

predicted a value for CO of $\Phi = 1.3$ in the same flow conditions, and this is not inconsistent with our experimental determination. Neither does the $\Phi \approx 3$ result disagree with von Rosenberg's², nor with Holbeche and Woodley's experimental results¹, within the respective combined accuracies. On the other hand, Russo's results³ of $\Phi = 100-1,000$ cannot be explained, and are the more surprising because he used an experimental temperature measuring technique identical to ours. None of these earlier investigations provided a more direct determination of Φ than ours, and they did not include an independent "calibration" of their respective experimental techniques, as we have done in our emission-absorption measurements with the CO₂/argon expansions.

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Diffuse Electrical Discharges in the Absence of Flames

KARLOVITZ^{1,2} has proposed and investigated a device in which a high voltage electrical discharge is superimposed on a flame seeded with an alkali metal salt. The distinctive feature of this device is the spreading of the discharge through the flame volume. Usually discharges tend to contract as a result of feedback, causing an increase in the temperature of the discharge, which in turn causes a further increase in conductivity. Addition of an easily ionizable salt (in this case potassium carbonate) overcomes this instability by providing a better conducting path at lower temperatures and reducing the temperature coefficient of electrical conductivity. Because the discharge is dispersed through the flame volume higher voltages are possible than with an arc column and consequently currents used are comparatively small (5-15 A). Most of the subsequent work on such systems has concerned electrical properties and heat-transfer measurements³⁻⁵.

During a study of chemical synthesis in propane-air flames augmented with an a.c. electrical discharge, we found it possible to turn off the propane supply without extinguishing the discharge, which continued in air and proved to be stable over a wide range of flow rates. Diffuse discharges were subsequently established in nitrogen, oxygen and argon by cutting off the fuel supply to an augmented propane-air flame and gradually replacing the air by the gas to be augmented. The apparatus was a simple tunnel-burner with a central rod electrode having a copper head as anode and a graphite cathode mounted externally to the burner. It was very similar to that used by Fells *et al.*⁶, but operated horizontally and on a.c. power.

The current-voltage characteristics of these discharges were similar to those for augmented flames, except in the case of argon where the voltage was somewhat smaller because of its small ionization potential (Ar 11.6 eV, O₂ 13.6 eV, N₂ 15.5 eV). Typical values are shown in Table 1. No details of similar discharges have been reported as far as we know. Montes and Cushing⁷ refer briefly to the augmentation of nitrogen-argon mixtures to 1,300 K

Table 1. TYPICAL CURRENT-VOLTAGE VALUES

	Voltage (volts)	Current (amps)
Typical flame	810	10.2
Nitrogen	880	9.9
Oxygen	700	10.8
Air	840	10.0
Argon	300	10.9

Total gas flow rate 170 l./min. Rate of seed addition 0.6 g/h.

Table 2. BURNER OPERATING CONDITIONS

	Burner dimensions (cm)
Burner tunnel diameter	2.54
Anode diameter	1.90
Distance between electrodes	15-25

Gas flow rates for stable operation 20-150 l./min.

but give no further details. T. J. Hirt (private communication) has operated a device of this type and shown that with an a.c. discharge the temperature variation at 3,200 K is less than 10 per cent. He found it necessary to preheat the gases to be augmented by using a plasma torch, and suggested that heat retention in the burner tended to smooth out the variation which might be expected from a.c. power. No preheat was used in our work and the burner would operate satisfactorily without any warming-up period. This suggests that Hirt's conclusions are not directly applicable.

The fact that discharges of this type can be established in inert gases indicates that chemi-ionization is not of great importance. Fells *et al.*⁴ tentatively suggested its significance but were inclined to favour vaporization of the copper anode and/or electron emission from the graphite cathode to account for observed conductivities. In the argon discharge the low voltage compared with air, oxygen and nitrogen at the same rate of seed addition suggests that the contribution from thermal ionization of the argon to the overall conductivity of the system is of the same order as the contribution from thermal ionization of the seed material. The discharge in argon would operate without seeding but in an unstable way. Without seed it was greenish in colour and this was attributed to vaporization of the copper anode.

It seems that the conductivity of diffuse a.c. electrical discharges in the absence of flames results from thermal ionization of the seed material and, in the case of argon, from thermal ionization of the augmented gas. The need for continuous seeding to maintain a stable discharge in an a.c. system precluded any significant conclusions about the importance of anode vaporization. That such discharges can be maintained without preheating suggests that the slight fall in temperature between cycles of alternating current found by Hirt may not be caused by heat retention in the burner but may be characteristic of the system. Conductivity remains high enough for the maintenance of a diffuse discharge in the absence of a flame. Such discharges, which can attain a temperature of several thousand degrees Kelvin while avoiding the extreme temperature gradients and high electrode losses of the arc, furnish an attractive environment for a range of chemical reactions.

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