PROBLEM OF COMBUSTION STABILITY OF NATURAL GAS IN A FLUIDIZED BED

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The results are given of the analytical and experimental determination of the conditions of onset of stable combustion of natural gas in a fluidized bed.

The combustion of natural gas in a fluidized bed at temperatures of 925-1100°K and at atmospheric pressure creates specific difficulties associated with combustion instability. Under these conditions, the combustion of the mixture takes place predominantly in the bubbles, which is accompanied by intense pressure pulsations in the bed and considerable chemical underburning. Stable combustion, according to the data of [1, 2], starts at a bed temperature of 1025-1175°K. The large spread of the values obtained for the temperature of onset of stable combustion obviously is caused by the different conditions of conducting the experiments and the nonidentical determination of the onset of stable combustion. In this paper, the possibility is considered of an approximate calculation of the temperature of transition of combustion to the stable regime T^{cr}_{bed} as a function of the pressure, particle size, filtration velocity, and fuel mixture composition.

The calculation was started from the assumption that combustion converts to the stable state if the temperature of the bed particles exceeds the autoignition temperature of the mixture in the given specified conditions. The autoignition temperature of the mixture is found by the thermal explosion theory [3] from the conditions of equality of heat release and heat removal and their first derivatives with respect to temperature, i.e.,

$$q_+ = q_-, \tag{1}$$

$$\frac{dq_+}{dT} = \frac{dq_-}{dT}. \tag{2}$$

The heat release during combustion is determined by the flow velocity and by the thermal effects of the oxidation reaction of the natural gas hydrocarbons

$$q_+ = k \exp(E/RT)\, Q_p\, [\mathrm{CH}_4]^n\, [O_2]^m. \tag{3}$$

For this, we shall suppose that the components of the mixture during the induction period are not consumed.

In order to calculate the rate of oxidation of methane, of which natural gas consists predominantly, we have used the kinetic equations of the authors of [4]

$$-\frac{d\mathrm{CH}_4}{d\tau} = 10^{13.2\pm 0.2} \exp\left(-\frac{48400 \pm 1200}{RT}\right) [\mathrm{CH}_4]^{0.7} [O_2]^{0.8}, \tag{4}$$

which is true for T = 1030-1230°K and $\alpha_{mix} > 0.5$. For convenience of the calculations, this equation can be converted to the following expression:

$$-\frac{d\mathrm{CH}_4}{d\tau} = 3.7\cdot 10^{10} \exp\left(-\frac{48400}{RT}\right) \frac{\alpha_{mix}^{0.8}}{(1+\alpha_{mix} V_0)^{1.5}} \left(\frac{P}{T}\right)^{1.5}. \tag{5}$$

The heat of reaction in the period preceding autoignition of the gas is expended on heating the mixture from the temperature of the particles T_s to the temperature of preexplosive warmup T_{ig}, and is removed by the fluidized bed particles. The heat expended on heating up the mixture is insignificant and it can be neglected.

The heat removal by the particles was calculated by two methods. By the first method,

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$$q_{-} = \alpha F (T_{ig} - T_s). \tag{6}$$

Substitution of Eqs. (3) and (6) in the system of equations (1) and (2), and solution of the latter, gives the relation between T_s and T_{ig}

$$T_{ig} = T_s + \frac{RT_s^2}{E}. \tag{7}$$

In a fluidized bed, the role of the walls confining the gas flow is fulfilled by the particles. The intense intermixing of the particles permits the assumption of isothermicity of the bed and thereby the equality $T_s = T_{bed}$.

The heat-transfer coefficient from gas to particles was determined through the criterial equation [5]

$$\mathrm{Nu} = (\mathrm{Re}/\varepsilon)^{0.55} \mathrm{Pr}^{1/3}, \tag{8}$$

which satisfies our conditions ($\mathrm{Re} \leq 200$).

The specific surface of the particles depends on their size and the porosity of the layer

$$F = \frac{6(1-\varepsilon)}{d_s}. \tag{9}$$

According to the second method, we shall suppose that the gas is moving through the bed along a cylindrical channel and that between the particles and the gas only conductive heat exchange takes place. This latter does not contradict our conditions, as the Gr criterion in the range of parameters investigated is considerably less than $4 \cdot 10^4$, i.e., the values for which it is necessary to take account of heat exchange by convection. In this case, the heat removal can be determined from the critical condition of ignition [3]

$$\delta = \mathrm{const} = \delta_{cr}, \tag{10}$$

where

$$\delta = \frac{Q}{\lambda} \cdot \frac{E}{RT_s^2} k \exp(-E/RT_{ig}) r^2. \tag{11}$$

The quantity δ is a dimensionless parameter, the critical value of which for a cylindrical vessel of infinite length is equal to 2. From Eqs. (10) and (11) and taking Eq. (1) into consideration, we obtain

$$q_{-} = \delta_{cr} \frac{\lambda}{r^2} \cdot \frac{RT_s^2}{E}. \tag{12}$$

Equating the expressions for the heat release and heat removal, and using relation (7), we obtain a transcendental equation which it is convenient to solve by a graphical method. For

TABLE 1. Effect of Pressure and Particle Size on the Temperature of Transition of Combustion to a Stable Cycle, T_{bed}^{cr}, °K ($\alpha_{mix} = 1.0$)

d_s, mm	P, bar				
	1	3	5	10	15
0,5	1308	1173	1113	1053	1020
	1227	1167	1133	1095	1073
0,75	1235	1123	1063	1013	983
	1203	1143	1103	1073	1053
1,0	1193	1088	1033	983	950
	1180	1123	1087	1058	1040
2,0	1093	993	958	893	853
	1137	1080	1053	1033	1013

Note: Upper row — calculation of q_{-} by Eq. (12); bottom row — by Eq. (6).

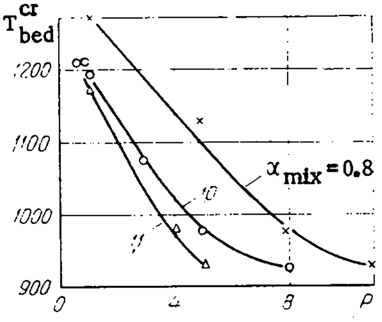

Fig. 1. Dependence of temperature of bed at which combustion converts to the stable regime on the pressure P, bar; T^{cr}_{bed}, °K; N = 2.5; $d_s = 0.75$ mm.

this, having assigned the temperature of the bed, we find the corresponding values of q_+ and q_-. The intersection of the relations obtained for $q_+ = f(T_{bed})$ and $q_- = f(T_{bed})$ on the graphs corresponds to the critical temperature of the bed for which the mixture is ignited and combustion converts to a stable regime. The results of the calculations carried out are given in Table 1. It can be seen from the table that the temperature of the bed at which combustion converts to a stable regime decreases significantly with increase of pressure and with increase of particle size. The effect of pressure is expressed through an increase of the rate of burnup of methane and, thereby, the heat release. Coarsening of the particles reduces the heat removal from the reaction zone, due to reduction of the heat-exchange surface.

An increase of the velocity of the fluidizing gas leads to an intensification of the interphase heat exchange and thereby to an increase of T^{cr}_{bed}. Some increase of porosity of the bed, leading to a decrease of the heat exchange surface, does not change the nature of this relation. Thus, an increase of the fluidization number in the calculations from 2 to 8 leads to an increase of T^{cr}_{bed} by 15-60°. The higher values refer to the coarser particles.

When assessing the results of the calculations, it must be taken into account that they have been obtained for the case of a uniform particle distribution in the volume and the instantaneous removal of heat by the particles from the zone of combustion into the depths of the bed. In actual conditions, combustion can take place in rarefied zones where the particle concentration is significantly less than the average over the bed. This is particularly characteristic for bubble-cap gas-distributors. A reduction of the particle concentration reduces the heat removal from the combustion zone. The low mobility and low heat conductivity of the particles may also contribute to this reduction. Taking these factors into consideration, it can be supposed that the actual temperature of transition of combustion to a stable regime may be lower than the calculated temperature. Thus, the values of T^{cr}_{bed} given in Table 1 are boundary values, i.e., those for which the mixture certainly should be ignited.

The experimental determination of the temperature of onset of stable combustion was conducted in an apparatus with an inside diameter of 132 mm [6]. A bubble-cap grid with a useful cross section of 0.36% served as the gas distributor. Globular corundum of two fractions, with an equivalent particle diameter of $d_s = 0.75$ and 1.22 mm, was used as the fluidized material. The fuel was natural gas, with a lowest calorific value of 36.6 MJ/m³. The experiments were conducted under the following conditions: pressure P = 1-20 bar, temperature in the core of the bed $T_{bed} = 923\text{-}1423$°K, air flow rate coefficient $\alpha_{mix} = 0.65\text{-}1.5$, and fluidization number N = 2-5.

For the onset of stable combustion, a temperature of the bed was assumed at which bursting of the gas into bubbles ("flocs") ceased, and the composition of the combustion products was stabilized within the limits of the bed. The pressure pulsations in the bed were measured by means of tensometric sensors by the procedure of [7]. The presence of sections of stable composition of the combustion products was determined by the analysis of gas samples, taken over the height of the bed.

The values of T^{cr}_{bed} obtained as a function of the pressure for an air flow rate coefficient of 0.8, 1.0, and 1.1, and $d_s = 0.75$ mm are shown in Fig. 1. It can be seen that at atmospheric pressure quite good agreement is obtained between the experimental and calculated values of the temperature of stable combustion transition. In the region of higher pressures, the values of T^{cr}_{bed} obtained experimentally are somewhat lower than the calculated values.

This discrepancy is less when the heat removal is calculated by Eq. (12) which, obviously, better describes the conditions of heat exchange in the fluidized bed than Eq. (6). The difference between the experimental and calculated values of T^{cr}_{bed} can be explained by the limited applicability of Eq. (4) for low temperatures. As shown above, with increase of pressure, the autoignition temperature of the mixture is reduced to 930-1000°K. At these temperatures, an increase of the order of the methane oxidation reaction to values of 2.65 occurs [8]. The order of the reaction in Eq. (4) is equal to 1.5.

The effect of the air flow rate coefficient on the temperature of stable combustion transition, as can be seen from Fig. 1, is more marked than is obtained by the analytical calculation. This is particularly marked for rich fuel mixtures.

NOTATION

d, diameter; E, energy of activation; F, specific surface of particles; Gr, Grashof number; k, reaction velocity constant; N, fluidization number; Nu, Nusselt number; P, pressure; q_+, heat release; q_-, heat removal; Pr, Prandtl number; R, universal gas constant; Re, Reynolds number; r, characteristic dimension; T, temperature; V_0, theoretical air flow rate necessary for combustion; α, heat transfer coefficient from gas to particles; α_{mix}, air flow rate coefficient; ε, porosity of bed; λ, thermal conductivity of gas; τ, time; indices: ig, ignition; cr, critical; s, particles.

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