

Effect of Tube Size and Obstacles on Explosion Limits in Flowing Gases

Jeroen W. Bolk and K. Roel Westerterp

Chemical Reaction Engineering Laboratories, Dept. of Chemical Technology, University of Twente,
7500 AE Enschede, The Netherlands

In a large pilot plant the upper explosion limit of ethene–air–nitrogen mixtures was experimented in 3.0-m-long and 21-, 50-, and 100-mm-dia. tubes at different flow rates, pressures, and temperatures. The upper explosion limit, influenced by the gas velocity, becomes smaller and shifts to higher oxygen concentrations for increasing flow rates. The results of these tubes could be correlated based on the tube Reynolds number. A cooling effect of the tube wall, which might influence the explosion region, was not observed. An increase in pressure lowers the critical oxygen content as does an increase in temperature, thus the explosion region becomes larger. Different obstacles were tested, which alter the hydrodynamics. Reaction fronts could only propagate for increased oxygen concentrations through a structured Sulzer laboratory gauze packing or a sudden reduction in diameter from 50 to 20 mm. In the experiment, where the tube was completely filled with glass spheres, propagation of reaction fronts through this packed bed was not possible, even at very low gas velocities and very high oxygen concentrations. In a deep dead zone connected to the tube, the gas was ignited and reacted without igniting the gas outside the dead zone. After some time the reaction stopped because of oxygen starvation. However, dead zones with another geometry, where renewal of the combustible gas can occur, still may be dangerous.

Introduction

Gas-phase oxidation reactions are widely applied in the chemical industry. These reactions can be dangerous, so to guarantee safe operation, most partial oxidation processes are operated outside the explosion region.

Explosion limits are usually determined in well-defined laboratory equipment, such as explosion bombs or tube, and in most cases a spark is used as the ignition source (see, e.g., Lewis and Von Elbe, 1961; Zabetakis, 1965; Lovachev et al., 1973). Explosion limits can be influenced by the geometry and the size of the equipment. Since heat from the flame front in a tube is transferred to the tube walls by radiation, conduction, and convection, a reaction front may be quenched, if the tube is too small. According to Zabetakis (1965) at atmospheric pressure and room temperature a 2-in.-ID vertical tube can be used to measure explosion limits for hydrocarbons without any effect on the tube wall. Jones

and Kennedy (1930) determined flammability limits of ethene mixtures at atmospheric pressures, and found these limits narrow as the tube diameter was increased, rapidly at first and more slowly at higher diameters. Tests were performed in tubes with diameters varying between 14 and 75 mm, and an increase in the tube diameter above 50 mm did not show more than a few tenths of a vol % decrease in the flammability limit. White (1924) performed experiments in tubes with internal diameters of 25, 50 and 75 mm, respectively. Explosion limits were independent of the tube size for diameters above 50 mm. Below this critical value the measured explosion regions were smaller, due to the cooling effect of the tube wall.

Starke and Roth (1989) investigated the flame behavior during explosions in cylindrical enclosures with obstacles: due to the obstacles the combustion process was enhanced significantly, because of an increased mixing rate. Phylaktou and Andrews (1991) placed a single baffle in a 76-mm-dia. tube with a total length of 1.64 m and quantitatively determined

Correspondence concerning this article should be addressed to K. R. Westerterp.

the characteristics of explosions in methane-air mixtures. Flame speeds and rates of pressure rise were enhanced downstream of the baffle, and the effects became larger with increasing baffle sizes. For large blockages local quenching of the flame was found in the high turbulence regions. Grätz (1995) placed a Sulzer Mellapack M500.Y with lengths of 400 and 600 mm and a Sulzer Mellapack M250.Y with a length of 1,000 mm in a pipe with an internal diameter of 100 mm and a total length of 33 m; in this pipe a mixture of 50% acetylene and 50% of oxygen was ignited outside the packing at a pressure of 0.2 MPa. Depending on the conditions, a deflagration or detonation propagated through the tube. This type of packing could not stop a detonation, but it was effective in stopping a deflagration.

Siccama and Westerterp (1993) developed an alternative method for determining explosion limits that resembles the industrial situation better and in which a combustible gas mixture of ethene, air, and nitrogen in their case flows through empty tubes and is ignited with an electrically heated wire. Siccama and Westerterp (1993) and Bolk et al. (1996) showed that the upper explosion limit strongly depends on the flow rate of the gas; an increase in flow rate makes the explosion region smaller.

Bolk and Westerterp (1999) developed a CFD model that could explain both qualitatively and quantitatively the relation between the upper explosion limit and the geometrical

and operating conditions for a flow of ethene, air, and nitrogen mixtures through a 21- and 50-mm-diameter tube. They also derived an expression relating the explosion limits for flow and nonflow conditions with the chemical and physical properties of the system and with the tube Reynolds number.

In this article we describe the extension of the work of Bolk and Westerterp (1999) to different tube diameters, to tubes filled with packing, or tubes with restrictions and to the influence of dead zones. Also a structured Laboratory Sulzer gauze packing has been used, which can be seen as a kind of flame arrester. In a partial oxidation process the system can be filled with packing to prevent reaction fronts from propagating through the installation; a structured Sulzer packing is very suitable for this, because it exhibits a low pressure drop.

Experimental Installation and Procedure

Experiments have to be performed under conditions like those in an industrial ethene oxidation process, which means that high pressures, temperatures, and gas flow rates are needed. In that case once-through flow through the explosion tubes is impossible, because the gas consumption will become disproportionately high. Therefore, a large recycle installation has been constructed in a concrete bunker in our specially equipped high-pressure laboratory. The installation is fully computer controlled (see Figure 1).

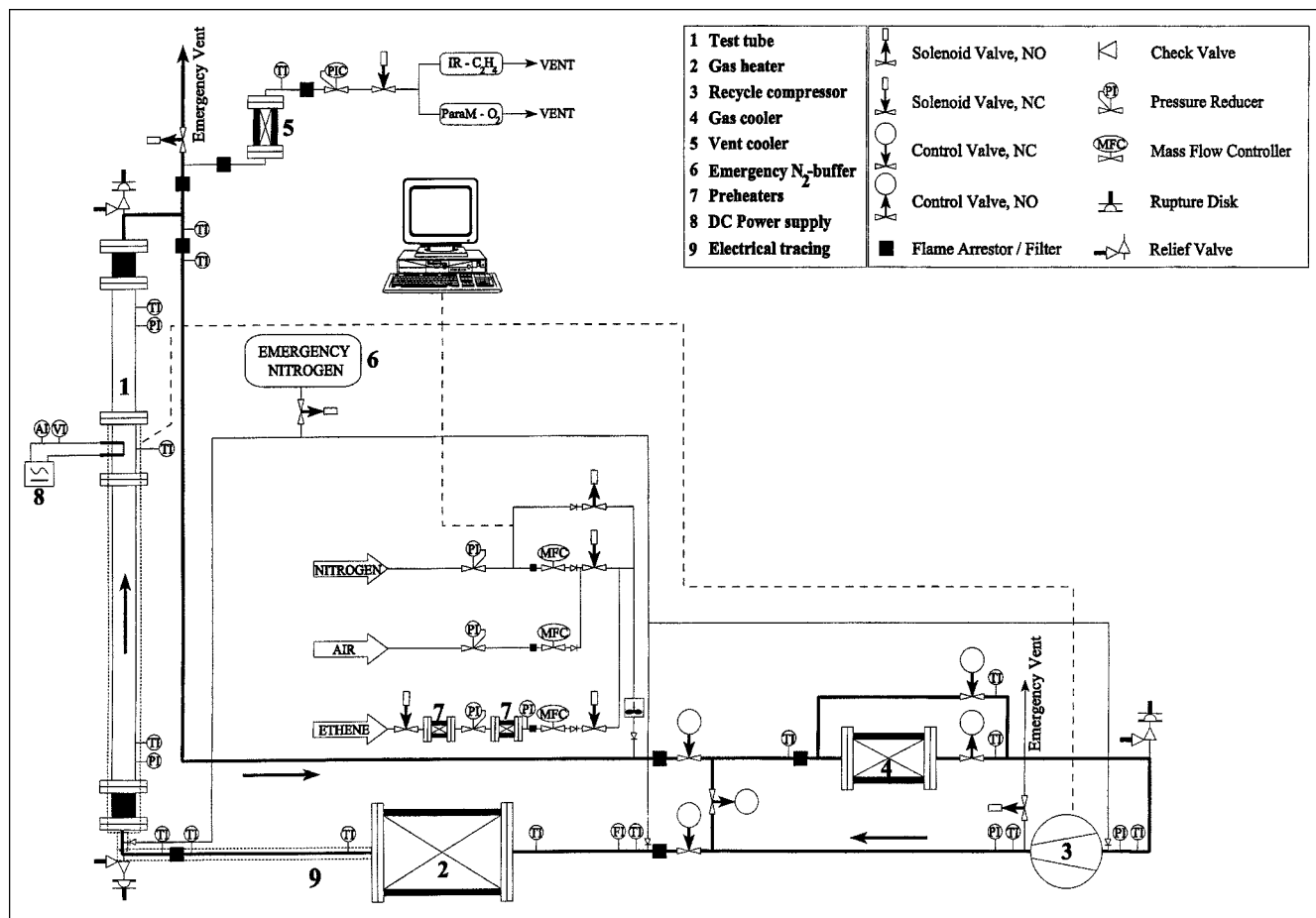


Figure 1. Test installation.

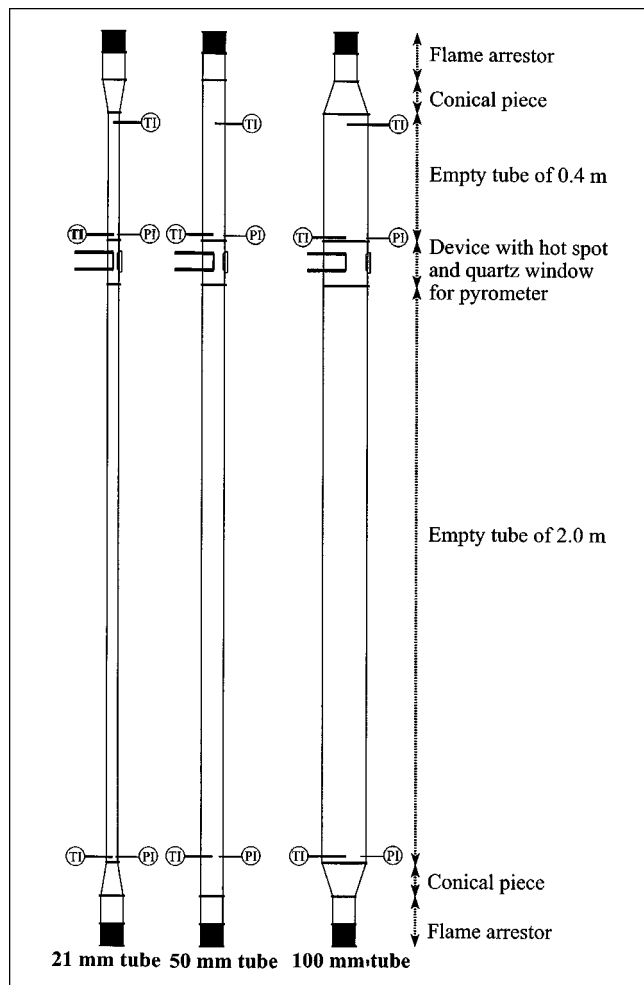


Figure 2. Three different test tubes.

Three different tubes with internal diameters of 21, 50 and 100 mm were used (see Figure 2). An electrically heated vertical wire with a length of 40 mm and a diameter of 0.6 or 1.0 mm was used as the ignition source in the tube. It was made of Kanthal A1, a special alloy of mainly Fe, Cr, and Al. The recycle compressor has a maximum flow capacity of 30 m³/h at all pressures. Heating and cooling equipment was installed in the recycle system to protect the compressor, which cannot handle temperatures above 425 K, and to heat the gas to a maximum temperature of 573 K. A detailed description of the installation is given in Bolk and Westertep (1999) and will not be given here. A detailed description of the experimental procedure can also be found there.

The 50-mm tube was used to study the effect of obstacles on the critical oxygen concentration in the explosion point. The experimental procedure is exactly the same as in the situation with no obstacles. In a first set of experiments a structured Laboratory Sulzer gauze packing 170 mm long and 35 mm in diameter was fitted in the tube at two positions: one close to and above the ignition wire at a distance of 0.06 m, while in the other position the packing is located further away, at a distance of around 0.25 m (see Figure 3). The packing was fitted in a tube with an internal diameter of 35 mm and an outer diameter of 50 mm; this tube was placed in the 50-

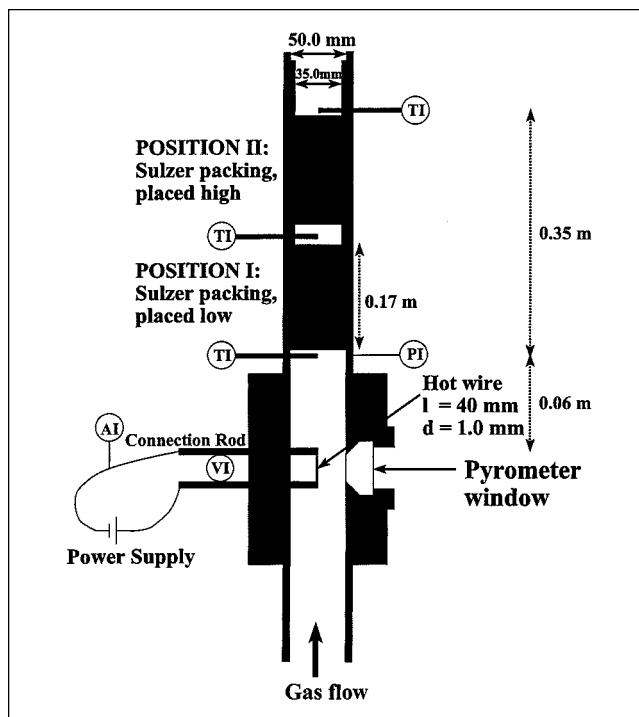


Figure 3. Setup with Laboratory Sulzer gauze packing as obstacle.

mm explosion tube. The critical oxygen concentration was determined at room temperature and at different pressures and gas velocities.

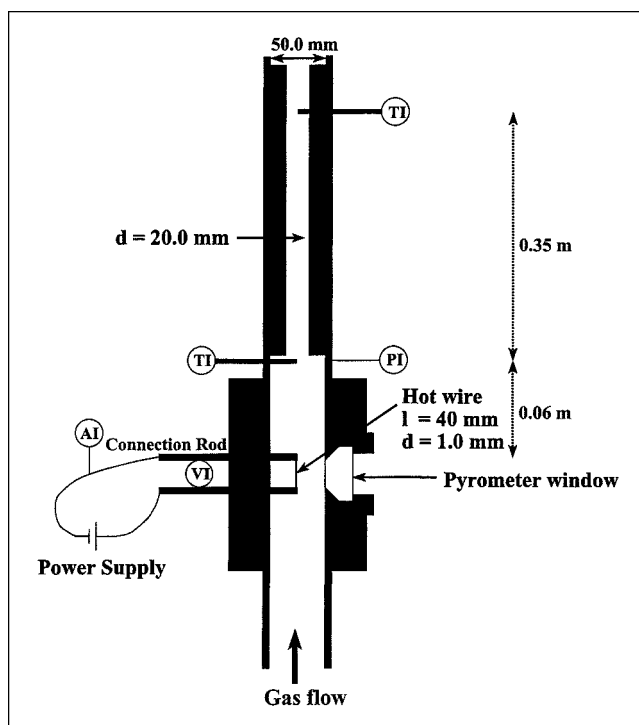


Figure 4. Setup with diameter reduction from 50 to 20 mm.

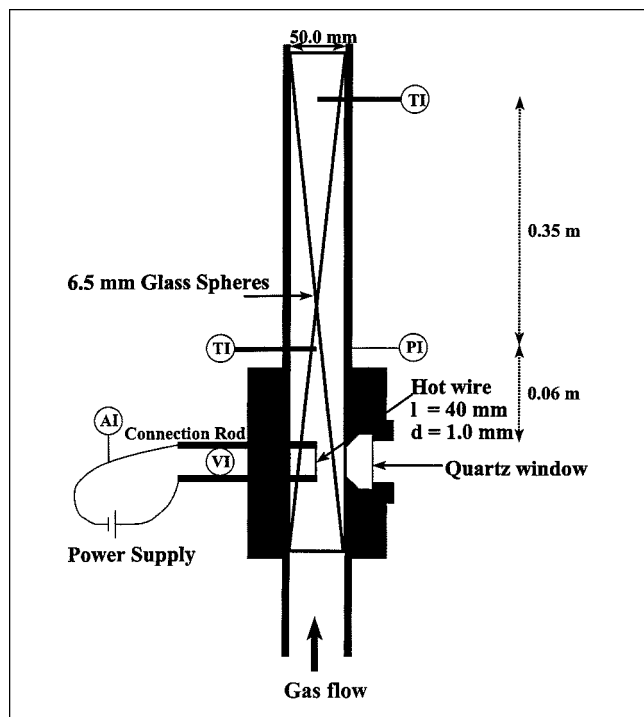


Figure 5. Setup with 50 mm tube filled with glass spheres of 6.5 mm.

In a second set of experiments, a sudden reduction in diameter from 50 to 20 mm was placed in the tube (see Figure 4). The critical oxygen concentration was determined by observing the temperature rise measured with a thermocouple placed in the 20-mm tube part.

In a third set of experiments the 50-mm tube was filled completely with glass spheres 6.5 mm in diameter (see Figure 5). Type K thermocouples with a diameter of 0.5 mm were mounted in the bed at distances of 0.06 and 0.40 m, respectively, from the ignition source. Propagation through the bed could be observed by a temperature increase in the bed, and these conditions were determined.

In Figure 6 an overview is given of the configuration for investigating the danger of dead zones in a fourth set of experiments. Perpendicular to the 50-mm tube a 20-mm-dia. tube 0.2 m long was connected. In this side tube a vertical ignition wire 20 mm long and 1.0 mm in diameter and a thermocouple to measure the temperature to detect an ongoing reaction were placed. A small gas flow, controlled by an electronic Brooks mass flow controller, could be fed through the side tube to refresh the gas. If the temperature in the main tube rose suddenly more than 200 K, propagation into this tube was supposed to occur and the experiment stopped. Experiments have been performed with and without gas flow through the side tube at different pressures and gas velocities.

Results and Discussion

In all experiments the ethene concentration was kept constant at 25 vol %. For the influence of the oxygen concentration, see Eq. 27 in Bolk and Westerterp (1999), which reads

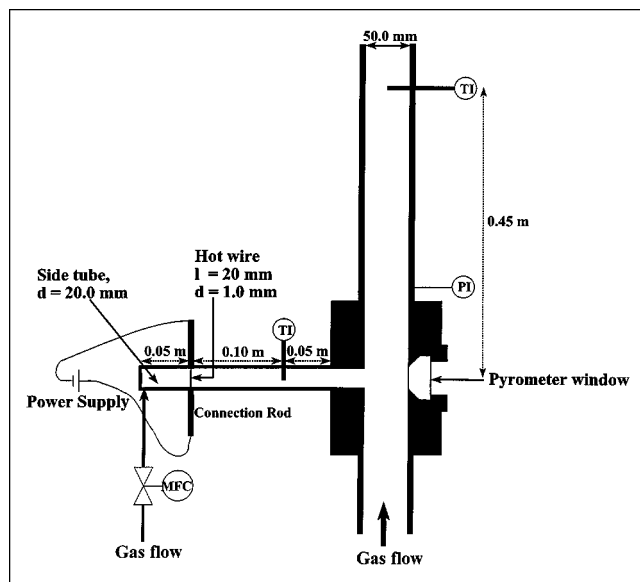


Figure 6. Experimental configuration with ignition from a dead zone.

$$\left(X_{O_2, Re} - X_{O_2, Re \rightarrow 0} \right) = 1.53 \times 10^{-3} \frac{RT_{\text{flame}} C_p}{P \Delta H_{\text{comb}}} \times (T_{\text{flame}} - T) Re^{1.0}. \quad (1)$$

The power supply to the wire and the oxygen concentration were varied to obtain the typical explosion diagram (see Figure 7). In this diagram three different regions can be seen: no reaction, local reaction, and explosion. No reaction occurs when the power supply rate to the wire, and thus the temperature of the wire, is too low for ignition. Increasing the power supply to the wire causes a temperature increase and initiation of combustion reactions at or above a specific supply rate, which we defined as the minimum power supply rate. This minimum is independent of the oxygen concentration and can be represented by a horizontal line in the explosion diagram of Figure 7.

Under these conditions a local reaction starts around the heated wire or there is an explosion through the tube, de-

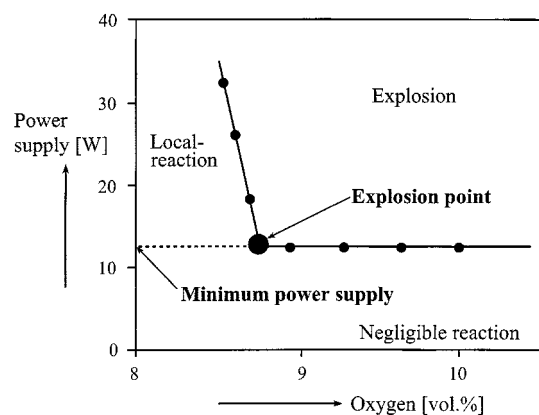


Figure 7. General explosion diagram.

pending on the oxygen concentration. At low oxygen concentrations the heat produced by the combustion reactions is low and counterbalanced by the heat removal from the reaction front to the unburnt gas (see Semenov, 1928; Frank-Kamenetskii, 1969). These heat removal processes are enhanced significantly in a turbulent flow (see Bird et al., 1960). The reaction zone remains confined to a specific volume around the ignition wire. Increasing the oxygen concentration will increase the heat produced by the combustion reactions, and at a critical concentration, this heat cannot be removed anymore, so a reaction front starts to propagate through the entire tube. The boundary between the local reaction and explosion in the explosion diagram of Figure 7 can be given by an almost vertical line with a negative slope. We defined the intersection between the lines that separate the local reaction and explosion region and the minimum power supply as the explosion point (see Siccama and Westerterp, 1993). A physical explanation of the three different regions in the explosion diagram is also given in Bolk and Westerterp (1999).

In our case an experiment is terminated as soon as the gas is ignited, in order to prevent flames propagating into the recycle system. In industrial plant operation it is important to know the maximum pressure rise after such an ignition. Therefore, an explosion installation without recycle was constructed to measure these maximum pressure rises in the 21-mm tube. Results are given in the Appendix.

Effect of tube diameter

At room temperature and pressures of 0.5 and 1.0 MPa experiments have been performed at different gas flow rates in the 21-, 50- and 100-mm tubes, respectively. Results are given in Figure 8. In the 21-mm and 50-mm tubes the gas velocity has been varied between 0.25 and 2.5 m/s, and in the 100-mm tube between 0.2 and 0.8 m/s. The critical oxygen concentration at the explosion point is the same in all the tubes for equal Reynolds numbers, which are based on the inner-tube diameter and the density and viscosity of air at initial conditions.

An explosion can only propagate when the heat generated by the combustion reactions is larger than the heat removed via the turbulence from the flame front to the unburnt gas.

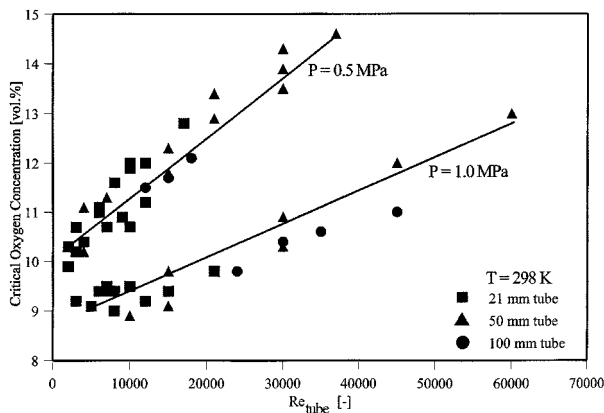


Figure 8. Pressure influence on the critical oxygen concentration at the explosion point in different test tubes.

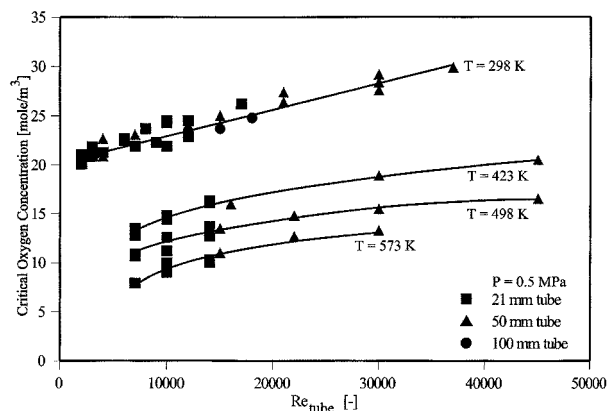


Figure 9. Temperature influence on the critical oxygen concentration at the explosion point in different test tubes.

At equal Reynolds numbers the flow regime in the tubes, and therefore the turbulence, is the same, and the heat removal processes from the flame front to the unburnt gas are equal. At the equal heat removal rate, the combustion reactions must produce an equal or greater amount of heat to create an explosion; thus, the same critical oxygen concentrations are found in the different tubes (see Bolk and Westerterp, 1999).

Wall effects could not be observed: in that case, the critical oxygen concentrations in the 21-mm tube should be higher compared to those in the other tubes, due to the additional cooling at the wall. At atmospheric pressures the explosion limits are influenced by the tube size for diameters smaller than 50 mm. However, we performed experiments at a minimum pressure of at least 0.5 MPa, and at these higher pressures the influence of the tube wall is negligible: apparently the critical diameter for wall cooling being important is now below 21 mm. Only some results in the 100-mm tube at a pressure of 1.0 MPa deviate, especially at high Reynolds numbers. There is no clear explanation for this effect.

The influence of the gas temperature has been investigated only in the 21- and 50-mm tube. Results for a pressure of 0.5 MPa are given in Figure 9. Again the critical oxygen concentrations for the two tubes coincide at the same Reynolds number. An increase in temperature decreases the critical oxygen concentration and the explosion region is enlarged, as was observed by Bolk and Westerterp (1999). Critical oxygen concentrations in the three tubes are equal for the same Reynolds number. All experimental data in the three tubes are covered well by the correlation of the oxygen concentration at the explosion point with the process variables in the tube flow (see Eq. 1).

In the remaining experimental program, only the 50-mm tube was used, in which different obstacles have been placed to study the effect on the upper explosion limit. In all situations a vertical Kanthal A1 wire 40 mm long and 1.0 mm in diameter was used.

Effect of structured Sulzer packing

The structured Laboratory Sulzer gauze packing was placed at two positions in the tube: as close to the ignition source as possible (at 0.06 m), and further away (at 0.25 m). The exper-

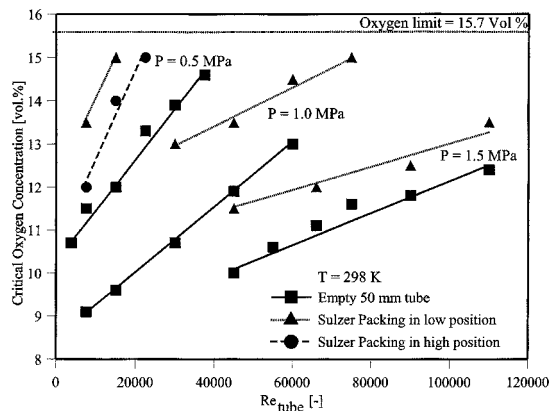


Figure 10. Effect of Sulzer packing on critical oxygen concentrations.

iments were performed at room temperature and pressures of 0.5, 1.0 and 1.5 MPa: results are given in Figure 10. The critical oxygen concentrations are given for which the reaction propagated through the packing.

The packing acts as a flame arrestor, in which the heat removal processes are enhanced, due to energy losses by distortion of the flow, heat transfer, and friction, and the reaction fronts may be quenched. At a pressure of 0.5 MPa the highest critical oxygen concentrations are found for the packing placed in the low position; apparently this is the best position to quench reactions. Close to the ignition source, the reaction fronts are probably not yet fully developed and restricted to a confined volume. Further away from the ignition source, these fronts have probably already developed over the full tube diameter, and quenching becomes more difficult.

The effect of pressure was investigated, with the Sulzer packing placed only in the low position. For increasing pressures the difference between the critical oxygen concentrations with and without the packing becomes smaller. Thus, quenching by the Sulzer packing is less effective at higher pressures: at high oxygen concentrations and high pressures it is still possible for the reaction fronts to propagate through this type of packing.

Glycol and polyglycols will always be present in ethene oxide plants, due to the reaction between ethene oxide and the water, formed by the total oxidation of ethene. Furthermore some soot may be formed. These substances will build up in the recycle system, and their deposits on a packing or flame arrestor may become a practical problem.

Effect of an abrupt reduction in tube diameter from 50 to 20 mm

Experiments for a sudden reduction in diameter from 50 to 20 mm have been performed at room temperature and at pressures of 0.5, 1.0 and 1.5 MPa; results are given in Figure 11. In this figure the critical oxygen concentrations are given for reactions propagating into the 20-mm tube and compared to those found in the empty 50-mm tube. In both cases the Reynolds number is based on a tube diameter of 50 mm. Due to the increase in velocity in the smaller 20-mm tube, the heat removal processes are enhanced. To counterbalance, the heat production rate must be increased to sustain an explo-

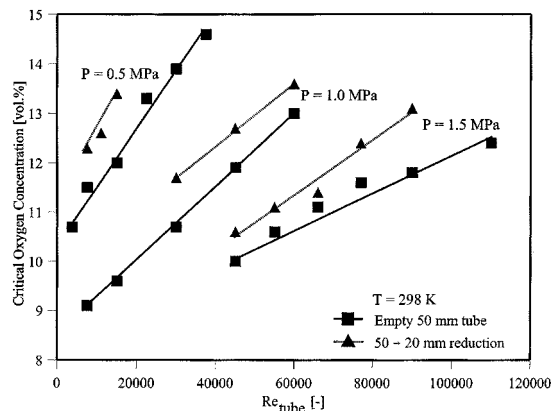


Figure 11. Effect of diameter reduction from 50 to 20 mm on critical oxygen concentrations.

sion; thus, higher critical oxygen concentrations are required. Only a small pressure influence can be observed, because at the three pressure levels the critical oxygen concentration does not change much; in Figure 11 this difference varies between 0.5 and 1.5 vol %. The required increase in $X_{O_2, Re} - X_{O_2, Re \rightarrow 0}$ cannot be predicted simply on the basis of the change in the Reynolds number (Re) as given in Eq. 1. We need to know that the ignition source is located in the 50-mm tube, whereas the 20-mm tube starts only at a distance of 60 mm after the heating wire.

Effect of a packed bed

For the tube filled with glass spheres 6.5 mm in diam the conditions have been determined at which reactions propagate through the bed at room temperature and for pressures of 1.0 and 1.5 MPa. No temperature and pressure increase was observed in the bed at superficial gas velocities of 0.25 m/s and higher, not even with oxygen concentrations up to 15 vol %. Therefore at an even lower superficial gas velocity of 0.1 m/s, experiments were performed for the conditions given in Table 1.

The power supply to the wire was at its maximum in experiments II, III, and IV. Indeed, under these conditions a local reaction was initiated around the ignition wire, but it could not propagate through the bed. With the thermocouple placed 0.06 m above the ignition source, a maximum temperature increase of approximately 100 K was measured, and with the thermocouple placed at a distance of 0.40 m, there was no temperature increase at all. Only a small and smooth pressure increase was measured in the tube, whereas the oxygen concentration in the recycle stream decreased.

Table 1. Experimental Conditions in the Packed Bed with Bed Porosity at 50%

Exp.	P [MPa]	O_2 [Vol %]	Power Supply [W]	Re_p	Re_t
I	1.0	15.0	53	380	5,900
II	1.0	14.5	80	380	5,900
III	1.5	12.0	76	570	8,900
IV	1.5	14.5	83	570	8,900

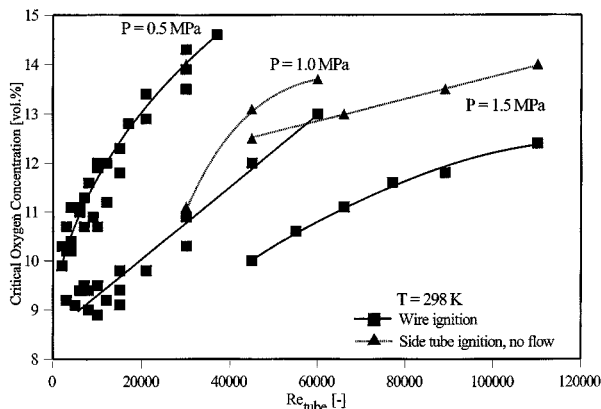


Figure 12. Critical oxygen concentrations for dead zone ignition, no gas flow through dead zone.

After every experiment the tube was opened and the packed bed inspected. Locally around the ignition source soot was found on the glass spheres, and after experiment IV locally molten glass spheres were found around the ignition wire. At places further away from the ignition source, the packed bed was still unchanged and no formation of soot was visible.

In the bed the flow is disturbed and much turbulence created, which enhances the heat removal processes. Furthermore, the glass spheres can be regarded as a heat sink, because their heat capacity is much larger than that of the gas, and heat is transferred from the gas to the packing. All these processes prevent the propagation of the reaction: the reaction zone remains localized around the ignition source. At a pressure of 1.0 MPa the $Re_{particle}$ is 380 and the Re_{tube} is 5900, and at a pressure of 1.5 MPa these values are 570 and 8,900, respectively. Thus the flow is turbulent in all cases.

Effect of dead zones

Experiments were performed at room temperature and pressures of 0.5, 1.0, and 1.5 MPa to investigate what happens in a dead zone: to this end a side tube was mounted on

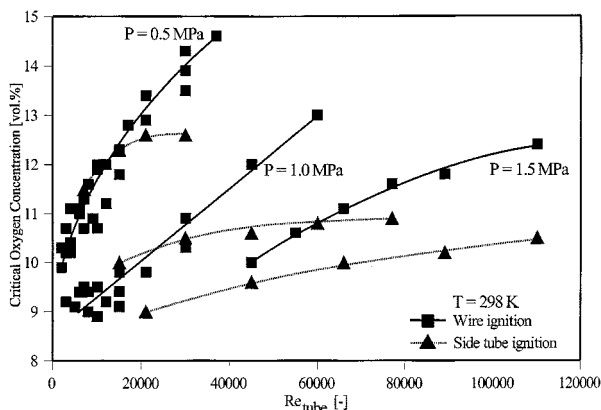


Figure 13. Critical oxygen concentrations for dead zone ignition, with gas flow through dead zone.

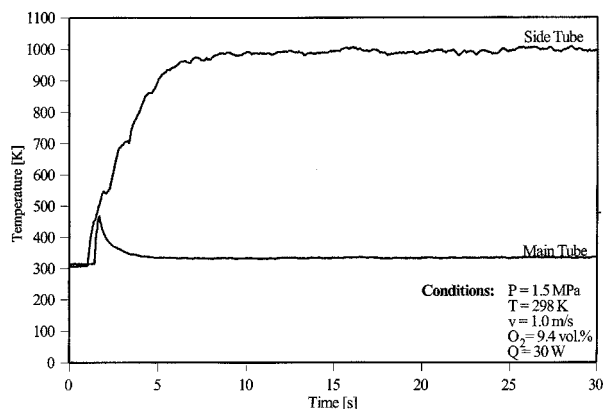


Figure 14. Temperature profiles in case of a combustion reaction in the dead zone without ignition of the gas in the main tube.

the main tube (see Figure 6). Experiments with no gas flowing through the side tube were performed (see Figure 12), and afterward with gas flow (see Figure 13).

In the side tube the gas was ignited with an electrically heated wire 20.0 mm long and 1.0 mm in diameter. Once the gas has been ignited, a reaction front starts to propagate through the side tube. In case of no gas flow, the reaction will stop as soon as the oxygen has been consumed. In the case of gas flow through the side tube, a completely different phenomenon was observed. Once the gas is ignited, it is constantly refreshed, and therefore the reaction will not be extinguished. The maximum gas velocity in the side tube was 0.2 m/s. A continuous flame was created in the side tube, which can be seen as a jet of hot gas entering the main tube. Under most conditions this flame cannot ignite the gas in the main tube, whereas temperatures may increase to more than 1,000 K in the side tube (see Figure 14, in which the temperatures in the main tube and the side tube have been plotted as a function of time). In the main tube a small temperature increase is measured, which dies out. In the side tube the temperature gradually increases to around 1000 K, due to the combustion reaction, which is continuously fed by fresh gas entering the side tube.

In Figure 12 critical oxygen concentrations are given for the case of no gas flow through the side tube. These concentrations are higher compared to the situation where gas is ignited in the main tube with a vertical wire as the ignition source. The local heat flux coming out of the side tube is apparently less severe than around the heated ignition wire.

The opposite occurs for gas flow through the side tube, where the critical oxygen concentrations are lower (see Figure 13). Now, instead of a short plow of hot gas, a continuous flame extends out of the side tube into the stream in the main tube. Apparently the heat flux out of the side tube is now more severe than around the heated ignition wire. Thus, the case of constant hot gas feed into the main tube is dangerous. Fortunately, the chances that a stream of combustible gas enters through a dead zone are very low in practice.

Our results, of course, are only valid for our type of dead zones with a large L/D ratio. Dead zones with a different geometry, for example, with a low L/D ratio like a manhole,

will behave differently. Mixing the combustible gas from the main tube into the dead zone may be possible, in which case a reaction can still propagate from the dead zone into the main tube.

Conclusions

The influence of hydrodynamics on the upper explosion limit of ethene-air-nitrogen mixtures was studied. Experiments were performed at different gas flow rates in three specially designed explosion tubes, with internal diameters of 21, 50 and 100 mm. In all cases the flow was turbulent. We could very clearly distinguish between ignition, local reaction, and deflagration or propagation of the reaction through the tube. The results in the three different tubes can be correlated by Eq. 1. No effect of the tube wall was observed. Experiments in quiescent stagnant mixtures give the lowest critical oxygen concentration, which makes this the worst-case situation for the design of industrial processes, with the highest operating costs.

Different obstacles were placed in the 50-mm tube. These obstacles altered the hydrodynamics of the system and increased the heat removal rate. A special structured Laboratory Sulzer gauze packing, which can be seen as a flame arrestor, was placed in the tube. The best position to quench reactions is as close to the ignition source as possible. The reaction fronts have not yet fully developed and still can be quenched easily. This quenching effect is less effective at higher pressures.

A sudden increase in gas velocity in a tube diameter going from 50 to 20 mm results in critical oxygen concentrations that are around 1.0 vol % higher, compared to the empty 50-mm tube, for all three pressure levels measured.

Also the tube was completely filled with glass spheres. No propagation through the glass bed was observed, even not at $u_s = 0.1$ m/s. A local reaction, which cannot propagate through the bed, occurs around the ignition wire; under certain conditions the local temperature has been so high that the glass spheres have melted in a restricted volume near the wire.

Dead zones will always be present in commercial equipment. If no fresh gas is mixed into the dead zone, gas ignited in the dead zone will consume all the oxygen present without igniting the gas in the main stream. The gas in the main tube can only be ignited for oxygen concentrations significantly higher than those in gas ignited in the main gas stream. These results were obtained in a deep dead zone with a large L/D ratio. Dead zones with a different geometry will behave differently.

The results obtained in this study can be used to redesign partial oxidation plants, such as the partial oxidation of ethene to ethene oxide. Apparently, they can be safely operated with higher oxygen concentrations, provided the gas flow rates are kept high. Dangerous areas in the plant can also be avoided by filling them with structured material or just with glass or metal spheres, because in such areas flame propagation becomes very difficult and even impossible.

Acknowledgments

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Notation

- C_p = heat capacity, J/kg · K
 d = diameter, m
 ΔH_{comb} = heat of combustion, J/mol · O₂
 R = gas constant, 8.314, J/mol · K
 T = temperature, K
 v = velocity, m/s
 X = volume fraction oxygen at the upper limit
 μ = viscosity, Pa · s
 ρ = density of the gas, kg/m³

Subscripts and superscripts

- flame = flame
 O₂ = oxygen

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Appendix: Maximum Pressure Rise after Ignition

In our setup propagation of reaction fronts could be avoided and formation of soot reduced outside the explosion tubes. Due to the experimental procedure maximum pressure rises were only a few tenths of a bar. A smaller installation without recycle was constructed in which the maximum pressure rise after ignition can be measured. The explosion tube with an internal diameter of 21 mm was placed in a concrete bunker. The installation is fully computer controlled, and is given in Figure A1. An electrically heated vertical wire 40 mm long and 1.0 or 1.5 mm in diameter, made of Kanthal A1, was used as the ignition source. Experiments were performed only at room temperature with upward gas flow. Maximum flow rate through the tube was restricted to 0.75 m/s at $P=1.0$ MPa due to the size of the mass flow controllers, which gives a maximum Reynolds number of 10,000.

An experiment is started by flushing the installation with nitrogen to remove all oxygen. Then the gas composition and velocity are set by adding air and ethene via calibrated mass flow controllers. The gases are mixed in a gas-mixing section, identical to the one in the large installation, and fed directly

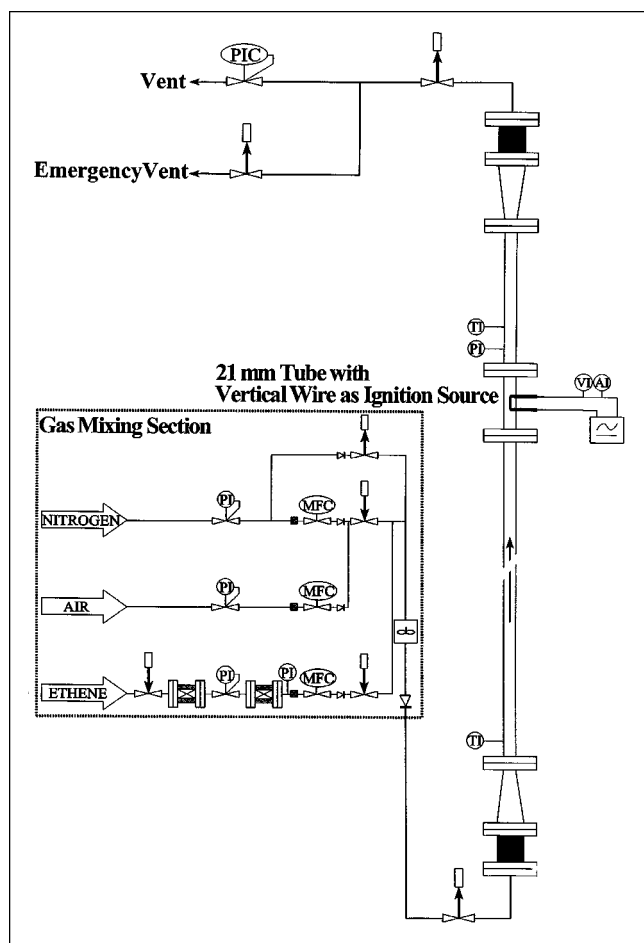


Figure A1. Experimental installation.

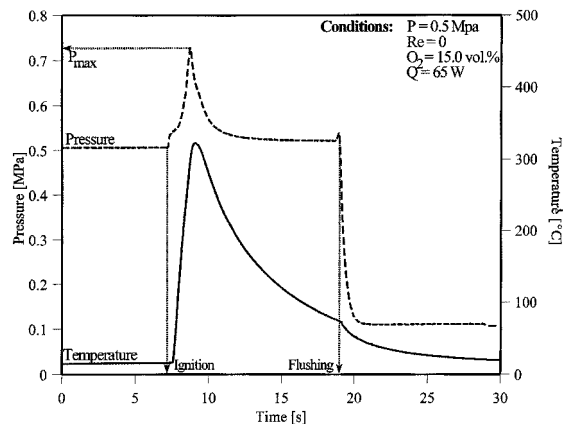


Figure A2. Pressure and temperature increase in a closed tube.

to the tube. The ethene concentration is kept constant at 25 vol %. Subsequently a valve in the emergency vent is closed and the pressure is automatically set by throttling an electronic back-pressure controller in the vent (see Figure A1). In the case of an experiment at zero gas velocity in a closed tube, two valves are closed at both ends of the tube just before the wire is heated.

An explosion experiment can then be executed: the wire is heated electrically for 30 s. The pressure and temperature in the tube downstream of the wire, the power supply to the wire, and the surface temperature of the wire are measured and recorded with a rate of 136 Hz. The gas mixture is considered to be ignited when the temperature rises more than 200 K or the pressure suddenly increases more than 0.05 MPa at least. After that the power supply to the wire is switched off after a short period that varies from zero to several seconds, and then after at least 10 seconds, all valves are set in the fail-safe position to flush the installation with nitrogen. During the time between ignition and flushing, the pressure rises can be measured. Experiments were performed at different gas velocities and in stagnant gas at pressures of 0.5, 1.0, 1.5 and 2.0 MPa in a closed tube.

Experiments were performed in the explosion region with oxygen concentrations somewhat higher than the critical oxygen concentration at the explosion point. Increasing the oxygen concentration by some percent, and therefore moving further into the explosive region, does not change the results significantly; approximately the same maximum pressure rise is measured in all situations. The conversion of oxygen is probably incomplete.

A typical time duration of the pressure and temperature of a stagnant gas in a closed tube is given in Figure A2. At $t = 7.2$ s, the gas is ignited and the pressure and temperature increase are measured. The maximum pressure rise to 0.73 MPa is recorded 1.5 s after ignition, after which both the temperature and pressure decrease in time. At $t = 18.7$ s, all valves are set in the fail-safe position and the installation flushed with nitrogen.

Similar experiments were repeated with gas flow through the tube. Typical results are given in Figure A3. At $t = 8.0$ s, the gas is ignited and the pressure and temperature increase measured. The power supply to the wire is switched off 3 s

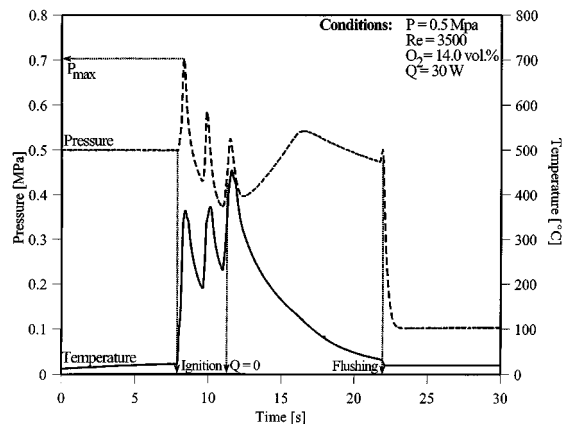


Figure A3. Pressure and temperature increase in an open tube.

after this ignition, and in the intermediate time combustible gas is still fed to the tube with the hot wire. The back-pressure controller in the vent responds to the pressure increase in the tube, with the result that after 1.5 and 3.0 s, a second and third ignition are measured with accompanying pressure and temperature increases. After the third ignition the power supply to the wire is switched off, and consequently no ignition is observed anymore. The experiment is stopped 10 s later: all valves are set in the fail-safe position and the installation is flushed with nitrogen. The maximum pressure rise to 0.71 MPa is measured after the first ignition. The results are given in Table A1 for experiments at other initial pressures, oxygen concentrations, and gas velocities: the maximum pressure rises are approximately 1.5 times the initial pressure.

The theoretical maximum pressure rise can be calculated assuming adiabatic conditions and total combustion with conversion of all the oxygen present (see Westerterp et al., 1987). For an initial oxygen concentration of 8.5 vol %, the maximum pressure rise calculated is 5.3 times the initial pressure; this value is 8.6 for an oxygen concentration of 15.0 vol %.

Table A1. Maximum Pressure Rise After Ignition

P_0 [MPa]	O_2 [vol %]	$[O_2 - O_{2,0}]/O_{2,0}$	Re	P_{max}/P_0
0.5	15.0	0.44	0	1.46
	11.6	0.12	2,000	1.32
	11.6	0.12	3,500	1.38
	14.0	0.35	3,500	1.42
	11.7	0.13	5,000	1.40
1.0	12.5	0.20	7,000	1.43
	12.0	0.40	0	1.47
	14.0	0.63	0	1.46
	10.0	0.16	4,000	1.29
	9.9	0.15	7,000	1.30
1.5	10.9	0.27	10,000	1.40
	8.8	0.17	6,000	1.28
2.0	9.4	0.25	10,000	1.36
	12.0	0.79	0	1.56
2.0	12.5	0.87	0	1.79
	8.6	0.28	2,750	1.20
	8.5	0.27	5,500	1.22
	8.8	0.31	8,250	1.26

Note: $T = 298$ K.

Zabetakis (1965), Hashiguchi et al. (1966), and Crescitelli et al. (1977) performed similar experiments in highly explosive bombs or vessels and recorded pressure increases of 3.7 to 8.5 times the initial pressure.

Thus, in our installation the experimentally recorded maximum pressure rises are much smaller than in experiments in explosive bombs. The calculated pressure rise is incorrect, because complete combustion to CO_2 and H_2O does not occur, large amounts of soot are precipitated in the tube, and moreover, the combustion is not adiabatic. In a tube with a small diameter the surface-to-volume ratio is considerably higher than in explosive bombs, so much heat is taken up by the tube wall. In our tube the A/V ratio is 190.5 m^{-1} , whereas in explosive bombs, this value is 40 m^{-1} at most.

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