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COMBUSTION, EXPLOSION, AND SHOCK WAVES

Flame Propagation in Gaseous Nitrous Oxide

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Abstract—The characteristics of the propagation of a nitrous oxide decomposition flame in a tube with an internal diameter of 70 mm were measured. It was demonstrated that the pattern of flame propagation and the extent of burnout are determined by the convective motion of the flame kernel because of a very slow burning of nitrous oxide. The laminar flame speed estimated from pressure oscillograms and calculated using thermal theory of flame propagation was found to be ~1 cm/s. The critical diameter of flame quenching in channels were measured to decrease from 10 to 4 mm as the pressure was increased from 15 to 20 atm. Because of the possibility of reignition of the fresh mixture behind the flame arrester by the outflowing combustion products, the channel should be significantly longer than 200 mm.

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INTRODUCTION

Nitrous oxide is an ideal object for testing thermal theory of flame propagation, since the kinetics of its dissociation and the subsequent reactions is well known over a wide range of temperatures and pressures. What is especially important is that the reaction scheme is simple enough to assume that the propagation of the flame is largely controlled by thermal factors, without a noticeable influence of active species diffusion and chain reactions.

From the practical point of view, studying the regularities of the propagation of a nitrous oxide decomposition flame is of considerable interest, since they determine the critical conditions of combustion, knowledge of which is required to work out recommendations on explosion safety during transportation, storage, and handling of nitrous oxide.

The most important characteristics of flame propagation are the laminar flame speed and the minimum diameters of channels in which the flame still propagates. The first parameter determines the rate of pressure rise in a vessel upon nitrous oxide ignition and the character of the ignition process itself, since at low flame speeds, convection is a factor that largely determines whether the flame can be initiated by an ignitor. The second characteristic is extremely important for explosion safety, since it provides information needed for developing flame arresters. In the present work, we obtained experimental and theoretical estimates of both these characteristics.

EXPERIMENTAL

The experiments were performed in a vertical tube with an inner diameter of 70 mm and a length of 600 mm. Nitrous oxide was initiated by electrical explosion of a Constantan wire, 0.03 mm in diameter, positioned near the lower endplate of the tube. The wire was exploded by a discharge of a 16-μF capacitor. In some experiments, the mixture was initiated near the upper end plate. The time evolution of the pressure was measured with a pressure transducer; a typical pressure time history for an initial pressure of 10 atm is shown in Fig. 1. As can be seen, within first 0.4 s, the pressure changes only slightly, within the next 0.4 s, its growth rate increases to its maximum value, after which the pressure rises nearly linearly with time to its maximum value (~120 atm). At initial pressures of 5 to 15 atm, the maximum relative pressure rise in this bomb was virtually constant, 10–13, which corresponds to a complete decomposition of N_2O. At a higher initial pressure, the pressure time history was somewhat different (Fig. 2): a quasi-plateau with a very slow pressure rise appeared within the range of intense pressure rise. The pressure rise delay and quasi-plateau level, unlike the pressure rise rate at the final stage and the maximum pressure attained, were dependent of the ignition energy. A similar pressure time history (with a plateau) was also observed when nitrous oxide was initiated near the upper endplate.

Experiments with initiation in bombs of different lengths showed that there is an optimal length, dependent on the initial pressure, at which nitrous oxide decomposed completely. For example, at the initial pressure of 3 atm, this length for a 70-mm-in-diameter tube is 30 cm.

Experiments on determination of the critical diameter of flame quenching were performed using two schemes. In the first scheme (Fig. 3), the tube consisted of two sections, 300 and 600 mm in length. A

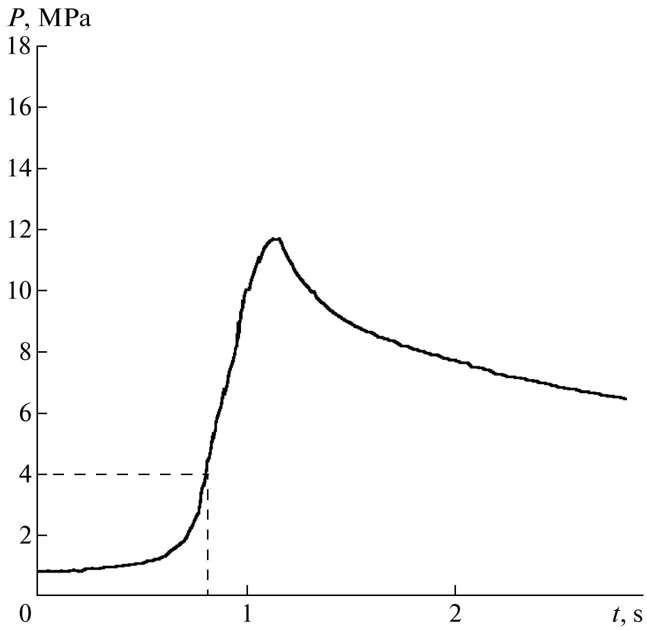

Fig. 1. Pressure oscillogram for the ignition of nitrous oxide near the lower endplate of the tube at an initial pressure of 10 atm.

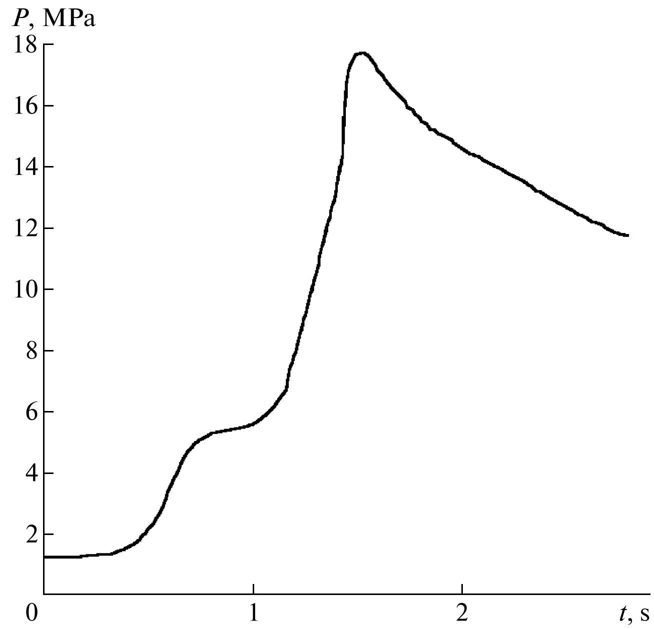

Fig. 2. Pressure oscillogram for the ignition of nitrous oxide near the lower endplate of the tube at an initial pressure of 14 atm.

block with channels of the same diameter was inserted into the longer section. Whether the flame passed through the flame arrester or not was determined from the character of pressure rise. It turned out that the outcome depended not only on the diameter of the channels but also on their length. For example, at the initial pressure of 8 atm, the flame passed through channels 8 mm in diameter and 100 mm in length but failed to do so in the case of 15-mm-in-diameter and 200-mm-in-length. Note, however, that, for channels of the same length, the result was quite expected: the flame passed through a channel with a larger diameter at a lower initial pressure.

In the second experimental scheme, a 200-mm-in-diameter metal cylinder with perforated holes of various diameters was closely attached to the upper endplate of the tube. At the end of each hole, flame indicators were placed, which changed their state under the action of high temperature and oxidative medium. This scheme made it possible to determine the critical diameter d_{cr} in one experiment. The quenching diameters measured using the second scheme were larger than those determined employing the first one. Note, however, that, when plotted in the logarithmic coordinates, the pressure dependences of this characteristic were parallel to each other, more specifically, the quenching diameter was inversely proportional to the initial pressure to the power of 1.7 (Fig. 4).

RESULTS AND DISCUSSION

Both the pressure time history and dependence of the degree of decomposition on the bomb length were found to be dependent on the flame speed, since pressure records showed that nitrous oxide combustion was a slow process, obviously subjected to a strong influence of convection. Indeed, first estimates within the framework of thermal theory yielded a very low laminar flame speed of nitrous oxide decomposition [1]. This was convincingly confirmed by experiments [2] in which pressure time histories and cinematographic records of a nitrous oxide flame propagating from the center of a spherical bomb 184 mm in diameter were obtained. At the initial pressure of 26 atm, only a twofold pressure rise was observed, with a burnout time of 1.35 s. This means that the degree of decomposition was as low as 10%, a result that can be accounted for by assuming that the volume of burning nitrous oxide rapidly lifted, reached the upper wall, and spread over it, being cooled in the process, so that the combustion extinguished some time later. Similar data on the maximum pressure rise were obtained in [3] at markedly lower initial pressures.

Note that the degree of pressure rise at high initial pressure can substantially exceed the above-mentioned values and those calculated using the equation of state of an ideal gas, since at high pressure the equation of state of nitrous oxide differs significantly from the equation of an ideal gas. Calculations performed in [4] (Fig. 5) demonstrated a considerable increase in the explosion pressure P_e to the initial pressure P_i as the latter approaches the critical point. When a cor-

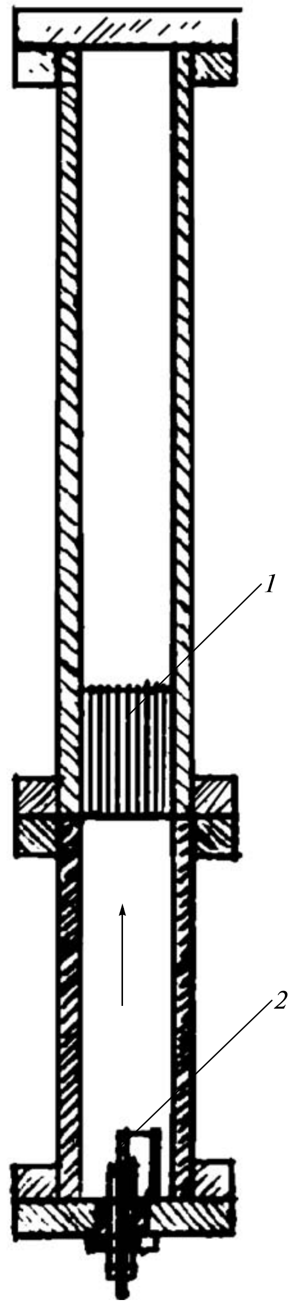

Fig. 3. Schematic of the setup for determination of the critical quenching diameter for a nitrous oxide decomposition flame: (*1*) flame arrester and (*2*) igniter.

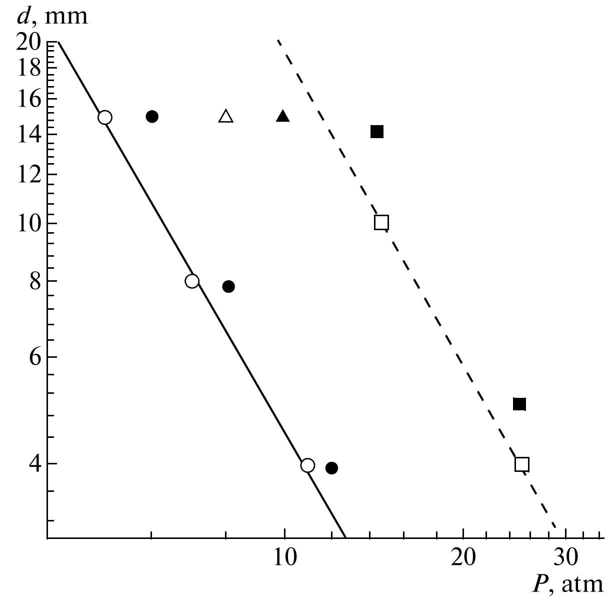

Fig. 4. Experimentally measured pressure dependences of the critical diameter for the quenching of a nitrous oxide decomposition flame passing through a flame arrester of length ℓ installed between two sections of the tube ((Δ) ℓ = 200 mm and (○) ℓ= 100 mm) and (□) a flame arrester positioned face-to-face with the upper endplate. The open and closed symbols denote flame passage and extinction, respectively.

rection for the nonideality of the state of nitrous oxide at high pressure is introduced, the extent of decomposition in spherical bombs becomes still lower.

The laminar flame speed for nitrous oxide decomposition was estimated from pressure oscillograms recorded in experiments with ignition from above. For a pressure rise range within which the flame front was expected to be planar, the laminar flame speed was determined from the pressure rise rate. At the initial pressure of 60 atm, it was found to be 1 cm/s. Estimates based on an analysis of oscillograms showed that, within the range of dynamic pressure from 50 to 130 atmospheric, the laminar flame speed is proportional to $P^{-1.25}$. The initial portions of pressure records for ignition from below also make it possible to estimate the laminar flame speed of nitrous oxide decomposition. Although an analysis of these portions yielded approximately similar results, the smallness of the pressure rise makes it impossible to ensure a satisfactory accuracy of determination of the flame speed; in addition, the convective lift of the flame kernel may substantially change the shape of the flame.

We also calculated the laminar flame speed within the framework of the thermal theory with consideration of the kinetics of nitrous oxide decomposition at high pressure. It was taken into account that the second order of the reaction of decomposition of nitrous oxide held only up to 20 atm, gradually approaching the first order at higher pressures. The calculations performed showed that, at pressures below 20 atm, the laminar flame speed is nearly constant, ~1 cm/s. At higher pressures, it must be inversely proportional to $P^{0.5}$. That the experimentally measured pressure dependence is stronger can be explained by two factors: (1) at high pressure, the gas density in the formula for the flame speed is proportional to the pressure to a power larger than unity and (2) the curvature of the flame, which was considered planar in calculations, depends on the initial pressure. In our opinion, the

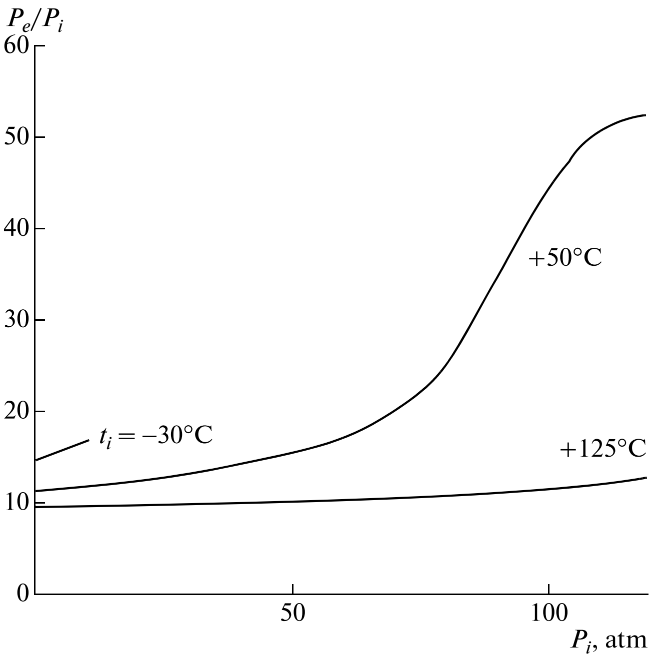

Fig. 5. Relative pressure of nitrous oxide decomposition in a closed volume with consideration given to the nonideality of the initial state of the gas; t_i is the initial temperature.

second factor is more important, since the density increase associated with the nonideality of gaseous nitrous oxide is expected to be small. Overall, estimates of the laminar flame speed of nitrous oxide decomposition as determined from pressure time histories are in satisfactory agreement with the predictions of thermal theory.

That the nitrous oxide decomposition flame propagates slowly makes the flame kernel produced by ignition from below leave the site of ignition before it reaches the tube wall. At a low initial pressure (for the run the pressure record of which is displayed in Fig. 1), convective motion manages to transport the flame kernel closely to the upper endplate rather rapidly, so that the flame surface area increases continuously, due to both the propagation of the flame toward the tube walls and the spread of hot combustion products over the endplate; therefore, oscillogram features monotonic pressure rise. At higher pressure (Fig. 2), the velocity of convective lift of the flame kernel is lower because of a higher gas density and, hence, a greater resistance to its motion (as can be seen from a comparison of Figs. 1 and 2), and, therefore, the flame reaches the tube walls earlier. Upon doing so, it becomes flatter, and the pressure rise slows down sharply. This explains why a quasi-plateau appears in the pressure record. For ignition from above, the quasi-plateau in the pressure time history begins to form when the flame touches the lateral walls of the tube and becomes flatter. The subsequent increase in the pressure rise rate is associated with the inverse curvature of the flame front caused by the cooling of the combustion products by the lateral walls of the tube and by convective flows from the tube axis to the periphery.

Due to a strong influence of convection on the propagation of the nitrous oxide decomposition flame, the problem of explosion safety of this gas is intimately related to the actual geometry of the volume it occupies. Indeed, when produced near the upper wall, the flame kernel should be large enough to forestall its quenching by the wall.

Therefore, in all experimental studies, the pressure at which the flame was capable of propagating downward was above 10 atm; i.e., it was necessary to ensure a high density of the starting hot kernel and a narrow reaction zone in the flame (which decreases with increasing pressure), so that heat losses could not extinguish the incipient flame. Ignition at a certain distance from the upper wall sometimes gives rise to a flame kernel, which rapidly reaches the upper wall, and the chance that it will spread over the entire volume is determined by whether it is large enough to survive cooling by the upper wall. When ignited at a large distance from the upper wall, the flame can spread throughout the entire volume (at a sufficient ignition energy and a high pressure), reach the upper wall and then propagate downward, or extinguish because of a too fast lift of the flame kernel accompanied by a supercritical extension of the frame. This, as pointed out in [5], explains why an optimal channel height for the complete decomposition of nitrous oxide exists. The large scatter in the values of the minimum pressure at which nitrous oxide was considered to burn in the works of different authors can also be explained by the above processes, since in many cases the extent of decomposition was small: once ignited, the flame soon extinguished.

As to the critical diameters of flame propagation in channels, the differences in their values obtained by the two different methods can be explained by the flame being reignited by the outflowing combustion products behind the flame arrester even if it extinguished while passing through the channel. This conclusion is supported by the dependence of the quenching diameter of a channel on its length. Indeed, as the length was increased to 200 mm, the quenching diameters determined by the first method approached the corresponding values measured by the second method, which involved no outflow of the combustion products into the unburnt gas.

We estimated the quenching diameter based on thermal theory of flame propagation [6] and the measured laminar flame speed of nitrous oxide decomposition. At a pressure of 10 atm, d_{cr} was 1.8 cm, in close agreement with the measured value. According to thermal theory, the quenching diameter should be inversely proportional to the pressure at less than 20 atm and to the square root of the pressure at higher values. In all probability, the reason why the experimental and theoretical dependences deviate from each other is the same as that underlying the stronger decrease of the laminar flame speed with increasing pressure compared to the theoretical predictions, more specifically, the pressure dependence of the flame front curvature. Indeed, the behavior of the experimentally measured laminar flame speed suggests that, with increasing pressure, the flame front becomes flatter (because of a slower convective motion) within the pressure oscillogram portion used to calculate the laminar flame speed, and, therefore, the pressure rise in a closed vessel slows down. The relative heat losses for a less curved flame are lower, and, consequently, the flame can propagate in a narrower channel, as observed in experiment.

CONCLUSIONS

The experiments performed revealed a complicated pattern of the propagation of nitrous oxide decomposition flame due to a strong influence of convection on the flame kernel motion and its shape. According to experimental data and theoretical estimates, the laminar flame speed for nitrous oxide is as low as 1 cm/s, and, therefore, convective flows should strongly affect the characteristics of the nitrous oxide decomposition flame. In this case, in contrast to fast-burning mixtures, the explosion safety of nitrous oxide is determined not only by the characteristics of the gas itself, but also by the parameters of the setup on which its explosibility is tested, since the convective motion of the flame kernel and the very thick reaction zone make an incipient flame vulnerable to extinction on the walls. For this reason, when ignited with a powerful source, nitrous oxide can decompose only to a small extent, a behavior observed in all experiments on its combustion.

While the simple mechanism of nitrous oxide decomposition makes it possible to apply thermal theory of flame propagation and obtain satisfactory agreement with experiment in laminar flame speed and critical quenching diameter at fixed initial conditions, the measured pressure dependences of these two characteristics differ significantly from those predicted by the theory. It makes sense to explain this discrepancy by the flame shape being different in experiment and calculations; generally, the flame shape should depend on the initial pressure, since the convective motion pattern should depend on the gas density.

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