THE FLAMMABILITY LIMITS OF GASEOUS MIXTURES IN POROUS MEDIA

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Introduction

In recent years, there has been considerable interest in gas combustion in porous media. An extensive review of the subject is given by Howell *et al.* [1]. Combustion in porous media is relevant to the design of porous burners for efficient heat transfer and for extending the flammability limits of gaseous hydrocarbon mixtures in a heated porous bed. Another important application of flame propagation in porous media is the use of packed beds as flame-arresting devices. Various applications of combustion in porous media have been discussed by Echigo [2] and Hanamura *et al.* [3].

Although the flammability limits of gaseous hydrocarbon mixtures have been well documented, there are few data available on flame propagation and limits of combustible mixtures in porous media. The basic mechanisms of flame propagation and extinction in a porous medium are not well established although narrower flammability limits are expected due to significant heat losses. Thus, the objective of the present study is to obtain experimental data to assess the effect of heat losses on the flammability limits of gaseous mixtures in a porous medium.

Experimental Details

The experiments are carried out in a 1.8-m long transparent Plexiglas tube of 5-cm internal diameter. The tube was constructed to match the standard flammability apparatus of Coward and Jones to permit a direct comparison of data [4]. A schematic of the apparatus is illustrated in Fig. 1. The combustible mixture is



Fig. 1 Schematic of apparatus

prepared by a continuous flow method where the concentrations of fuel and oxidizer (air) are monitored using calibrated flow meters. The fuel and air streams are mixed in a chamber with turbulence-generating baffles to enhance mixing prior to entering the flame tube. The combustible mixture is flowed through the flame tube for sufficient time to allow the displacement of ten tube volumes, ensuring a homogeneous mixture in the tube. Prior to ignition, the ignition end of the tube is opened to the atmosphere while the downstream end remains closed. Thus, in all experiments the flame propagates from the open end to the closed end of the tube. The mixture is ignited with an electrically heated 0.25-mm gauge tungsten wire. The wire is located at the top of the tube for downward flame propagation and at the bottom for upward flame propagation. Methane-air and propane-air mixtures are used in the present study.

The porous media is composed of a packed bed of solid spheres supported on a wire screen located 0.9 m from the end of the tube. A range of materials with varying thermal conductivities are tested (i.e. glass, stainless steel, and brass). The sphere diameters vary between 9.53 mm and 12.7 mm. Preliminary tests indicate that the flammability limits are independent of the height of the packed bed section for column heights greater than approximately one tube diameter (i.e. 5 cm). Thus, all

experiments are conducted with a one-tube diameter height of porous media. Failure or successful transmission of the flame through the porous media is determined by visual inspection, following the standard procedure used in flammability tests.

Results and Discussion

To verify the accuracy of the present results, experiments were first carried out to determine the flammability limits of CH_4 -air and C_3H_8 -air mixtures in an open tube (i.e. without the porous media section). The criterion used for determining limits is based on whether the flame quenches before it propagates through the entire length of the tube. In some cases, however, the limits were determined by failure to ignite the mixture itself. The results of the open tube experiments are summarized in Table 1. The limits obtained are in good agreement with published data. For example, the upward lean flammability limit for CH_4 -air is found to be 5.35%, in accord with the value of 5.3% given by Coward and Jones [4]. A downward limit of 5.98% for CH_4 -air is obtained which agrees well with the value of 5.85% reported by Strehlow and Jarosinski [5]. In addition, the results show clearly the effect of buoyancy on the flame propagation: the flammability limits for upward flame propagation are wider than those for downward flames. All the results obtained are highly reproducible.

In determining the flammability limits with porous media, the combustible mixture is ignited upstream of the column of solid spheres. Thus, the limit obtained corresponds to the critical mixture in which flame transmission through the porous medium is no longer possible. This choice of methodology is justified since upstream ignition ensures that a fully self-sustained laminar flame is first established prior to encountering the packed bed section. The range of sphere diameters tested varies from 9.53 mm to 12.7 mm. This rather narrow range of sphere sizes is due to the fact that a flame propagating in a stoichiometric mixture of CH_4 -air or C_3H_8 -air is quenched by a packed bed of 7.94 mm spheres thus setting the lower limit of sphere sizes. For the upper limit, spheres larger than 12.7 mm become comparable to the tube diameter (50 mm). Using such large spheres presents a difficulty in obtaining a packed bed in a 5-cm diameter tube. For sphere sizes below 12.7 mm, experiments show that the randomness in the packing of the spheres does not influence the flammability limits.

In general, upon ignition of the combustible mixture, a blue laminar flame propagates at relatively constant speed upstream of the porous medium. Periodic bright flashes are observed behind the flame front in the more sensitive mixtures near stoichiometric composition. Cellular flame structures are also observed in lean CH_4 -air mixtures and in rich C_3H_8 -air mixtures in accordance with laminar flame instability theory. Flames propagating in more sensitive mixtures are seen to generate acoustic vibrations indicating that the frequency of flame oscillations is close to the basic acoustic modes of the tube. Propagation and quenching of the flame in the porous medium can be observed when the packed bed is transparent (i.e. glass spheres). In all tests in which the flame successfully transmits through the porous medium, the flame undergoes rapid acceleration as it emerges from the porous medium. This is due to the increase in flow resistance provided by the porous bed to the downstream expansion of the products.

Typical data for the flammability limits of upward flame propagation in CH_4 -air and C_3H_8 -air mixtures in porous media are summarized in Table 2. Table 3 lists the results obtained with downward propagating flames. In all experiments, the results are very reproducible. Intuitively, it is expected that a material with high thermal conductivity will have narrower limits. However, the experimental results indicate that the flammability limits are relatively insensitive to the material of the solid spheres (Fig. 2). For example, the downward lean flammability limit for CH_4 -air in a packed bed of 9.53-mm glass spheres is found to be 7.86% compared to the limit of 7.66% obtained with 9.53-mm brass spheres. The downward rich limits for CH_4 -air in the same packed beds of glass and brass spheres are 10.87% and 10.96%, respectively. Glass is significantly less conductive than brass but its limits are found to be narrower. Figure 2 also indicates that buoyancy forces do not appear to affect the flammability limits of gaseous mixtures in the porous media. Unlike the results in the open tube, the downward limits are not narrower than the upward limits indicating that the buoyancy effect is insignificant in a porous bed. In addition, from Table 2, the upward lean flammability limit for CH_4 -air in a packed bed of 12.7mm stainless steel spheres is found to be 6.30% compared to the limit of 6.52% obtained with 12.7-mm glass spheres. The limits for C_3H_8 -air in the same packed beds of steel and glass spheres are 3.11% and 2.96%, respectively. Thus, the results are inconsistent for different fuels.

The present experimental results indicate that heat loss to the porous media does not appear to be a dominant flame quenching mechanism. This result is in accordance with flame quenching in small tubes where the tube wall material has been found to play a small role in the flame quenching distance [6]. The present results suggest that a more probable flame quenching mechanism is due to flame stretching and the high degree of flame curvature as the flame negotiates its way through the connecting channels between the closely packed

spheres of the bed. The average channel dimension in a close-packed bed of spherical particles is $0.3d_s$, where d_s is the sphere size [7]. For the 7.94-mm diameter spheres, the average dimension of the channels in the packed bed is about 2.4 mm, close to the quenching distance of stoichiometric hydrocarbon-air flames (on the order of 1 mm). Previously published data for quenching distance as a function of CH₄ concentration in air are shown in Fig. 3 [8]. By estimating the average channel dimension between the closely packed spheres as a characteristic quenching distance, the experimental results obtained are in fairly good agreement with the published data. Thus, the limits in a porous bed are better correlated to the quenching distance of the mixture rather than to the global mechanism of heat loss to the porous bed.

To complement the experimental study, a numerical simulation of flame propagation in a porous medium has also been carried out. The model assumes a one-dimensional tubular reactor where a planar reaction front propagates into a porous medium with increased heat and momentum transport. On a qualitative basis, the numerical results are in accordance with experimental results even though the three-dimensional effects of flame propagation through the channels of the porous bed (considered to be the dominating mechanism in the quenching process) are not included in the theoretical model. Further refinement of the numerical simulation based on more experimental data is necessary to arrive at a proper theoretical description of the phenomenon.

References

- 1. Howell, J.R., Hall, M.J., and Ellzey, J.L. Progress in Energy and Combustion Science, 1996, pp. 22-121.
- 2. Echigo, R. Radiation Enhanced/Controlled Phenomena of Heat and Mass Transfer in Porous Media. ASME/JSME Thermal Engineering Joint Conference, 1991, Vol. 4, pp. xxi-xxxii.
- 3. Hanamura, K., Echigo, R., and Zhdanok, S.A. Super-Adiabatic Combustion in Porous Media. *International Journal of Heat and Mass Transfer*, 1993, Vol. 36, pp. 3201-3209.
- 4. Coward, H.F., and Jones, G.W. Limits of Flammability of Gases and Vapors. U.S. Bureau of Mines Bulletin, 1952, No. 503.
- 5. Jarosinski, J. and Strehlow, R.A. Lean Limit Flammability Study of Methane-Air Mixtures. *Archivum Combustionis*, 1981, Vol. 1, No. 3-4, pp. 203-215.
- 6. Holm, J.M. Phil. Mag., 1932, Vol. 14, pp. 18-56.
- 7. Lee, J.J. Detonation Mechanisms in a Condensed-Phase Porous Explosive. PhD thesis, University of Sherbrooke, Sherbrooke, Quebec, Canada, 1997.
- 8. Blanc, M.V., Guest, P.G., von Elbe, G., and Lewis, B. Minimum Ignition Energies and Quenching Distances of Mixtures of Hydrocarbons and Ether with Oxygen and Inert Gases. *Third Symposium on Combustion Flame and Explosion Phenomena*, 1949, pp. 363-367.

Table 1: Summary of lean (ϕ <1) and rich (ϕ >1) flammability limits for CH₄-air and C₃H₈-air mixtures in an open tube

Fuel	Direction of	Published	Results [%]	Experimental Results [%]		
	Flame Propagation	(\$<1)	(\$>1)	(\$<1)	(\$>1)	
CH_4	upward	5.3	15.0	5.35	15.03	
CH_4	downward	5.85	N/A	5.98	13.84	
C_3H_8	upward	2.2	9.5	2.25	9.47	
C_3H_8	downward	N/A	N/A	2.23	6.85	

Table 2: Summary of lean (ϕ <1) and rich (ϕ >1) flammability limits for upward flame propagation in CH₄-air and C₃H₈-air mixtures in porous media

	Flammability Limits [%]								
Fuel	9.53-mm spheres			11.11-mm spheres			12.70-mm spheres		
	Glass	Steel	Brass	Glass	Steel	Brass	Glass	Steel	Brass
CH ₄ (\$<1)	7.93	7.38	7.03		6.89		6.52	6.36	
CH ₄ (\$>1)	10.51	10.42	10.23		11.32		11.14	11.58	
C ₃ H ₈ (\$<1)	3.45	3.53	3.50				2.96	3.11	
C ₃ H ₈ (\$>1)	6.04	5.97	6.45				6.79	7.12	

Table 3: Summary of lean (ϕ <1) and rich (ϕ >1) flammability limits for downward flame propagation in CH₄-air and C₃H₈-air mixtures in porous media

	Flammability Limits [%]								
Fuel	9.53-mm spheres			11.11-mm spheres			12.70-mm spheres		
	Glass	Steel	Brass	Glass	Steel	Brass	Glass	Steel	Brass
CH ₄ (\$<1)	7.86	7.45	7.66	7.38	7.17		6.59	6.59	
CH ₄ (\$>1)	10.87	11.06	10.96	11.14	11.14		11.93	12.02	
C ₃ H ₈ (♦<1)	3.37	3.22	3.28	3.31	3.22		2.91	2.84	
C ₃ H ₈ (\$>1)	6.18	6.11	6.18	6.24	6.45		6.52	6.45	



Fig. 2 Lean (ϕ <1) and rich (ϕ >1) flammability limits for CH₄-air mixtures in packed bed sections of 9.53 mm diameter spheres. Three sphere materials are tested: brass, steel, and glass.



Fig. 3 Characteristic quenching distance (approximated as $0.3d_s$) as a function of CH₄ concentration in air. The triangles represent data obtained with packed beds of steel spheres.