CALCULATION OF UPPER FLAMMABILITY LIMITS OF METHANE-AIR MIXTURES AT ELEVATED PRESSURES AND TEMPERATURES

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INTRODUCTION

Knowledge of the flammability limits of gaseous mixtures is important for the safe and economic operation of many industrial processes. There are several standardised experimental methods available today to determine these limits [1-4]. These experiments are, however, cumbersome and time-consuming, especially at the elevated conditions of temperature and pressure at which many industrial processes are operated. Therefore, other methods are being sought with which safe and accurate limits can be calculated. To guarantee a safe operation the calculated limits should at the least be wider than the experimentally determined ones. They cannot, however, be too wide, since this would lead to a non economic operation. The aim of this study is to evaluate different approaches for the calculation of the limits of flammability of methane-air at initial pressures up to 10 bar and temperatures up to 200°C. One possible method is to numerically calculate premixed flames, with the inclusion of a heat loss term in the energy conservation equation [5-7]. Other possibilities include the application of a limiting flame temperature [8] or a limiting burning velocity [9] below which flames are unable to propagate.

NUMERICAL CALCULATIONS

The numerical calculations are performed using CHEM1D [10], a one-dimensional (1D) flame code capable of solving 1D mass, energy and species conservation equations with detailed transport and chemical kinetics models. Two different flame geometries are used, namely 1D planar premixed flames and quasi 1D spherically expanding flames. Radiative heat loss is modelled by means of the optically thin limit. The GRI 3.0 mechanism [11] was used, however, without the reactions describing the nitrogen chemistry. The resulting mechanism contains 217 elementary reactions amongst 35 species.

The flammability limits for the planar flames are determined by considering whether an unsteady flame calculation reaches steady state or not.

In the spherical flame calculations, flames which propagate over the entire computational domain, i.e. over a distance of 100 mm, are considered to be flammable. The ignition is modelled as an energy input of 2.1 J during a period of 10 ms in a spherical volume with a radius of 5 mm.

RESULTS AND DISCUSSION

In Figure 1 a comparison is made between the numerically calculated upper flammability limits and a set of experimentally determined ones [12]. The experiments were performed in a closed spherical vessel with an internal volume of 8 l. The mixtures were ignited by fusing a tungsten wire, placed at the centre of the vessel, by electric current. This gave an energy input of 10 J during a period of 40 ms. As can be seen in Figure 1, both the numerical calculations for planar flames and the experiments yield a straight line relationship between the upper flammability limit and initial temperature. The numerical limits are, however, far wider than the experimental ones. This disparity increases with increasing initial pressure, thereby rendering the numerical results useless for practical purposes at elevated pressures. The limited number of results obtained so far for the spherically expanding flames show a slight improvement. This was expected, since the flame stretch to which these flames are subjected, makes them less reactive, i.e. it lowers their burning velocity.

Apart from values for the flammability limit, the numerical calculations also give values for the burning velocity and the maximum flame temperature. Table 1 gives an overview of these values calculated at the experimentally determined upper flammability limit. Often cited in literature is the modified law of Burgess and Wheeler [13], which describes a linear relationship between the flammability limits and initial temperature. This law is based upon a constancy of the adiabatic flame temperature at
They found the following dependence:

\[ S_{u,\text{lim}} \propto \left( \frac{g \alpha^2}{d} \right)^{1/4}, \]  

(1)

in which \( g \) is the gravitational acceleration, \( \alpha \) is the thermal diffusivity and \( d \) is the tube diameter. From equation 1 it can be calculated that:

\[ S_{u,\text{lim}} \propto p^{-1/2}, \]  

(2)

in which \( p \) is the pressure.

This correlation is checked in Figure 2 which gives the numerically calculated burning velocities as a function of pressure. Through the data points a relationship of the form \( S_{u,\text{lim}} = a \cdot p^b \) line is fitted with the least squares method. The exponent \( b \) is found to be -0.45, -0.47 and -0.51, respectively at 25°C, 100°C and 200°C. The limiting burning velocity approach, thus, seems the most promising for calculating the flammability limits at elevated conditions of pressure and temperature.

CONCLUSIONS

At atmospheric pressure the numerical calculation of flammability limits by inserting a radiative heat loss term into the energy conservation equation is satisfactory. The same holds for the application of the modified law of Burgess and Wheeler. At elevated pressures, however, large deviations occur. The numerical calculations yield values which are too conservative, while the modified law of Burgess and Wheeler (or the application of a constant limiting flame temperature upon which this law is based) leads to underestimation of the limits. The concept of a limiting burning velocity \( S_{u,\text{lim}} \) however, seems promising, as the dependence of \( S_{u,\text{lim}} \) on pressure, given by equation (2), is confirmed for the numerically calculated values of the burning velocity at the experimentally determined flammability limits. Additional calculations are being done at higher pressures and temperatures, as well as for propane-air mixtures, to further corroborate these findings.

REFERENCES

Figure 1: Comparison of numerically and experimentally determined upper flammability limits of methane-air mixtures

Table 1: Calculated values of the burning velocity $S_u$ and the maximum temperature $T_{\text{max}}$ for 1D planar premixed flames at the experimentally determined upper flammability limits of methane-air mixtures

$$S_u, \lim = a \cdot p^b$$