

Heat and Mass Transfer

Nickolai M. Rubtsov

The Modes of Gaseous Combustion



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The author dedicates this book to the memory of Doctor of Sci. Professor Ideya M. Naboko, the remarkable coauthor and the generator of plans which did not manage to be carried out completely.

Ideya M. Naboko was a well-known expert in the area of physical gas dynamics, combustion, explosion, and detonation of gases, methods of investigation of fast processes. Her works, especially in the field of non-stationary jet currents, are used in adjacent areas of science for the solution of applied problems. For these achievements, Ideya M. Naboko in 1986 received an award by the USSR Council of Ministers.

Since 1994, Ideya M. Naboko has applied her experience and knowledge to the study of nonstationary modes of combustion and explosion of gases in volumes of unconventional geometric. Her results are stated in more than 160 scientific works and were continually represented at Russian and international conferences. Ideya M. Naboko was a member of the organizing committee of the annual Academic Space Conferences of Federal Space Agency. Nine

people from her wards defended Ph.D. theses, and later four became doctors of science.

Ideya M. Naboko was a vigorous and not indifferent person and took all the latest events in the country and, especially, related to the Russian Academy of Sciences very much to heart.

Preface

The book is aimed at the analysis of contemporary problems in combustion science, namely flame propagation, detonation, and heterophaseous combustion based on the works of the author; from a certain viewpoint, it is an auto survey. In this book, some modern problems in the area of gas combustion, as well as methods allowing to calculate and estimate limiting conditions of ignition, and flame propagation on the basis of experimental results obtained substantially by the author of the book are considered. The book may be useful for experienced students and qualified scientists in the area of experimental studies of combustion processes.

An approximate analytical approach for estimation of the effectiveness of the influence of inhibitor additives on flame velocity and flame propagation limits was suggested for combustion of rich H_2 + air mixtures. The method is based on the model of a narrow reaction zone and takes into account peculiarities of the branched chain mechanism of H_2 oxidation. It is shown that the occurrence of flame propagation limits depending on the amount of inhibitor is caused by a positive feedback between flame velocity and the amount of active centers of combustion, being terminated *via* an inhibitor. According to the feedback, the influence of an inhibitor leads to lowering of combustion temperature and flame velocity as well. The method is proposed for the analysis of experimental data on the limits of flame propagation in hydrogen–air mixtures at atmospheric pressure in the presence of small quantities of active additives (inhibitors). The mechanism of the occurrence of an upper concentration limit of flame propagation at atmospheric pressure taking into account effective heat losses in the termolecular recombination $\text{H} + \text{O}_2 + \text{M} \rightarrow \text{HO}_2 + \text{M}$ is suggested for combustion of an H_2 + air mixture.

Regularities of formation of spherical flames in stoichiometric mixtures of natural gas and isobutylene (iso- C_4H_8) with oxygen and Kr or CO_2 additives in the constant volume reactor were established by means of color speed cinematography at 100 Torr and 298 K. The influence of additives of CO_2 , Ar, propene, and CCl_4 on initial stages and dynamics of flame front formation and the structure of laminar spherical flames in hydrogen–air, methane–air, and n-pentane–air mixtures were studied in a bomb of constant volume by means of color high-speed digital

cinematography at atmospheric pressure. Numerical investigation into specific surface effects in flame propagation of lean and rich laminar hydrogen flames at different wall boundary conditions and fuel/air ratios was performed by means of two-dimensional simulations. It was experimentally shown that the same lean ($H_2 < 10\%$) hydrogen–air mixture can be repeatedly ignited. Numerical simulation based on Boussinesq approximation was shown to be in qualitative agreement with the observed features of combustion. Features of spatial development of thermal ignition (so-called self-ignition or spontaneous ignition) in chain oxidation of hydrogen, natural gas, and isobutene with oxygen in the pressure range of 10–100 Torr and 750–1000 K have been revealed by means of high-speed color cinematography. It was shown that the features could be controlled by addition of chemical active gaseous additives (promoters and inhibitors). It is shown that the features of spatial development of spontaneous ignition of propane–air and n-pentane–air mixtures depend on the state of reactor surface, namely an ignition initial center originates on the reactor surface, then the flame front of the center propagates into volume with the normal velocity corresponding to the reactor walls temperature and gas mixture composition. The ignition of n-pentane–air mixture at low temperatures was experimentally studied in a rapid mixture injection static reactor. The ignition process was monitored using a high-speed color video camera. It is shown that introduction of platinum wire into the reactor eliminates the phenomenon of a negative temperature coefficient; however, Pt wire has no effect on the ignition delay time of thermal ignition of stoichiometric n-pentane–air mixture at lower temperatures. Spatial development of chain ignition in hydrogen–air mixtures in the vicinity of the third combustion pressure limit has been investigated by means of quick gas transfer with the use of high-speed color cinematography. It was shown that spatial development of ignition is determined by material and state of the reactor surface; a primary ignition center always occurs at reactor surface.

The approximate analytical method was applied for analysis of the problem on a local chain-thermal explosion in reaction of hydrogen oxidation in the presence of chemically active additive. It is shown that key parameters defining the critical size of a local source of ignition are the temperature in the center of local ignition zone; the quantity of the active centers of combustion created with the local source; and presence of active chemical additives in combustible gas mixture. Comparison to experimental data has shown applicability of the developed approach for the analysis of critical conditions of local ignition in combustible gas mixtures. It is experimentally revealed that the methane combustion inhibitor CCl_4 shows no effect on the lower ignition limit of hydrogen combustion. It is established that small amounts ($\sim 10^{-1}\%$) of chromium hex carbonyl promote combustion of $2H_2 + O_2$ mixture, which manifests itself in the increase in the propagation velocity of the flame, thus inhibition of oxidation of isobutene by this additive takes place. Therefore, the role of hydrogen atoms in hydrocarbon oxidation is not significant and may result at least in participating in longer reaction chains than in hydrogen oxidation. This means that the kinetic mechanism of inhibiting combustion of hydrocarbons by carbonyls suggested in the literature based on accounting for termination of hydrogen atoms should be refined.

By means of high-speed color cinematography, it was shown that the flames in lean H_2 -air mixtures at an initial stage propagate symmetrically and the flame radius can be estimated from the frames of speed filming. It is shown that sufficiently strict calculation of cellular structure of the flame front of lean hydrogen mixes requires consideration of a 3D problem, however, thermal diffusion instabilities at the initial stage of combustion have no effect on the velocity of flame which can be estimated assuming unperturbed flame front in the range of $8\% < H_2 < 15\%$. It was shown that the analysis of experimental data on flame propagation in lean mixtures does not allow taking apart the results of calculation by two-dimensional model with regard to convection and without convection. It was experimentally shown that isobutene additives in quantities below a concentration limit (up to 1.5 %) tend to increase, and CO_2 additives up to 15 %—to reduce the flame propagation velocity in lean H_2 -air mixtures. The reasons for the acceleration of combustion in the presence of hydrocarbon additive are considered. The method of high-speed cinematography was used in investigation of transition of spherical flame front to flat front in n-pentane-air and methane-air mixtures initiated by a spark discharge. Cellular flame structures were observed in the transition. Modeling based on compressible reactive Navier-Stokes equations at low Mach number showed qualitative agreement with experiment. Features of combustion in flame cells caused by hydrodynamic instability are experimentally established. It was shown that each flame cell represents a separate "chemical reactor"; in the cell, the process of complete chemical transformation occurs. It was shown that inhomogeneities detected in light emission that arise after contact of a flame front with the walls of cylindrical reactor can be correlated with the occurrence of acoustic waves by the example of combustion of hydrogen-air mixtures containing 30 and 15 % of hydrogen. It was revealed that flame velocities in stoichiometric hydrogen-air mixtures at central spark initiation do not depend on the material of inner reactor surface but on its shape.

It was shown that spark initiated flames of hydrogen-air mixtures (8–15 % H_2) pass through the close-meshed aluminum spherical obstacles of cell size $0.04\text{--}0.1\text{ mm}^2$; the flame of 15 % H_2 in the air after obstacle is accelerated; acoustic gas fluctuations occur in the reactor. The flame of 8 % natural gas-air mixture is not accelerated after obstacle; acoustic fluctuations are missing. It was shown that active centers of methane and hydrogen combustion, determining flame propagation, have different chemical nature. It was shown that spark initiated flames of diluted stoichiometric natural gas-oxygen mixtures in close-meshed aluminum spheres of mesh size $0.1\text{--}0.2\text{ mm}^2$ do not propagate through the spheres, but always propagate through planar meshed obstacles of the same mesh size. It was found that the features of flame propagation at simultaneous initiation at opposite butt-ends of the cylindrical reactor differ markedly from those at initiation from a single discharge. It is shown that the increase in warming up in hydrocarbons combustion at simultaneous initiation at opposite butt-ends of a cylindrical reactor by a factor of ~ 2 as compared to flame propagation from a single initiation source is due to a two-stage nature of the combustion process. It was shown that ignition of diluted methane-oxygen mix (total pressure up to 200 Torr) after a single obstacle can be

observed markedly far from an obstacle surface. The use of the meshed sphere as an obstacle leads to increase in the distance of flame emergence behind an obstacle in comparison with a round opening; two or more close-meshed obstacles strongly suppress flame propagation. It is experimentally shown that under the same conditions the limit of penetration of diluted methane oxygen flame through a confuser is markedly less than in the case of a plain orifice and is even less than in the case of a diffuser. Therefore, the diffuser is the most effective flame arrester.

It is illustrated that one-dimensional Zeldovich–von Neumann–Doering model of detonation wave gives a satisfactory approach for the description of a stationary detonation wave (DW), despite a large number of the approximations made at the derivation of the equations of the theory. Besides, according to modern literary data on numerical modeling, the neglect of transverse structure of detonation wave in one-dimensional model has no influence on the pressure value in the front of DW in comparison with multidimensional models. It is experimentally demonstrated that the acoustic resonator (Helmholtz's resonator) connected to a cylindrical reactor can cause reactor destruction at spark initiation of deflagration in lean (15 %) hydrogen mixture with oxygen. This points to a possibility of transition of deflagration to supersonic regime mode near the lower concentration limit of detonation even for small reactor where detonation is obviously impossible. On the basis of Zeldovich–von Neumann–Doering detonation theory with allowance for the theory of chain processes by the example of the oxidation of hydrogen-rich mixtures in the presence of chemically active additive (inhibitor), it is shown that taking into account reactions of inhibitor with chain carrier leads to "chemical" losses in addition to heat losses.

A heterophaseous branching-chain reaction of dichlorosilane oxidation is considered in terms of unsteady nucleation theory taking into account the condensation growth of nuclei. The transition between the homogeneous and heterogeneous phase formation; the dependence of the amount of aerosol on the initial temperature, pressure, and mixture concentration; both the kinetics of phase formation and of the disappearance of initial substance are qualitatively described. It is shown that small inhibitor (propene) additives reduce the mean size of aerosol particles. This result is also in qualitative agreement with experimental data. It is found that the amount of aerosol formed decreases with increasing initial temperature because of the increase in equilibrium vapor pressure of the new phase. The reactivity of the silicon dioxide aerosol obtained in the presence of Freon-12 is shown to vary reversibly. The role of surface diffusion and surface nucleation in the deposition of thin films is discussed. We have experimentally observed both the stable liquid and solid Coulomb crystals formed in the discharge-initiated combustion reaction between dichlorosilane and oxygen and the growth dynamics of formation of a rotating solid Coulomb crystal at the interface between the void and dusty cloud. It was shown that the gases evolving during thermal annealing of coal powder have an inhibiting effect on the ignition and combustion of hybrid gas suspension, containing natural gas. Investigation into flammability in oxygen of various types of coal with various content of volatiles at a total pressure of 85 Torr and initial temperatures in the range of 650–750 °C has been performed. It is shown that

ignition of separate particles of coal powder occurs right after injection of powder with oxygen without gas combustion. Then after an ignition delay the volatiles containing in a coal powder ignite, thus the ignition propagates over reactor volume. The more volatiles the coal powder contains the more intensive ignition is observed. It is shown that hybrid powder gas mixture PGM consisting of soot or graphite powders which do not contain volatiles, and a stoichiometric mixture of natural gas and oxygen, intensively ignite in conditions in which hybrid PGM, consisting of coal powder, and a stoichiometric mixture of natural gas and oxygen, does not ignite. It is shown that unlike graphite, soot powder promotes ignition of a stoichiometric mixture of natural gas and oxygen. Possible scenarios of ignitions occurrence in mines were analyzed. It is shown that hybrid PGM, consisting of soot powder and the stoichiometric mixture of natural gas and oxygen, intensively ignites in the absence of $C_{10}F_{18}$, however, in the presence of $C_{10}F_{18}$ combustion of natural gas is missing, the ignition of separate particles of soot powder is only observed.

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Introduction

There are three things you can watch forever: flame, water, and other people working. In this book, I will speak about different regimes of flame propagation and I will try to tell about the main factors influencing the flames. A tremendous variety of the modes of combustion registered by means of color high-speed filming presented in the book seems very surprising even for the specialist.

A flame does not arise spontaneously. We use a lighter to turn on the gas stove, or a match to kindle a fire. Many years ago, our ancestors used flint, or a lightning, irrespective of the person, lit combustible subjects. Anyway, without an ignition source a process of combustion will not occur. It concerns also an internal combustion engine and a fighting shell, and any devices using combustion. These are mines for coal mining and other minerals, pipelines and gas highways, and premises. So broad application and value of combustion in our life demands knowledge of the regularities of this phenomenon; the ignition on which it first depends whether combustion occurs or not. If ignition happens, flame propagation in gas mixture takes place. In certain conditions, the flame can be accelerated and there will be detonation—the complex consisting of the shock wave fed by the energy, which is given by the combustion process, and a combustion zone, which extends with supersonic speed. It is obvious that such processes leading to destruction of industrial objects, premises, and death of people have to be excluded. Emergence of combustion has to be controlled strictly; there must be reliable methods of prevention of combustion.

Methods of suppression of combustion can be divided as chemical ones with use of active additives that terminate reaction chains (it is well known that all gas combustion processes have branched chain nature), and physical ones (fire prevention devices). The ways of suppression by means of foamy and aerosol structures are intermediate, as the substances entered into a flame zone are usually chemically active. The active additives suppressing combustion (inhibitors) despite the seeming simplicity of their use, for example, mixing with combustible gas in advance, possess many shortcomings: inhibitors of hydrogen combustion, such as unsaturated hydrocarbons, are flammable and besides these are effective only in stoichiometric and rich mixes. As is shown below, in lean mixes these inhibitors, on

the contrary, accelerate burning. Carbonyls of metals are poisonous and cannot be used in the presence of human personnel. Safe (having high occupational exposure limit, OEL) freons are less effective, and, besides, at emergency ignition lead to the formation of very toxic HF and COF₂. Physical methods as barriers and grids can also serve along with overwhelming action as turbulizers of combustion and in certain conditions lead to acceleration of the front of combustion with undesirable consequences.

On the other hand, in many combustion devices, ensuring completeness of combustion (diesel engines) or combustion acceleration (the detonation engine) is required. It is achieved with the use of nozzles, turbulizers, and other obstacles providing flame acceleration. It is known that the majority of reactions of gaseous combustion is followed by formation of a solid disperse phase. Thus, the solid phase can arise directly in the course of combustion, or at oxidation of an active additive, and can also be present in advance in the gaseous mixture in the form of a disperse phase (for example, when using aerosol fire extinguishers). Industrially important processes of oxidation of hydrocarbons (e.g., synthesis of nanotubes) and silanes are combustion processes that are followed by the formation of a new phase. Oxidation of silanes is followed by formation of silicon dioxide and finds application in microelectronics as isolating coatings and as active elements in field-effect transistors. It indicates the importance of research of combustion of heterophase systems, gas–solid.

The problems listed above, which are reduced to providing complete control of process of combustion of the set combustible substances in the set conditions are those actual problems that researchers have been solving in combustion science up to now.

It is impossible to overestimate a huge contribution of the Soviet school of combustion to the problems considered in this book. We stand on the shoulders of giants such as academicians Ya.B. Zeldovich, N.N. Semenov, A.G. Merzhanov, Profs. D.A. Frank-Kamenetsky, O.M. Todes, A.S. Sokolik, etc. In their works, they developed theories of thermal explosion, local ignition, focal ignition, and flame propagation. These theories are written by “wide dabs” and leave still considerable scope for specification of the physical description of many phenomena occurring in the combustion of gaseous and solid systems.

Note that at a theory statement we tried to pay main attention not to numerical calculations, but whenever possible to analytical consideration. It is also related to the way that the development of an analytical approach is always followed by the creation of some ideology of understanding of essence of the process using certain reasonable assumptions, which allow getting the solution of a complex problem in a rather simple and clear analytical form. Thus, the reader has an opportunity to understand the nature of the phenomenon, without penetrating into a particular architecture of algorithms of a calculation method.

The author would also like to make the reader more familiar with virtually inaccessible works of Russian authors.

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