

Stability of Burner Flames for Binary and Tertiary Mixtures of Methane, Carbon Monoxide and Water Vapour

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The flame stability of binary and tertiary mixtures of methane, carbon monoxide and water vapour has been correlated using the velocity gradient concept for laminar flow proposed by B. Lewis and G. von Elbe'. The effect of composition changes in both dry and wet methane-carbon monoxide mixtures upon flash-back and blow-off is non-linear. Increases in water vapour content, either in the primary or secondary gases, increase the velocity gradient for both blow-off and flash-back of carbon monoxide and to a lesser degree decrease the velocity gradient for methane.

IT HAS been known for some time that the burning velocity of carbon monoxide is greatly increased by the addition of small amounts of compounds containing hydrogen. For instance, L. UBBELOHDE and O. DOMMER² found that the maximum burning velocity of carbon monoxide-air mixtures rises from 16 cm/sec for dry carbon monoxide to about 55 cm/sec for carbon monoxide with 9.4 per cent water. Beyond this concentration water no longer acts as a hydrogen donor but as a diluent. E. F. FLOCK and H. K. KING³ determined the effect of water vapour on the burning velocity of carbon monoxide-oxygen mixtures at various initial gas pressures. They found that the percentage of water vapour to give the peak carbon monoxide-oxygen burning velocity varied with pressure. For instance, at 100 mm pressure the burning velocity reaches a peak at 16 per cent water, whereas, at 760 mm pressure the peak is at 8.0 per cent water vapour. W. A. BONE⁴ found that for a mixture of 45 per cent carbon monoxide in air, the burning velocity determined with a 2.5 cm tube varied from 56 to 118 cm/sec as the percentage of moisture was changed from 0.70 to 8.0 per cent. K. BUNTE⁵⁻⁹ and his associates found that the burning velocity of carbon monoxide is accelerated not only by water vapour but also by hydrogen, as well as by organic compounds containing hydrogen. This is interpreted by Bunte as a sign that OH radicals as well as H atoms participate in effecting the burning velocity of the carbon monoxide combustion. K. BUNTE and E. HARTMANN¹⁰ found that 1 per cent hexane raises the burning velocity of carbon monoxide 29.5 per cent even though hexane had a lower burning velocity than the carbon monoxide used.

Despite the considerable amount of work that has been done on the effect of hydrogen-bearing compounds on the burning velocity of carbon monoxide, little information is available regarding the flame stability curves of carbon monoxide in combination with hydrogen-bearing compounds. The present paper endeavours to alleviate this deficiency by presenting data on the flame stability of binary and tertiary mixtures of methane, carbon monoxide and water vapour.

Blow-off and flash-back were determined by the conventional methods using the equipment and procedure previously described¹¹. Air cooled Pyrex No. 744 burner tubes of 1.273, 1.093, 0.930, 0.770 and 0.573 cm internal diameter were employed. Mass spectrometer analyses, on the dry basis for the 'pure grade' natural methane and the 'pure grade' cylinder carbon monoxide gases used in this investigation, are shown in Table 1.

Table 1 Analyses of gases

Natural methane		Cylinder carbon monoxide	
Constituent	%	Constituent	%
Methane	96.77	Carbon monoxide	98.50
Ethane	1.88	Hydrogen	1.01
Propane	0.11	Carbon dioxide	0.49
Oxygen	0.10		
Nitrogen	1.80		
Carbon dioxide	0.04		

For the tests with no moisture added to the gas streams, the air, methane and carbon monoxide were dried by passing them separately through drying tubes packed with granular calcium chloride, followed by activated alumina. For the tests with moisture added, the carbon monoxide and methane streams were saturated by passing the gases through standard saturators immersed in a constant temperature bath, and the primary air stream by passing the air through two saturator towers in series. The fuel gas saturation was found to be essentially constant over the range of gas flow

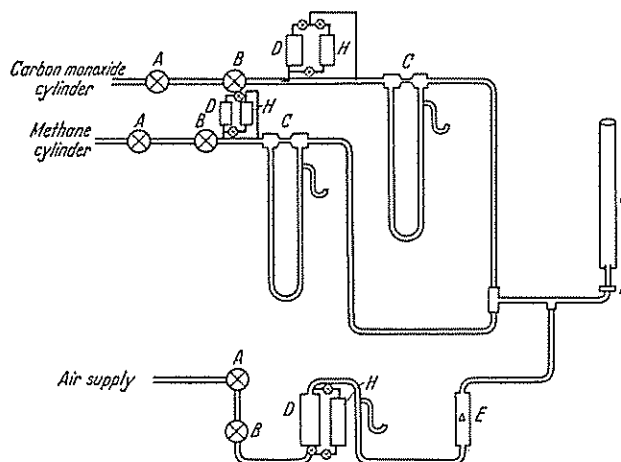


Figure 1 Schematic diagram of apparatus: A high pressure reducing valves B low pressure reducing valves C capillary flow meters D calcium chloride column E rotameter F flame check G burner tube H water saturators

rate used, at a value of 3.5 mol per cent water vapour (0.0225 lb H₂O/lb CO, 0.0394 lb H₂O/lb CH₄) and the saturation of the primary air stream was essentially constant at a value of 3.0 mol per cent water vapour (0.0186 lb H₂O/lb bone dry air). All runs were made at room temperature

which varied a maximum of 15°F from 71°F to 86°F. Figure 1 represents a schematic diagram of the apparatus.

Velocity gradients were calculated from the flow data using the relation $g=4V/\pi R^3$, where g is velocity gradient at the burner wall (sec^{-1}), V is the total flow in the burner tube (cc/sec), and R is the burner tube radius (cm).

DISCUSSION OF RESULTS

Flash-back and blow-off results for dry natural methane are shown in Figure 2a; this represents a graph of the velocity gradients at which flash-back and blow-off occur for different percentages of methane in methane-air mixtures. The percentage of primary air in the mixtures is indicated by arrows shown at the top of the figure. In all the flame stability graphs the velocity gradient scale covers the range from 100 to 10,000 and the abscissa is plotted in per cent of dry fuel gas. It will be noted that the flash-back peak comes at just under 100 per cent primary air. The

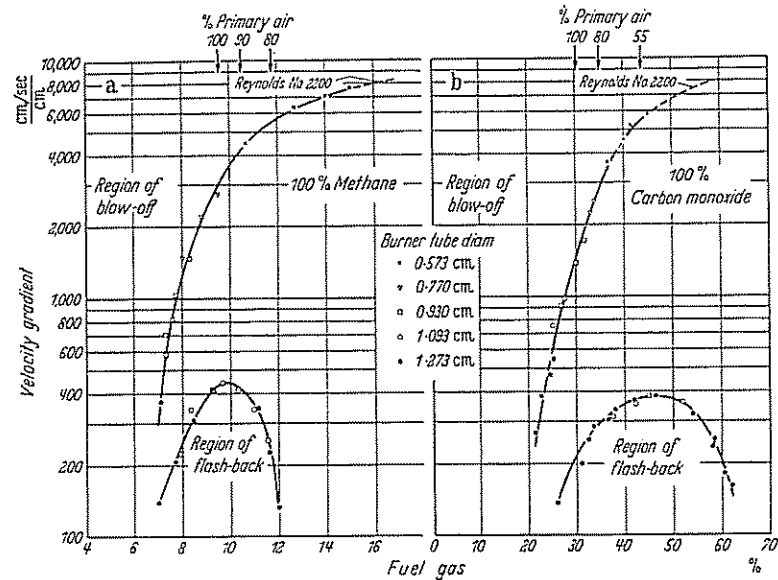


Figure 2. Critical velocity gradients at boundary of gas stream for flash-back and blow-off from cylindrical air cooled Pyrex burner tubes of different diameters (secondary air humidity 0.006 lb water/lb bone dry air): a primary air and methane dry b primary air and carbon monoxide dry

theoretical transition point from viscous to turbulent flow is indicated. The regions of blow-off and flash-back are marked. Outside of these areas the range of stable flame exists.

Similar data for dry carbon monoxide are shown in Figure 2b. A small variation in the amount of hydrogen in the carbon monoxide has an appreciable effect on both blow-off and flash-back. For instance, the cylinder of carbon monoxide used in subsequent studies on carbon monoxide-propane mixtures contained 1.80 per cent hydrogen and showed

a flash-back peak of 590 as compared to 380 in the present case with carbon monoxide containing 1.01 per cent hydrogen. It will be noted that the flash-back peak is shifted to a much richer gas mixture, 46 per cent, in comparison to 10 per cent for methane. This is equivalent to 45 per cent of the primary air theoretically required for complete combustion of carbon monoxide. It is also noted that the humidity of the secondary air is recorded since, as will be seen later, this has considerable effect on carbon monoxide flame stability.

In *Figure 3* flash-back data for dry methane-carbon monoxide mixtures are plotted in terms of velocity gradient *versus* per cent fuel gas in the fuel gas-air mixture. Experimental points are not shown on these summary curves, but in all cases the reproducibility of experimental data was good. It is readily apparent that increasing the methane content of the carbon monoxide-methane mixtures produces a marked shift in the flash-back peak and that the effect is non-linear. It is also apparent that a small addition of methane increases the velocity gradient at the flash-back peak considerably with the peak velocity gradient coming at about 7.5 per cent methane. Beyond 7.5 per cent methane the flash-back peaks decrease progressively to the value for natural methane.

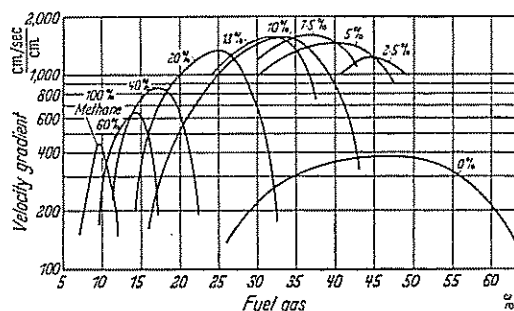


Figure 3 Critical velocity gradients at boundary of gas stream for flash-back at various mixture compositions of methane and carbon monoxide; primary air and fuel gas dry, secondary air humidity 0.005 lb water/lb bone dry air

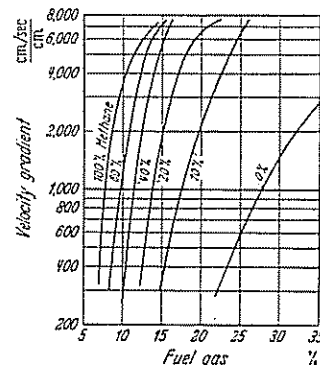


Figure 4 Critical velocity gradients at boundary of gas stream for blow-off at various mixture compositions of methane and carbon monoxide; primary air and fuel gas dry, secondary air humidity 0.005 lb water/lb bone dry air

In *Figure 4* blow-off data for dry methane-carbon monoxide mixtures are presented over the complete range of compositions. As for flash-back, it is apparent that the effect of methane additions upon blow-off is non-linear, since the first 10 per cent methane shift the curves as much as do the last 90 per cent. Also as the percentage of methane increases, the slope of the blow-off curves increases. It will also be noted that as the velocity gradient increases the slopes of the curves decrease.

In *Figure 5a* the effect of water vapour on methane flame stability is noted. Curves A represent the stability for dry methane and dry primary air

with a secondary air humidity of 0.006 lb water/lb air (H in the legend refers to secondary air humidity). Curves B represent dry gases again, however, the secondary air humidity has been increased to 0.015. No detectable change in either blow-off or flash-back curves was noted. For the last two curves, the secondary air moisture remained at 0.015. Saturating the methane with 3.5 mol per cent water, as shown in curves C has no noticeable effect on blow-off, but decreases the flash-back curve slightly. In curves D , both the methane and the primary air were saturated. The additional 3 mol per cent water vapour added by the primary air stream, about 25 per cent water on the basis of methane, quench the reaction considerably and produce a much lower blow-off and flash-back curve. Curves A and D represent the limits of flame stability that should be encountered in practice for methane.

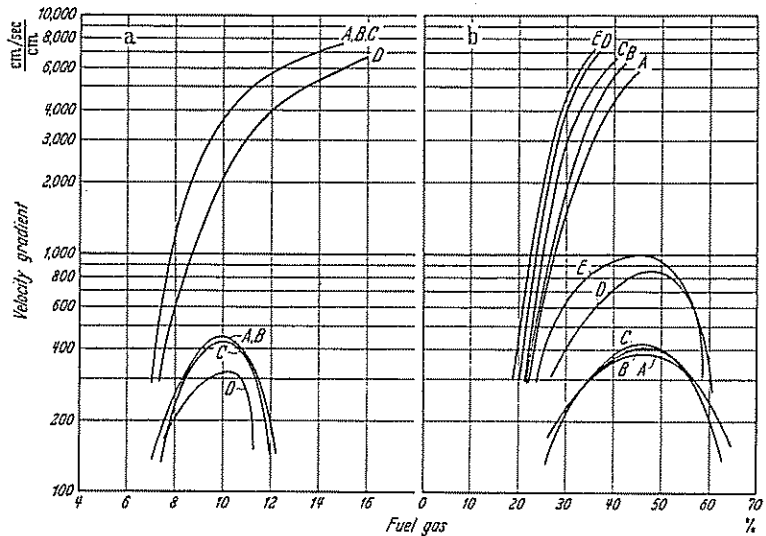


Figure 5 Effect of water vapour on flame stability of a methane (A dry gases H 0.006; B dry gases H 0.015; C saturated methane H 0.015; D saturated gases H 0.015) b carbon monoxide (A dry gases H 0.005; B dry gases H 0.010; C dry gases H 0.0165; D saturated carbon monoxide H 0.0165; E saturated gases H 0.0165)

In Figure 5b the effect of water vapour on carbon monoxide flame stability is noted. Moisture has just the opposite effect on carbon monoxide that it has on methane. Water acts as a reaction accelerator for carbon monoxide, as can be seen from the stability curves. Curves A , B and C are for dry carbon monoxide and dry primary air with variation in secondary air humidity of 0.005, 0.010 and 0.016 lb of water per lb of air respectively. It will be seen that increases in the secondary air humidity increase the blow-off appreciably and the flash-back only slightly. This is to be expected since at blow-off the dead space distance between the burner rim and the flame is much greater than at flash-back. Hence, more secondary air can enter the flame. In curves D , the carbon monoxide

was saturated with water vapour and an appreciable increase in blow-off and flash-back is noted. The increase in velocity gradient peak for flash-back from 420 to 850 strikingly illustrates the marked effect of water vapour. In curves E both the carbon monoxide and primary air are saturated and again the flame stability curves show an increase; however, it is seen here that the effect of water vapour is non-linear, being much greater at low concentrations. This is in direct comparison to the results for methane where water vapour has almost a linear effect on flame stability.

In Figure 6 flash-back data for mixtures of dry methane, saturated carbon monoxide and saturated primary air are plotted. Even though the methane was not saturated, it is present in such a small percentage in the fuel gas

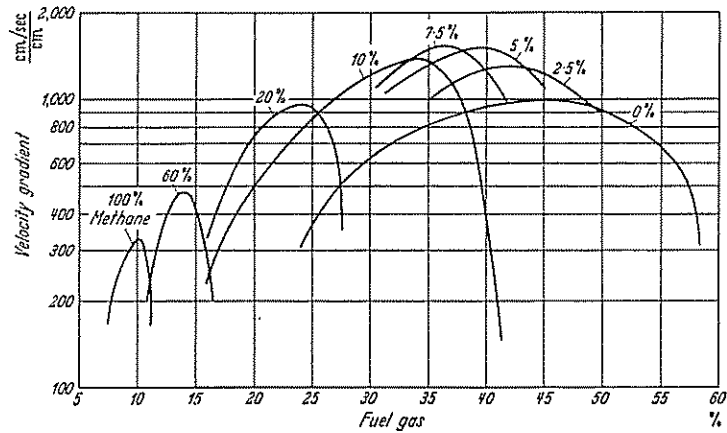


Figure 6 Critical velocity gradients at boundary of gas stream for flash-back at various mixture compositions of methane and carbon monoxide; primary air and carbon monoxide saturated, methane dry, secondary air humidity 0.0165 lb water/lb bone dry air

mixtures that the mixtures can be considered essentially saturated. Comparison of Figure 6 with Figure 3 shows the very large effect a small quantity of water vapour has on flame stability.

In Figure 7 a composite picture of the flash-back peaks for both dry and saturated methane-carbon monoxide mixtures is presented. For the dry mixtures a peak velocity gradient of 1,600 is reached at 7.5 per cent methane. When the carbon monoxide and primary air are saturated, the peak velocity gradient is 1,525 at about 6.5 per cent methane. Up to 6 per cent methane, the velocity gradient of the saturated gas is greater than the dry gas; beyond this point the saturated mixture peaks are lower than the dry gases. It is also interesting to note that for the dry gas streams, the first 5.0 per cent methane increase the velocity gradient 1,080 units, 216 units for each per cent methane, whereas for the saturated gas streams the first 5.0 per cent methane increase the velocity gradient 525 units or 105 units for each per cent methane. This fact gives an idea of the decreased effectiveness of methane added to saturated carbon monoxide.

There is also a shifting of the shape of the maximum portion of the flash-back peak. The lower methane side of the peak is raised so that there is a negligible difference between the 5.0 and 7.5 per cent points. Also in *Figure 7* are shown dashed straight lines connecting pure carbon monoxide and methane for both the dry and saturated mixtures. These lines represent the velocity gradients for mixtures that would be predicted on a molecular basis. For the dry mixtures it is seen that if the peak velocity gradients of any carbon monoxide-methane mixture were estimated on a molecular basis from the maximum velocity gradients of the initial gases, the predicted value would be considerably below the correct value, reaching a maximum difference for a 7.5 per cent methane mixture of over 300 per cent. In the case of the saturated mixture, the velocity gradient predicted on a molecular basis deviates from the experimental results by a maximum of 60 per cent too low at 7.5 per cent methane and 18 per cent too high at 55 per cent methane.

Figure 8 shows the blow-off curves of saturated gas mixtures. The spread between the 0 and 100 per cent methane curves is much less in

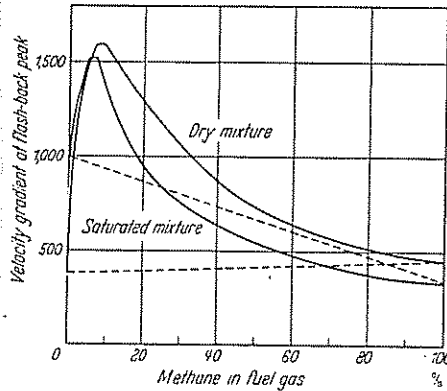


Figure 7 Comparison of flash-back peaks of dry and saturated methane-carbon monoxide mixtures

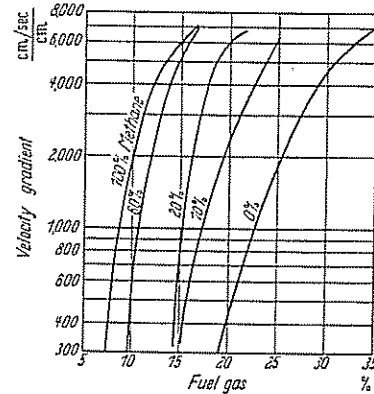


Figure 8 Critical velocity gradient at boundary of gas stream for blow-off with various mixture compositions of methane and carbon monoxide; primary air and carbon monoxide saturated, methane dry, secondary air humidity 0.0165 lb water/lb bone dry air

this case than for the dry gases. This is due to the marked shift at the lower methane concentrations towards blow-off at leaner gas mixtures when water vapour is present and a significant but less pronounced shift at the higher methane concentrations toward blow-off at richer gas mixtures. The results may be attributed to the fact that up to about 10 per cent methane, carbon monoxide still lacks sufficient hydrogen to completely accelerate the combustion reaction. Beyond this point the addition of hydrogen is of little use and quenches the reaction, if the hydrogen be supplied by a slower burning fuel or an inert gas.

CONCLUSIONS

The effect of composition changes in dry methane-carbon monoxide mixtures upon flash-back and blow-off is non-linear.

The peak velocity gradient for flash-back occurs at approximately 7.5 per cent methane in the dry methane-carbon monoxide mixture for gases of the composition studied.

Increasing secondary air moisture increases the velocity gradient for blow-off and flash-back of carbon monoxide.

Up to a concentration of 0.015 lb water/lb bone dry air, increasing secondary air moisture has no measurable effect on the velocity gradient for blow-off or flash-back of methane.

For carbon monoxide flames, changing secondary air moisture affects blow-off more than flash-back.

Water vapour added to the carbon monoxide and primary air streams increases the velocity gradient for blow-off and flash-back of carbon monoxide considerably.

Water vapour added to the methane and primary air streams decreases the velocity gradient for blow-off and flash-back of methane noticeably, but the change is not as great as for carbon monoxide.

Saturation of the carbon monoxide and primary air streams increases the flash-back velocity gradient up to 6 per cent methane in methane-carbon monoxide mixtures; beyond this point the velocity gradient is less than for dry gas mixtures.

For hydrocarbon-carbon monoxide mixtures flame stability curves cannot be predicted from the pure gases on the common mol per cent basis.

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