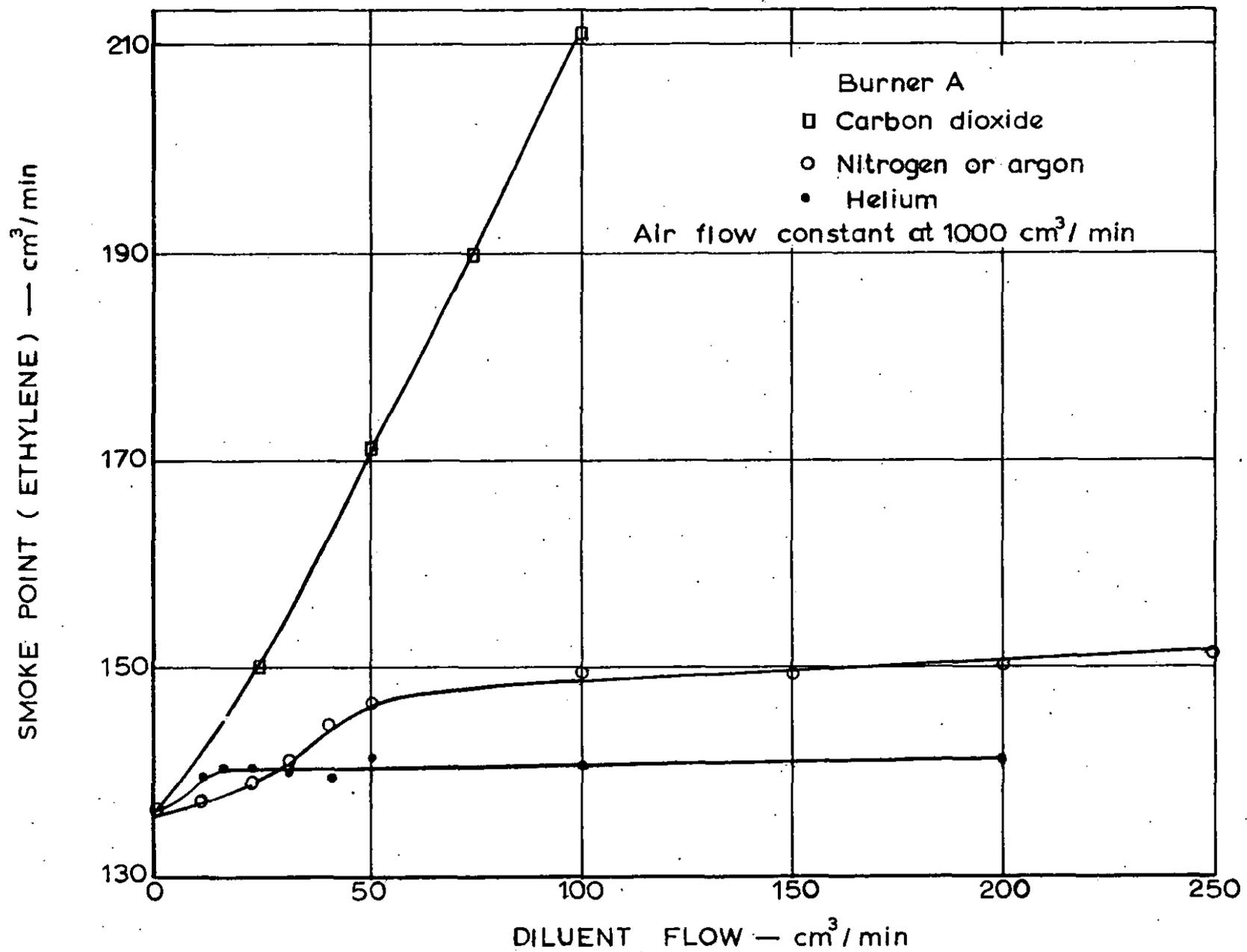
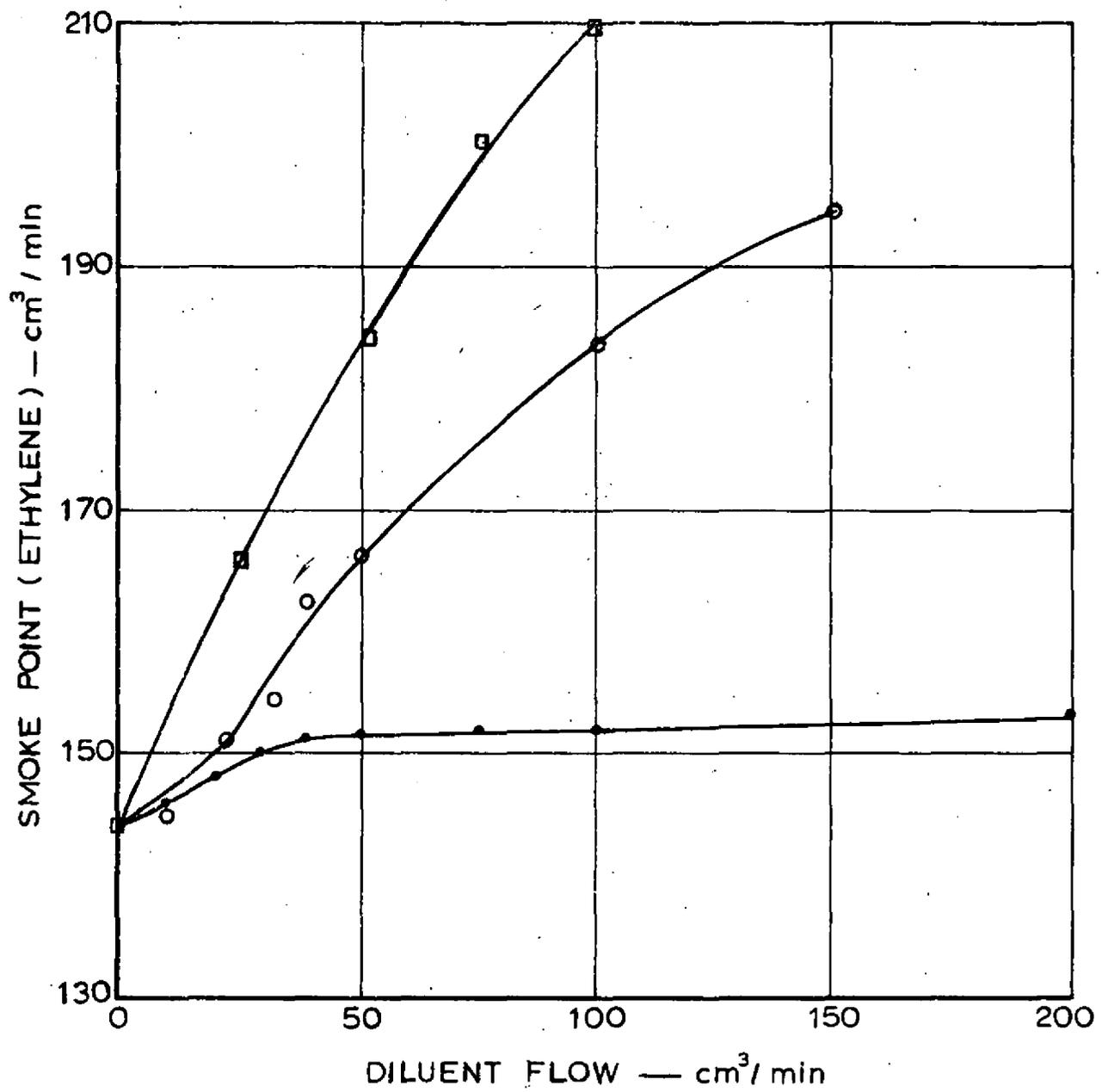


FIG.15. INFLUENCE OF DILUENT ON SMOKE POINT
DILUENT ADDED TO FUEL



Burner B
 □ Carbon dioxide
 ○ Nitrogen or argon
 • Helium
 Air flow constant at 1000 cm³/min

FIG.16. INFLUENCE OF DILUENT ON SMOKE POINT
 DILUENT ADDED TO FUEL

