

## HEAT AND MASS TRANSFER IN COMBUSTION PROCESSES

ASSESSMENT AND CONTROL OF DETONATION HAZARD  
OF SILANE-CONTAINING MIXTURESP. A. Fomin,<sup>a,b</sup> A. V. Fedorov,<sup>b</sup> D. A. Tropin,<sup>b</sup>  
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UDC 534.222

*A formula for calculating the induction period of a silane–air gas mixture has been proposed. The dimension of a detonation cell and the energy of direct initiation of gaseous detonation were assessed. Consideration has been given to the issue of control of the parameters of detonation of silane-containing mixtures. The parameters of Chapman–Jouguet detonation, the relative dimension of the cell of a detonation wave, and also the parameters of explosion at constant pressure and volume in a stoichiometric silane–air gas mixture with additions of chemically inert microparticles ( $Al_2O_3$ ) have been calculated.*

*A series of experiments was conducted on measurement of the pressure profile resulting from the explosion of the silane–air mixture in a volume of cubic shape with the known average fuel–oxidant ratio. The distribution of the fuel during the silane jet flowing out into this volume was visualized. The distribution of silane in a cloud in its outflow into the cubically shaped volume and in a high-speed jet flowing out into an unbounded space was calculated. The performed investigations can be useful in evaluating the comparative efficiency of explosion of clouds of silane– and hydrocarbon–air mixtures.*

**Keywords:** silane, induction period, detonation, cell, explosion, gas–particles mixture, explosion safety.

**Introduction.** Gaseous silane ( $SiH_4$ ) is widely used in the semiconductor and photochemical industries. Its capacity for self-ignition on contact with air even at room temperature is a source of constant explosion hazard [1]. This underlines the importance of investigating the parameters of explosion and detonation of silane–air mixtures for the solution of relevant problems of fire and explosion safety.

The dimension of a gaseous-detonation cell is one basic characteristic of detonation hazard of gas mixtures [2, 3]. In particular, it is by the cell's dimension that the geometric (on the tube diameter) limits of propagation of a detonation wave (DW), the energy of direct DW initiation, the possibility of the detonation going out successfully of the tube into the volume, etc. are determined. Unfortunately, the dimension of the cell and the quantity of the energy of direct initiation of detonation in silane–air gas mixtures of various stoichiometries has been neither measured experimentally nor calculated theoretically to date, which makes it difficult to assess the detonation hazard of such mixtures. One possibility of evaluating the dimension of a DW cell in silane-containing mixtures is to use the widely known Vasil'ev–Nikolaev model [4]. Within its framework, the cell dimension can be evaluated from the algebraic formula if the parameters of the Chapman–Jouguet wave and constants of the Arrhenius formula for calculation of the induction period of a chemical reaction behind the shock wave front are known. Calculation of the parameters of the Chapman–Jouguet wave is not a fundamental problem. Accordingly, to evaluate the dimension of a detonation cell in the mixtures in question, it is necessary to obtain an algebraic formula for calculation of the induction period of a chemical reaction in silane-containing gas mixtures at detonation pressures and temperatures.

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It is common knowledge that additions of chemically inert particles are an efficient method to monitor and control the processes of combustion and detonation of gas mixtures. Such additions decrease the pressure and temperature of explosion and hence lower thermal and mechanical load on industrial structures during emergency emissions and subsequent explosions of chemically active gases. The loss of DW energy by particle heating and acceleration may result not only in the reduction in the pressure, temperature, and rate of detonation, but also in the suppression of a DW [5–8]. At the same time, the influence of chemically inert additives on the parameters of explosive processes in silane–air mixtures has not been investigated either experimentally or theoretically to date. The parameters of instantaneous explosion of gases and mixtures of gases with chemically inert particles can be used to evaluate thermal and mechanical loads on solid surfaces of residential buildings and industrial structures (explosion at constant volume) and to evaluate the adiabatic temperature of combustion of a chemically active mixture (explosion at constant pressure). Such calculations of silane–air gas mixtures with additions of chemically inert particles are not presented in the available literature.

In the present work, we have proposed a formula for calculation of the induction period of a silane–air gas mixture of various stoichiometry at high pressures and temperatures and have made an estimate of the cell's dimension and the energy of direct detonation initiation. Consideration has been given to the issue of control of explosive processes in silane-containing mixtures: the Chapman–Jouguet parameters and the relative dimension of a DW cell, and also the parameters of explosion at constant pressure and volume in a stoichiometric silane–air mixture with additions of chemically inert microparticles ( $\text{Al}_2\text{O}_3$ ), have been calculated. Furthermore, a series of experimental and theoretical investigations associated with the assessment of the comparative efficiency of explosion of clouds of silane– and hydrocarbon–air mixtures has been presented.

**Modeling of Ignition of Silane.** The model of detailed kinetics of oxidation of silane [9] takes account of 140 chemical reactions (70 forward and 70 back) for 23 components. Among the drawbacks of this model can be, e.g., neglect of the reaction of formation of  $\text{SiO}_2$  in a solid state. The gaseous components  $\text{SiO}_2$  in the model is produced in elementary gas-phase reactions, and its enthalpy is taken equal to the enthalpy of  $\text{SiO}_2$  in a solid state. Nonetheless, the given model made it possible to describe experimental times of delay of the ignition of silane, hydrogen, oxygen, and nitrogen mixtures in reflected shock waves in the range of temperatures from 750 to 1050 K. The ignition-delay time was defined as the time it takes the temperature to increase by 5%. In [9], a study was also made of the ignition of a stoichiometric silane–oxygen mixture at low temperatures (of the order of 400 K). Dependences of the ignition-delay times on the mixture's pressure (in the range of pressures from 20 to 100 kPa) were obtained at three values of temperature: 373, 393, and 413 K. A sharp increase in the ignition-delay times (from  $10^{-4}$  to 10 s) at a pressure of 40 kPa was shown.

The ignition of silane at room temperatures was modeled in [10]. The kinetics model based on the model of [9] and containing 196 chemical reactions (98 forward and 98 back) was proposed. However, no consideration was given to the formation of  $\text{SiO}_2$  in a solid state. It has been shown that the existence of the ignition of silane at low temperatures depends on the ratio of the amount of the fuel to the amount of the oxidant, and if this ratio is large ( $[\text{SiH}_4]/[\text{O}_2] \geq 15$ ) the mixture is unstable, and ignition may occur. Furthermore, it has been obtained that a great influence on the ignition of silane at room temperatures is exerted by the presence of a steam in the mixture, and if the mixture contains no steam, we do not have ignition at all.

In [11], 201 reactions for 69 components can be counted in the model of kinetics of silane; complex silicon compounds (molecules containing several atoms of silicon) can also be formed. The time of reaching the OH maximum in ignitions behind shock waves in hydrogen–oxygen, silane–oxygen, and silane–hydrogen–oxygen mixtures was calculated. Good agreement between calculated and experimental data was only obtained for the hydrogen–oxygen mixture. For all the remaining mixtures, there is a considerable disagreement between calculations and experiment. The ignition of silane was modeled at low temperatures (375–450 K) and pressures from 0.01 to 10 atm. Dependences of the ignition-delay time on pressure at different temperatures were obtained. It has been established that these times are much (up to five orders of magnitude) larger than those obtained in [9].

Within the framework of our work, use was made of the least cumbersome kinetic scheme of oxidation of silane [9], with which detonation phenomena in silane-containing mixtures were described and on whose basis an algebraic formula has been obtained for evaluating the induction period.

**Model of Nonequilibrium Gas Dynamics of Silane Mixtures.** Consideration will be given to a shock tube filled with silane, oxygen, and nitrogen/argon mixture. Let a shock wave (SW) propagate over the mixture. At fairly large values of the SW Mach number, the parameters of the mixture may exceed critical values, and ignition will occur.

In a one-dimensional nonstationary formulation, the motion of the mixture is described by equations of nonequilibrium gas dynamics:

$$\begin{aligned}\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} &= 0, \\ \frac{\partial(\rho u)}{\partial t} + \frac{\partial(\rho u^2 + p)}{\partial x} &= 0, \\ \frac{\partial(\rho E)}{\partial t} + \frac{\partial[(\rho E + p)u]}{\partial x} &= 0,\end{aligned}\quad (1)$$

where the total energy is equal to

$$E = e + \frac{u^2}{2}.$$

The internal energy of a chemically active mixture is determined by the equation

$$e = c_v(T)T + \sum_{\alpha=1}^N \xi_{\alpha} h_{0,\alpha} - c_p(T)T_{00},$$

where  $c_v(T) = \sum_{\alpha=1}^N c_{v,\alpha}(T)\xi_{\alpha}$ . Data on the dependences of the heat capacity on temperature will be taken from [12, 13].

System (1) supplemented with the equation of state

$$p = \rho TR \sum_{\alpha=1}^N \frac{\xi_{\alpha}}{M_{\alpha}} \quad (2)$$

and with the kinetic equations of detailed chemical kinetics

$$\frac{d\xi_{\alpha}}{dt} = \frac{1}{\rho} M_{\alpha} \sum_{r=1}^l \rho^{m,r} (v'_{\alpha,r} - v_{\alpha,r}) \left[ k_r \prod_{\delta=1}^N \left( \frac{\xi_{\delta}}{M_{\delta}} \right)^{v_{\delta,r}} - k'_r \prod_{\delta=1}^N \left( \frac{\xi_{\delta}}{M_{\delta}} \right)^{v'_{\delta,r}} \right], \quad (3)$$

enables us, after formulating the relevant initial boundary-value problem, to calculate the pattern of SW propagation in a channel filled with this working mixture. The given model (1)–(3) will be called nonstationary.

**Kinetics of Combustion of a Silane-Containing Mixture.** Consideration will be given to a problem on ignition of silane mixed with air in a reflected SW near the end of the shock tube. Use will be made of a detailed kinetic scheme from [9], which takes account of 140 forward and back reactions of 25 components ( $\text{H}_2$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{OH}$ ,  $\text{O}$ ,  $\text{H}$ ,  $\text{HO}_2$ ,  $\text{H}_2\text{O}_2$ ,  $\text{SiH}_4$ ,  $\text{SiH}_3$ ,  $\text{SiH}_2$ ,  $\text{HSiO}$ ,  $\text{SiH}_2\text{O}$ ,  $\text{SiH}_3\text{O}$ ,  $\text{SiH}_3\text{O}_2$ ,  $x\text{SiH}_3\text{O}_2$ ,  $\text{SiH}_3\text{OH}$ ,  $\text{SiH}_3\text{O}_2\text{H}$ ,  $\text{SiH}_2\text{OH}$ ,  $\text{HSiOOH}$ ,  $\text{SiOOH}$ ,  $\text{SiO}$ ,  $\text{SiO}_2$ ,  $\text{N}_2$ , and  $\text{Ar}$ ). Study [9] gives the rate constants of forward and back reactions. The updated information on activation energies of certain reactions ( $\text{SiH}_3\text{O}_2 \rightarrow \text{SiH}_2\text{O} + \text{OH}$ ,  $\text{HSiOOH} + \text{O}_2 \rightarrow \text{SiOOH} + \text{HO}_2$ , and  $\text{HSiOOH} \rightarrow \text{SiOOH} + \text{H}$ ) will be borrowed from [10] and the enthalpies of formation of components, from [12]. As noted above, in the model of [9],  $\text{SiO}_2$  was only considered in a gaseous phase, but with the formation enthalpy corresponding to a solid phase. However, in the DW structure itself where temperatures are fairly high (2000–3000 K),  $\text{SiO}_2$  is in a gaseous state. Therefore, account will be taken of the formation of this component with an enthalpy taken for the gas phase.

**Results of Numerical Calculations of the Ignition-Delay Time. Verification of the Kinetic Scheme.** Consideration will be given to the ignition of the mixture behind the front of a reflected SW. The flow behind a reflected SW is known to be in a quiescent state. Therefore, from system (1), we only use the energy equation, which is transformed to a form describing the change in the temperature of the gas mixture:

$$\frac{dT}{dt} = - \frac{1}{c_v(T)} \sum_{\alpha=1}^N (c_{v,\alpha}(T)T + h_{0,\alpha} - c_p(T)T_{00}) \frac{d\xi_{\alpha}}{dt}. \quad (4)$$

Thus, the process of ignition of the mixture behind a reflected SW is described by the system of equations (2)–(4) and initial data at  $t = 0$ :  $u = 0$ ,  $p = p_r$ ,  $T = T_r$ , and  $\xi_{\alpha} = \xi_{\alpha,0}$ .

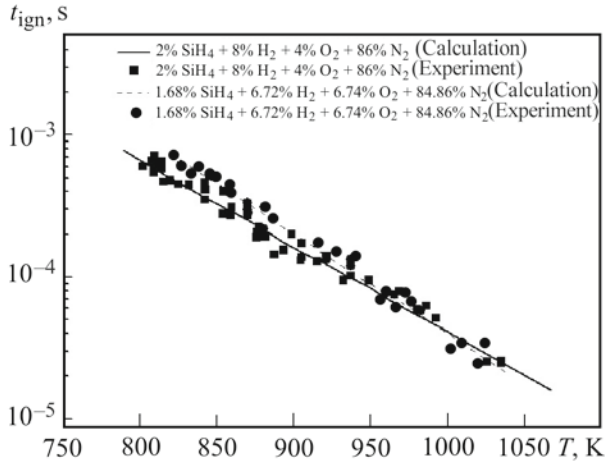


Fig. 1. Delay time of ignition of silane vs. temperature behind the reflected SW. Comparison with experiment [14].

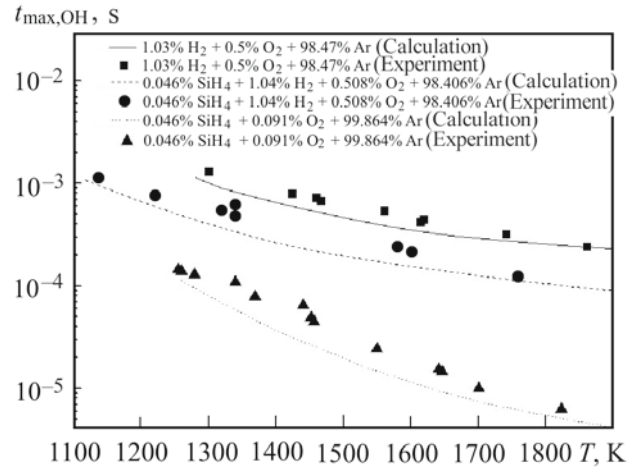


Fig. 2. Time of reaching the OH maximum vs. temperature behind the reflected SW. Comparison with experiment [15].

We consider the following criteria of determination of the ignition-delay time.

*Criterion 1.* The ignition delay corresponds to the time during which the maximum rate of growth in the temperature is attained. Figure 1 gives the dependences of the time of delay of the ignition of a silane–hydrogen–oxygen–nitrogen mixture on the temperature behind the reflected SW, calculated from the given model and obtained in experiment [14]. Mixture 1:  $\xi_{\text{SiH}_4} = 2.45 \cdot 10^{-2}$ ,  $\xi_{\text{H}_2} = 6.12 \cdot 10^{-3}$ ,  $\xi_{\text{O}_2} = 4.9 \cdot 10^{-2}$ ,  $\xi_{\text{N}_2} = 0.92038$ ,  $p_r = 1.26$  atm. Mixture 2:  $\xi_{\text{SiH}_4} = 2.02 \cdot 10^{-2}$ ,  $\xi_{\text{H}_2} = 5.055 \cdot 10^{-3}$ ,  $\xi_{\text{O}_2} = 8.11 \cdot 10^{-2}$ ,  $\xi_{\text{N}_2} = 0.893645$ , and  $p_r = 1.36$  atm. It can be seen that the given model well describes, from ignition criterion 1, ignition-delay times obtained experimentally.

*Criterion 2.* In [9], the ignition delay was associated with the time during which the mixture's temperature grew by 5% of the total temperature increase:  $T_{\text{ign}} = 0.05(T_e - T_r)$  (criterion 2). It has been shown that with the ignition delay defined in this manner, the calculated data are in good agreement with experiment.

*Criterion 3.* The time during which the maximum concentration of the radical OH is attained can also serve as one possible ignition criterion. Figure 2 gives the calculated dependences of the ignition-delay time on the temperature behind a reflected SW and experimental data [15] on the time of reaching the maximum excited radical OH\*. As can be seen, the calculated curves lie just below experimental points. Conceivably this might be due to the fact that it is the time of reaching the concentration maximum by the component of the gas mixture in different states, i.e., excited and unexcited, that is determined in calculations and experiments. It should be noted that similar calculations were carried out in [11] (the time of reaching the OH maximum was investigated), but for a more complicated model of detailed kinetics: 69 components and 201 reactions. Satisfactory agreement with experimental data was only obtained for the hydrogen–oxygen–argon mixture. For the remaining mixtures, a considerable disagreement with experiment (up to two orders of magnitude) was obtained. Thus, the comparatively simple model of chemical kinetics [9] is more adequate than [11].

**Mathematical Model of Calculation of the Parameters and Structure of a Detonation Wave.** Consideration will be given to the problem on the structure and parameters of a DW in a silane–air mixture. We will consider the process of DW propagation in a shock tube as stationary and pass to a new spatial variable related to the DW front  $\zeta = x - Dt$ , where  $D$  is the DW velocity. Then system (1) will be reduced to three algebraic conservation law:

$$\begin{aligned} \rho U &= \rho_0 U_0 = c_1, \\ p + c_1 U &= p_0 + c_1 U_0 = c_2, \\ e + \frac{p}{\rho} + \frac{U^2}{2} &= e + \frac{p_0}{\rho_0} + \frac{U_0^2}{2} = c_3, \quad U = u - D. \end{aligned} \quad (5)$$

We close this system by the equation for the mixture's velocity

$$\frac{dU}{d\zeta} = \frac{\frac{R}{c_v(T)M_{\text{mix}}} \sum_{\alpha=1}^N \frac{d\xi_{\alpha}}{dt} (c_{v,\alpha}(T)T + h_{0,\alpha} - c_{p,\alpha}(T)T_{00}) - RT \sum_{\alpha=1}^N \frac{1}{M_{\alpha}} \frac{d\xi_{\alpha}}{dt}}{U^2 - c_f^2}. \quad (6)$$

Supplementing Eqs. (5) and (6) with the chemical-kinetics equations (3) with account of the relation  $\frac{d}{dt} = (u - D) \frac{d}{d\zeta}$ , we obtain a system of ordinary differential equations for which the Cauchy problem with initial conditions for the mass concentrations  $\xi_{\alpha}|_{\zeta=0} = \xi_{\alpha,0}$  and the sought initial condition for the velocity  $u|_{\zeta=0} = u_f$  is posed.

**Formula for Calculation of the Delay Time of Ignition of Silane.** Calculations with detailed kinetic mechanisms (as has been done above) can sometimes be burdensome from the viewpoint of computational cost, particularly for two-dimensional and three-dimensional nonstationary flows. Therefore, it is expedient to construct a simple kinetic model that would satisfactorily describe the processes of chemical transformations of silane behind transmitted and reflected SWs in subsequent multidimensional calculations.

The entire process of chemical transformations of a reactive mixture of silane and oxidant in the presence of inert gas will be subdivided standardly into two stages. The first of them describes the induction period using the equation

$$\frac{d\beta}{dt} = - \frac{1}{t_{\text{ign}}}. \quad (7)$$

At the beginning of the induction zone, we have  $\beta = 1$ , and at the end,  $\beta = 0$ . For the region of ignition delay, we must know the dependence  $t_{\text{ign}} = t_{\text{ign}}(T, \xi_{\alpha})$ . Note that we have not encountered approximation formulas of this kind for silane in the available literature. Furthermore, the existing experimental data on ignition-delay times are rather scanty; all of them have been presented above when a detailed kinetic scheme of oxidation was verified. Therefore, we find the dependence  $t_{\text{ign}} = t_{\text{ign}}(T, \xi_{\alpha})$  by approximating calculated ignition-delay times for silane, oxygen, and inert-gas mixtures. From the analysis and approximation of numerous calculated data for nearly stoichiometric and fuel-deficient mixtures of silane and air (nearly stoichiometric mixtures:  $\xi_{\text{SiH}_4} = 0.1$ ,  $\xi_{\text{O}_2} = 0.2$ , and  $\xi_{\text{N}_2} = 0.7$ ; fuel-deficient mixtures:  $\xi_{\text{SiH}_4} = 0.1$ ,  $\xi_{\text{O}_2} \geq 0.2$ , and  $\xi_{\text{N}_2} \leq 0.7$ ) at a pressure of 1 atm behind the reflected SW, we obtained the following expression for the delay time of ignition of silane in air behind the reflected SW:

$$t_{\text{ign}} = 1.374 \cdot 10^{-10} \xi_{\text{SiH}_4}^{-0.84} \xi_{\text{O}_2}^{-0.07} e^{\frac{9695.45}{T}}. \quad (8)$$

To the stoichiometric composition of the silane–air mixture  $\text{SiH}_4 + 2(\text{O}_2 + 3.76\text{N}_2)$ , there correspond the concentrations  $\xi_{\text{SiH}_4} = 0.104$ ,  $\xi_{\text{O}_2} = 0.208$ , and  $\xi_{\text{N}_2} = 0.688$ . The ignition-delay time in (8) is measured in seconds.

Figure 3a gives a comparison of the calculated data obtained from the detailed kinetics and from formula (8) for stoichiometric, fuel-enriched, and fuel-deficient silane–air mixtures. It can be seen that approximation (8) describes well the calculated data not only for stoichiometric and lean mixtures (it is for these mixtures that it was obtained), but also for fuel-rich mixtures. The compositions and concentrations of the components of such mixtures are given in Table 1. Figure 3b gives the dependences of the ignition-delay time behind the reflected SW in the silane–oxygen–argon mixture which have been obtained from the detailed kinetics and approximation formula (8). The compositions of the mixtures are given in Table 2. It can be seen that formula (8) yields a somewhat overstated estimate for the ignition-delay time, but the slopes of the curves are similar to the slopes of the plots obtained using detailed-kinetics-based calculations of the problem. Thus, the reduced activation energy in formula (8) holds for such mixtures, too. However, (8) was obtained for silane–air mixtures, not for silane–oxygen–argon ones. Therefore, to minimize the disagreement between the data calculated from the detailed kinetics and approximation (8), we must modify it slightly. This can be done, e.g., by adding the multiplier  $(1 - \xi_{\text{Ar}})^2$  to the preexponential factor of formula (8). Then it will describe well ignition-delay times for silane–oxygen–argon mixtures and will automatically become dependence (8) for silane–air mixtures when  $\xi_{\text{Ar}} = 0$ . The calculations have shown that  $z \approx 0.004$ .

We take account of the dependence of the ignition-delay time on pressure. From the analysis and approximation of numerous calculated data for silane mixtures with oxygen, nitrogen, and argon at different pressures, we have obtained the following expression for the ignition-delay time behind the reflected SW:

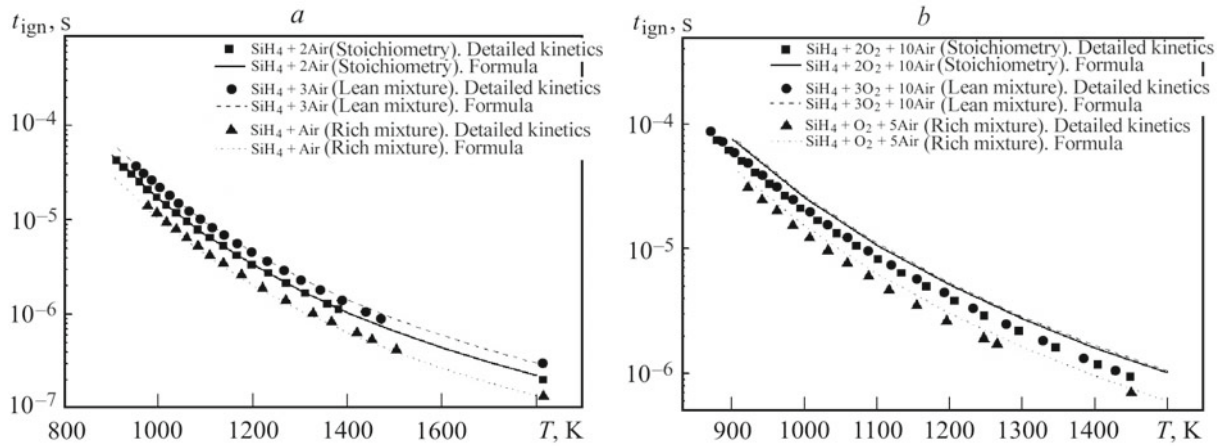


Fig. 3. Delay time of ignition of silane vs. temperature behind the reflected SW in silane–air (a) and silane–oxygen–argon mixtures (b). Comparison with approximation formula (8).

TABLE 1. Compositions of Mixtures of Silane and Air

Composition of the mixture	$\xi_{SiH_4}$	$\xi_{O_2}$	$\xi_{N_2}$
SiH <sub>4</sub> + Air (Rich mixture)	0.189	0.189	0.622
SiH <sub>4</sub> + 2Air (Stoichiometry)	0.104	0.208	0.688
SiH <sub>4</sub> + 3Air (Lean mixture)	0.072	0.216	0.712

TABLE 2. Compositions of Mixtures of Silane, Oxygen, and Argon

Composition of the mixture	$\xi_{SiH_4}$	$\xi_{O_2}$	$\xi_{Ar}$
SiH <sub>4</sub> + O <sub>2</sub> + 5Ar (Rich mixture)	0.121	0.121	0.758
SiH <sub>4</sub> + 2O <sub>2</sub> + 10Ar (Stoichiometry)	0.0645	0.129	0.8065
SiH <sub>4</sub> + 3O <sub>2</sub> + 10Ar (Lean mixture)	0.06	0.18	0.76

$$t_{ign} = 1.374 \cdot 10^{-10} \left( \frac{p}{p_0} \right)^{-0.47} \xi_{SiH_4}^{-0.84} \xi_{O_2}^{-0.07} (1 - \xi_{Ar})^{0.004} e^{\frac{9695.45}{T}}, \quad (9)$$

where  $p_0 = 1$  atm,  $p$  is measured in atmospheres. Figure 4 gives a comparison of the calculated data obtained from the detailed kinetics and from formula (9) for the stoichiometric silane–air mixture at pressures from 1 to 10 atm behind the reflected SW. It can be seen that approximation (9) describes well the calculated data in the range of pressures from 1 to 10 atm and temperatures from 1000 to 2200 K.

**Parameters of the Chapman–Jouguet Wave in a Silane–Air Mixture with Additions of Chemically Inert Microparticles and Without Them.** Calculation of the parameters of the Chapman–Jouguet wave in silane–air mixtures (without and in the presence of chemically inert particles) will be performed according to the model proposed in [5–8]. Recall that within the framework of this model, we made the following basic assumptions. The gas is ideal. The condensed phase is incompressible. The total volume of particles and the partial pressure of the condensed phase are negligible. The particles are in mechanical and thermal equilibrium with the gas (the velocity and temperature of the gas and particles are equal). The loss by friction and heat removal into tube walls is disregarded. The DW structure is in agreement with the Zel’dovich–von Neumann–Döring model. In the Chapman–Jouguet plane, the gas is in a chemical-equilibrium state. The range of applicability of such assumptions was analyzed in [5–8] in detail. In particular, the assumptions of thermal and mechanical equilibrium

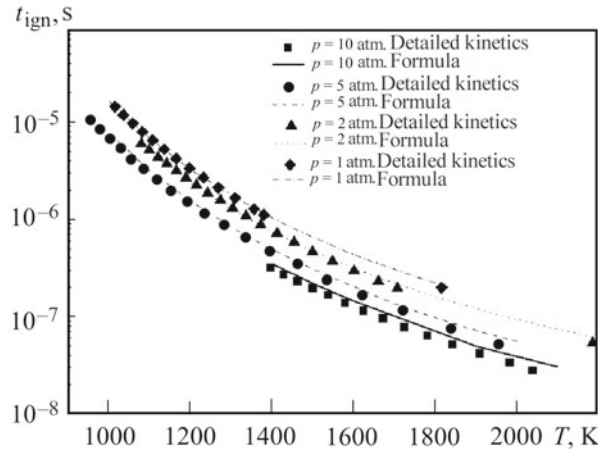


Fig. 4. Delay time of ignition of silane vs. temperature behind the reflected SW in the silane–air mixture. Comparison with approximation formula (9).

between phases hold for particles having a relatively small size (of the order of ten microns, i.e., microparticles). In this case the characteristic times of thermal and mechanical relaxation between phases are much shorter than the characteristic times of change in the parameters of the two-phase flow inside the DW reaction zone. Thermodynamic parameters of the mixture in chemical equilibrium were calculated from formulas [16–19] which are highly accurate and consistent with the second law of thermodynamics and take account of the Le Chatelier principle.

Results of calculating the detonation rate and the parameters of the wave in the Chapman–Jouguet plane in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles are presented in Fig. 5, and the parameters of the wave behind the leading shock front, in Fig. 6. Results of calculating the parameters of detonation in a silane–air gas mixture ( $G = 0$ ) of varying stoichiometry are presented in Figs. 7 and 8. It can be seen that the DW pressure and temperature in the Chapman–Jouguet plane in the mixture in question (17.89 atm and 2973 K) are close to the relevant parameters of the DW in stoichiometric hydrogen– and methane–air mixtures (15.6 atm, 2947 K and 19.13 atm, 3114 K [20]). Such high pressures and temperatures implemented in detonation processes should be taken into account in silane mixtures when the relevant heat and shock loads on industrial and civil structures are calculated.

From the formulas of the kinetic scheme [9] and the system of equations (15) and (16), we also calculated the parameters of detonation in the silane–air mixture at different fuel-oxidant ratios. The obtained results are in good agreement with the calculations given in Figs. 5 and 6 at  $\alpha = 0$  and in Figs. 7 and 8 at  $c = 0.0951$ . For example, the detonation rate, the pressure, and the temperature in the Chapman–Jouguet plane are equal to 1760 m/s, 17.5 atm, and 2727 K (calculation from [9], (5), and (6)) and to 1812 m/s, 17.89 atm, and 2973 K (Figs. 5 and 7) respectively.

**Dimension of the Cell and Energy of Direct Initiation of Gaseous Detonation in the Silane–Air Mixture.** To assess the dimension of a gaseous-detonation cell, we use the universally adopted model [4]:

$$b_p = 4 \frac{E_a}{T_{SW}} D t_{\text{ign,SW}} , \quad a_p = 0.6 b_p . \quad (10)$$

To calculate the induction period of a chemical reaction in a silane–air mixture, we use formula (9). Calculation results are presented in Fig. 9 (figures a–c complement each other). It can be seen that the dimension of the cell in silane–air mixtures is much smaller than that in analogous acetylene mixtures. For example, in a stoichiometric silane–air mixture, we have  $a_p = 2.62$  mm (Fig. 9c), whereas in an analogous acetylene mixture,  $a_p = 8$  mm [21, 22].

It is common knowledge [2, 3] that the energy of direct initiation of a gas mixture can be assessed from the formula

$$E^* = AD^2 a_p^2 ,$$

where the coefficient  $A$  will further be assumed constant. Therefore, the energy of direct initiation of the silane–air mixture  $E_s^*$  can be assessed as follows:

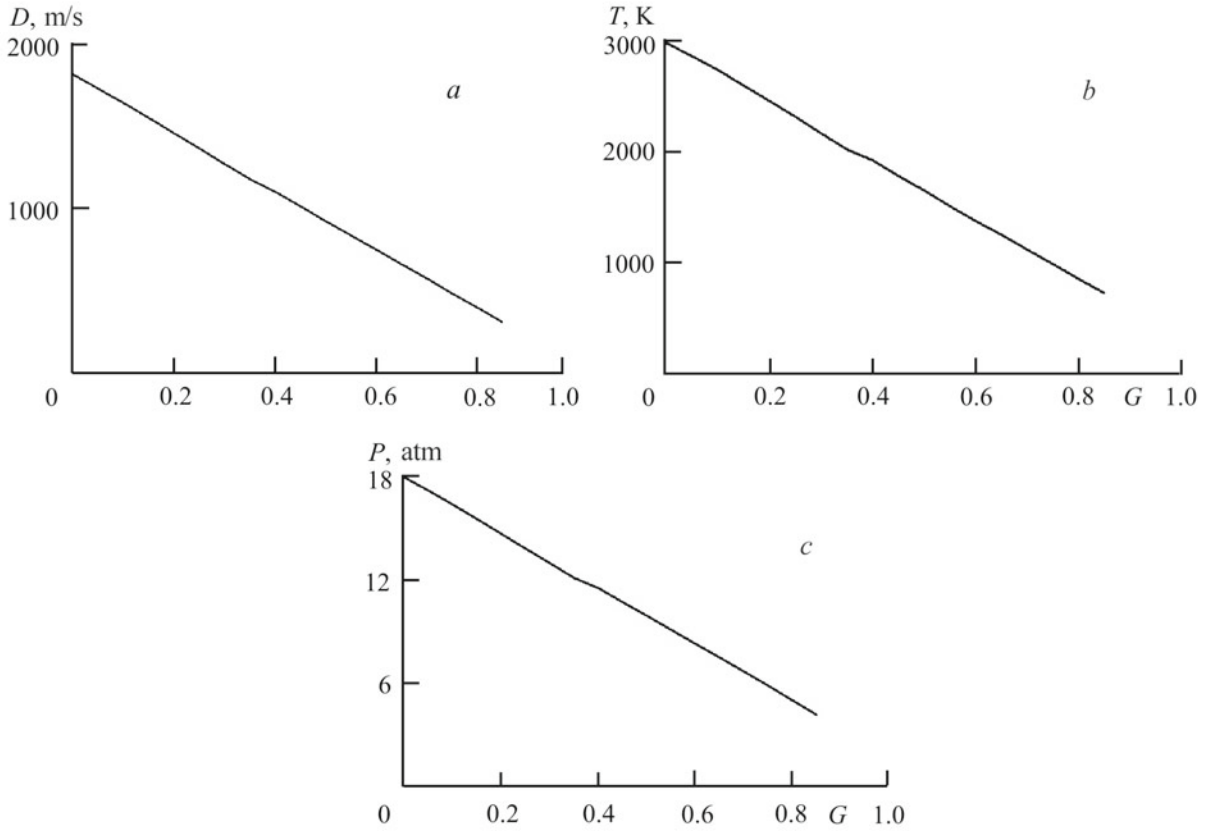


Fig. 5. Detonation rate (a) and parameters of the wave (b and c) in the Chapman–Jouguet plane in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles at  $T_0 = 293 \text{ K}$  and  $P_0 = 1 \text{ atm}$ .

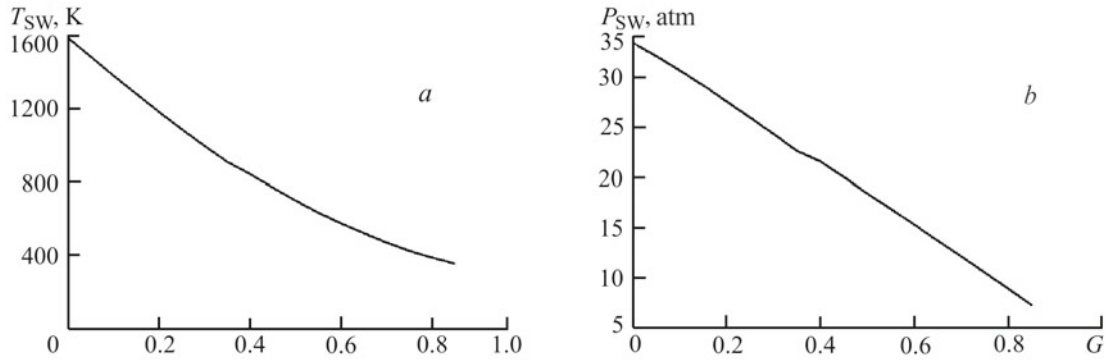


Fig. 6. Temperature (a) and pressure (b) behind the DW leading shock front in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles at  $T_0 = 293 \text{ K}$  and  $P_0 = 1 \text{ atm}$ .

$$\frac{E_s^*}{E_g^*} = \left( \frac{u_{D,s}}{u_{D,g}} \frac{a_{p,s}}{a_{p,g}} \right)^2.$$

As a gas with the known quantities  $E_s^*$ ,  $u_{D,g}$ , and  $a_{p,g}$ , we consider, e.g., a stoichiometric acetylene–oxygen mixture in which  $E_g^* \approx 0.4 \text{ g TNT}$ ,  $u_{D,g} = 1864 \text{ m/s}$ , and  $a_{p,g} \approx 8 \text{ mm}$  [20–22]. According to the above calculations, for the stoichiometric silane–air mixture we have  $u_{D,s} = 1812 \text{ m/s}$  and  $a_{p,g} \approx 8 \text{ mm}$  [20–22]. Therefore, we obtain

$$E_s^* = 0.4 \text{ g} \cdot \frac{(1812)^2}{(1864)^2} \frac{(2.62)^2}{(8)^2} = 0.4 \text{ g} \cdot 0.1 = 0.04 \text{ g TNT}.$$



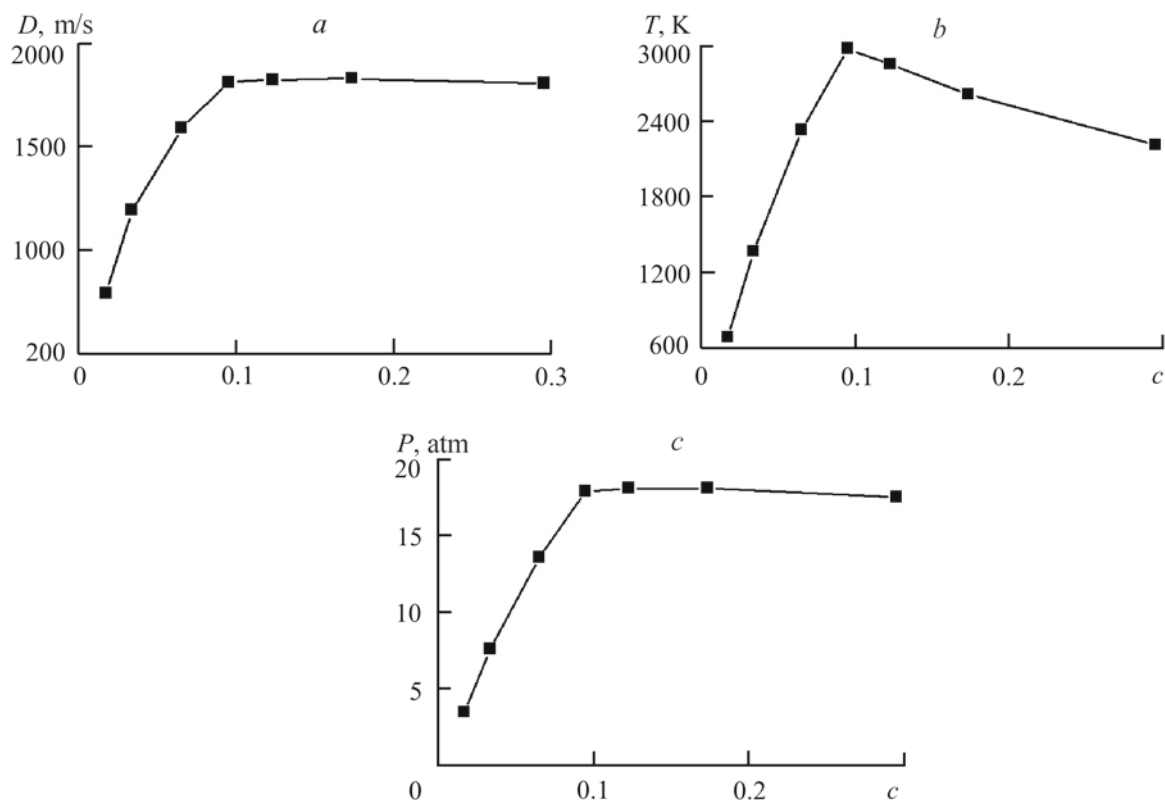


Fig. 7. Detonation rate (a) and parameters of the flow (b and c) in the Chapman–Jouguet plane in the silane–air mixture  $c\text{SiH}_4 + (1 - c)\cdot\text{air}$ .

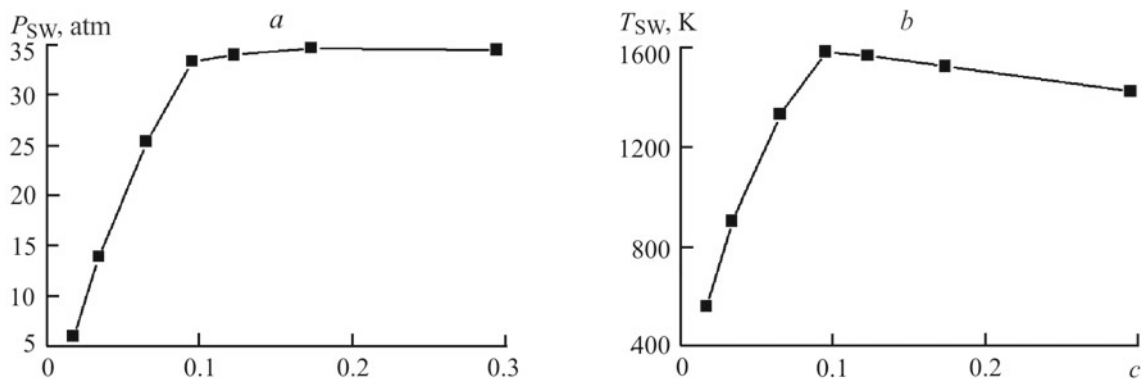


Fig. 8. Parameters of the Neumann peak in the DW in the silane–air mixture  $c\text{SiH}_4 + (1 - c)$  air: a) mixture pressure and b) mixture temperature.

It can be seen that the calculated energy of direct initiation of the stoichiometric silane–air mixture is one order of magnitude lower than the analogous energy in the relevant acetylene mixture.

**Dimension of the Cell of a Detonation Wave in the Mixture of a Gas with Chemically Inert Particles.** In accordance with [5–8], we assume that the dimension of a detonation cell in a mixture of a gas with particles has the same dependence on the wave parameters as the dimension of the cell in gas mixtures. This means that the above-presented formula for calculation of the dimension of a gaseous-detonation cell is assumed true for the considered mixtures of a gas with particles as well. Thus, (9) and (10) yield

$$\frac{b}{b_p} = \frac{D}{D_p} \left( \frac{P_{sw,p}}{P_{sw}} \right)^{0.47} \frac{T_{sw,p}}{T_{sw}} e^{E_A \left( \frac{1}{T_{sw}} - \frac{1}{T_{sw,p}} \right)}$$

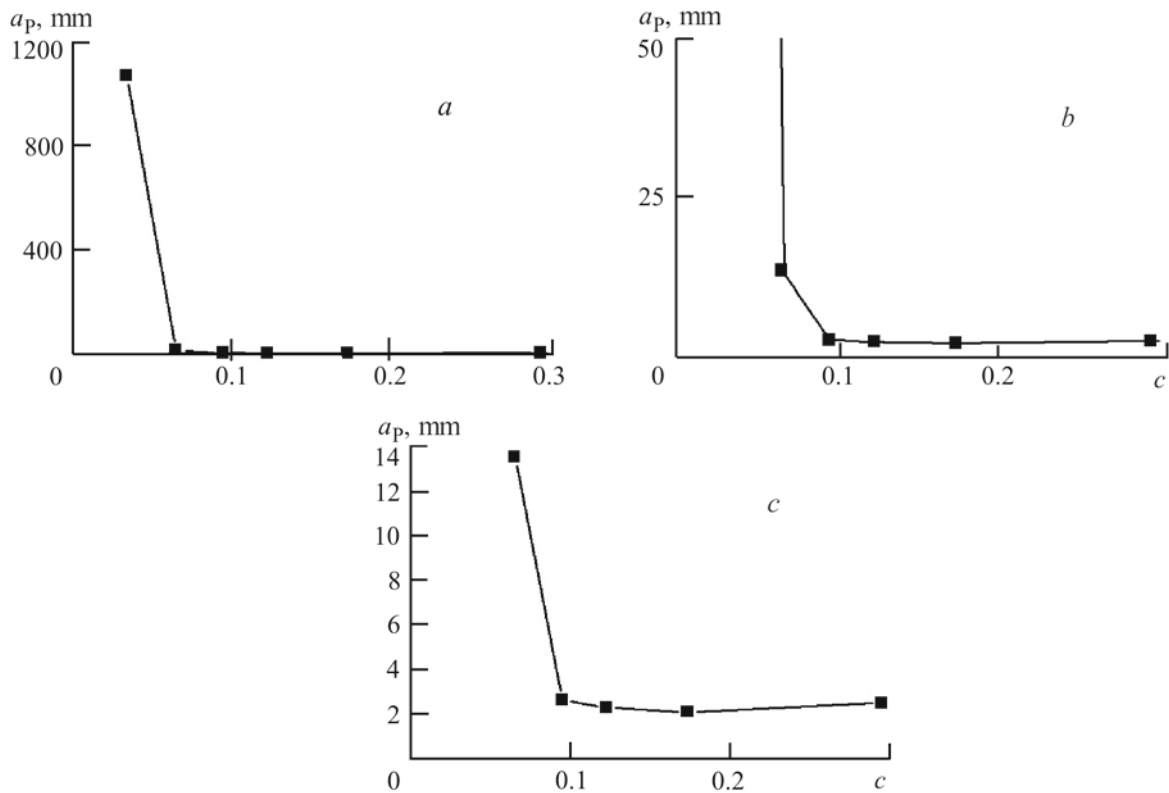


Fig. 9. Dimension of the detonation cell in the silane–air mixture  $c\text{SiH}_4 + (1 - c)\text{air}$  for different scale on the ordinate axis.

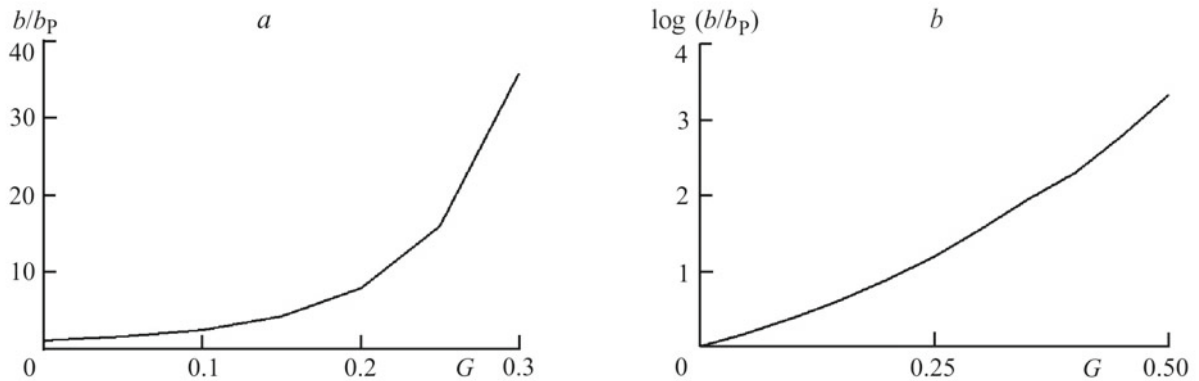


Fig. 10. Relative dimension of the DW cell (a) and the logarithm of the relative dimension of the DW cell (b) in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles at  $T_0 = 293 \text{ K}$  and  $P_0 = 1 \text{ atm}$ .

The wave parameters without a subscript correspond to the two-phase mixture. The transverse dimension of the cell in the gas without particles is assumed known. Also, we will assume that  $a/a_p = b/b_p$ . The result of calculating the relative dimension of the DW cell in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles from the above formula and from the data presented in Figs. 7 and 8 is shown in Fig. 10.

**Calculation of the Explosion of a Mixture of a Chemically Reactive Gas with Chemically Inert Microparticles at Constant Pressure and Temperature.** Within the framework of the employed model of a two-phase mixture, a calculation was done of the parameters of explosion of a stoichiometric silane–air mixture without and with additions of chemically inert microparticles ( $\text{Al}_2\text{O}_3$ ) at constant pressure ( $P = \text{const}$ ) and volume ( $\rho = \text{const}$ ). The explosion of a silane–air mixture in a constant-volume chamber was investigated in [23]. In our calculations, it was assumed that as a result of instantaneous

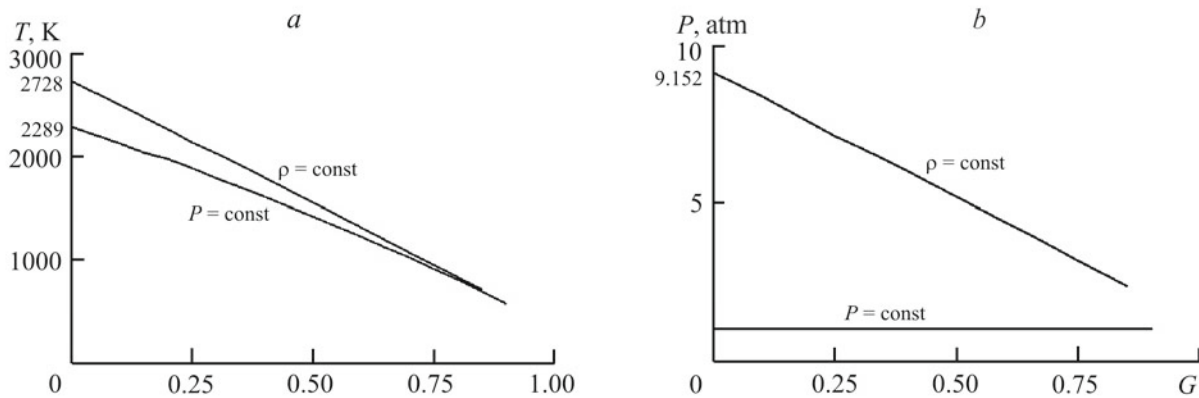


Fig. 11. Temperature (a) and pressure (b) of explosion at constant pressure and volume in a stoichiometric silane–air mixture with  $\text{Al}_2\text{O}_3$  microparticles at  $T_0 = 293$  K and  $P_0 = 1$  atm.

chemical transformations, the mixture changes to a state of chemical equilibrium. For the calculations, we used the laws of conservation of internal energy (at  $\rho = \text{const}$ ) and enthalpy (at  $P = \text{const}$ ). The mixture's thermodynamic parameters were calculated from the formulas of [16–19]. Calculation results are presented in Fig. 11. It can be seen that in the case of a purely gas mixture ( $k = 0$ ) the explosion pressure at  $\rho = \text{const}$ , which is equal to 9.15 atm, is comparable with the relevant quantities realized in explosions in stoichiometric hydrogen–air and methane–air mixtures (8.01 atm and 9.77 atm, respectively) [20]. This constitutes additional evidence in favor of the high potential hazard of silane–air mixtures.

As follows from Figs. 5, 6, 10, and 11, increase in the weight fraction of particles leads to a substantial decrease in the detonation rate, pressure, and temperature behind the leading shock front and in the Chapman–Jouguet plane, a strong growth in the dimension of the detonation cell, and a significant reduction in the pressure and temperature of the mixture in explosion. Thus, adding particles can serve as one tool for control of the parameters of explosive processes and can minimize the detonation hazard of silane–air mixtures.

**Experimental and Theoretical Study of the Explosion of Clouds of a Silane–Air Mixture.** One of the most important engineering problems of explosion safety is to assess the pressure profile of a compression wave produced by the explosion of a cloud of a gas mixture, which is formed, e.g., during the escape of combustible gas from a high-pressure cylinder and its mixing with the environment or the evaporation of liquid fuel in its leakage from a pipeline or a tank. Investigations of this kind have been carried out to date for a wide range of hydrogen– and hydrocarbon–air mixtures. Formulas to calculate spatial profiles of pressure as a function of the kind of fuel and its total mass in the cloud and explosion conditions were obtained [24–26]. Recent studies have shown that the obtained formulas are not always applicable to a silane–air mixture [27]. One reason for this inconsistency, as we think, can be that one of the main products of explosion of the silane–air mixture is  $\text{SiO}_2$  molecules which condense on expansion of the explosion products. This results in the change in the adiabatic exponent in the mixture and in the molar mass of the gas, and also in additional heat release due to the phase transition. Another possible reason can be that the range between the upper and lower concentration limits of ignition of silane is much wider than in typical hydrocarbons [28]. As a result, the percentage of silane experiencing explosive transformation is much higher than the analogous quantity in the case of typical hydrocarbons.

As a first step to elucidate the reason for the above inconsistency, we conducted a series of experiments on measurement of the pressure profile produced by the explosion of a cloud of a silane–air mixture, which is formed during the escape of silane from a high-pressure cylinder into a half-closed space of cubic shape ( $30 \times 30 \times 30$  cm wire skeleton covered with a polyethylene film) (Fig. 12a). With this method of obtaining a silane–air cloud, the average concentration of the fuel in the mixture is computed quite simply and accurately. To experimentally model the process of mixing of silane with air, the cubic volume was filled with smoke which, unlike silane, is visible (Fig. 12b).

To determine the spatial distribution of silane in a cubic volume more accurately, we numerically modeled the escape of silane into a closed volume. We used the ANSYS Fluent 14.5 computer package for calculations. The jet flowed out from the center of one side of the cube through a circular hole. Upward and downward from the site of efflux of the jet, there were holes through which the air displaced by the silane flowed out into the surrounding space. Calculation results are in qualitative agreement with the conducted experiment on filling the considered volume with smoke (Fig. 12).

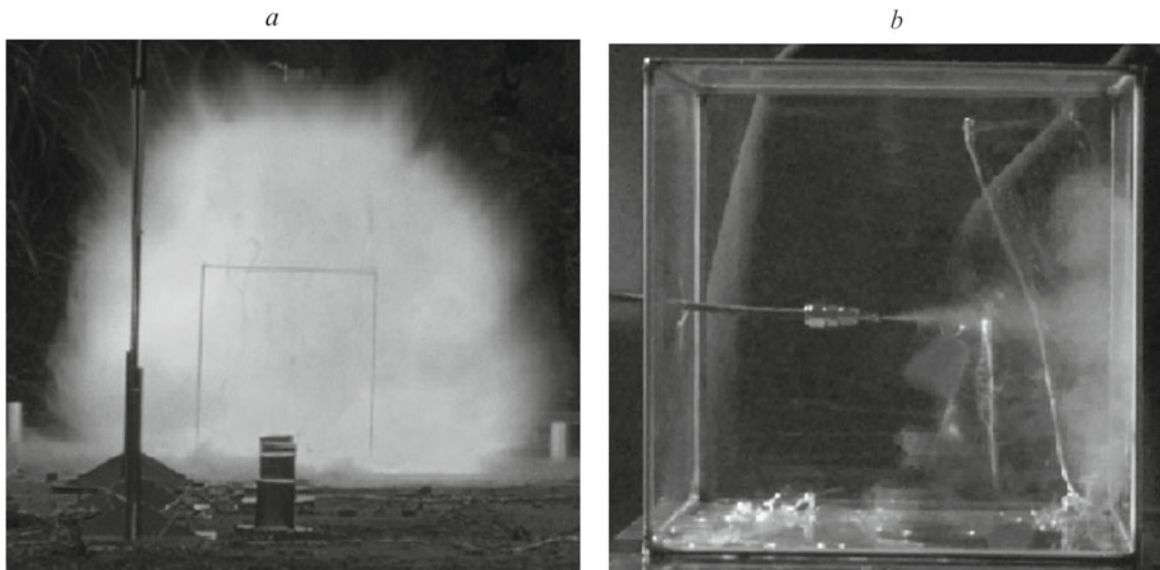


Fig. 12. Explosion of the cubically shaped volume filled with silane–air mixture (initiation was at the center) (a); the process of filling the cube with smoke (b).

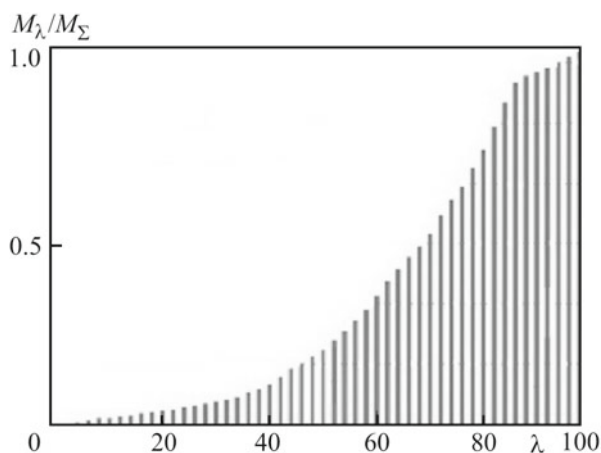


Fig. 13. Integral characteristic of spatial distribution of silane in the cloud.

Consideration was also given to the efflux of a subsonic jet of silane into an unbounded space [29, 30], and the possibility of modeling its ignition was studied within the framework of the phenomenological approach. Use was made of Reynolds-averaged nonsteady Navier–Stokes equations within the framework of the ANSYS Fluent 14.5 applied software package. The hole diameter and the efflux velocity were equal to 3.86 mm and 2 m/s respectively. The integral result of calculating the efflux is shown in Fig. 13. The figure corresponds to an instant of time of 0.1 s after the beginning of efflux. The chemical composition of the mixture inside the cloud is  $1/100 \cdot \text{SiH}_4 + (1 - 1/100) \cdot \text{air}$ ; the stoichiometric coefficient 1 depends on the spatial distribution of silane. It can be seen that since  $\lambda_{\min} = 1.37\%$  and  $\lambda_{\max} = 96\%$  [1], in the example in question the total mass of silane in the cloud, which is between its ignition concentration limits and, accordingly, is potentially capable of exploding, is equal to  $M_{\Sigma}$ , in practice:

$$M_{\text{exp},l} = (M_{\lambda_{\max}} - M_{\lambda_{\min}})M_{\Sigma} \approx M_{\Sigma} .$$

This example demonstrates that the efficiency of explosion of the silane–air cloud may substantially exceed the relevant index during the explosion of typical hydrocarbons, since the range between concentration limits in such mixtures is much narrower than in silane.

## CONCLUSIONS

1. On the basis of detailed kinetic calculations, we have proposed an algebraic formula to assess the delay time of self-ignition of silane-containing mixtures. This enabled us to assess, from the universally adopted model, the dimension of a detonation cell and the energy of direct initiation of gaseous detonation in silane–air mixtures of varying stoichiometry.

2. It has been shown that the cell dimension and the energy of direct initiation of a DW in silane–air mixtures are much smaller than analogous quantities in relevant acetylene mixtures of high detonation power. This points to the high detonation hazard of silane-containing mixtures.

3. Consideration has been given to the issue of control of the parameters of detonation and explosion of a silane–air mixture by adding chemically inert  $\text{Al}_2\text{O}_3$  microparticles. The parameters and relative dimension of a DW cell, and also the parameters of explosion were calculated at constant pressure and volume. The results of the work can be used to solve explosion-safety problems and to assess the heat and mechanical load on industrial structures due to the self-ignition of silane–air mixtures.

4. We have conducted a series of experiments on measurement of the pressure profile from the explosion of a cloud of a silane–air mixture, which is formed during the escape of silane into a half-closed space of cubic shape. The process of filling the cubic volume with fuel has been visualized. The half-closed volume used enables us to monitor the average fuel-oxidant ratio in the mixture.

5. We have calculated the distribution of silane in the cloud formed during its escape into an unbounded space. It has been shown that in this case there can be the explosion of the entire, in practice, silane mixed with air. As a result, the efficiency of explosion of the silane–air cloud may substantially exceed the relevant index in explosion of typical hydrocarbons.

6. The results of the conducted experiments and calculations of the spatial distribution of silane in the cubic volume and the jet can be used for analysis of the reasons for the variation of the pressure profile from the explosion of the silane–air clouds from the well-known dependences obtained for hydrogen– and hydrocarbon–air mixtures.

This work was carried out with partial financial support under the projects of the Siberian Branch of the Russian Academy of Sciences and the National Scientific Council of Taiwan-2016, the Russian Science Foundation (grant No. 16-19-00010), and the Russian Foundation for Basic Research (grant No. 15-08-01947-a).

## NOTATION

$A$ , coefficient dependent on the physiochemical properties of the mixture;  $a$ , transverse dimension of the cell in the gas with particles, mm;  $a_p$ , transverse dimension of the cell in the gas without particles, mm;  $b$ , longitudinal dimension of the cell in the gas with particles, mm;  $b_p$ , longitudinal dimension of the cell in the gas without particles, mm;  $c_1$ ,  $c_2$ , and  $c_3$ , mass, momentum, and energy fluxes through the SW;  $c_f^0$ , frozen velocity of sound in the mixture, m/s;  $c_p$ , specific heat at constant pressure, J/K/kg;  $c_v$ , specific heat at constant volume, J/K/kg;  $c_{v,\alpha}$ , specific heat at constant volume of the component  $\alpha$ , J/K/kg;  $D$ , detonation rate, m/s;  $E$ , total energy, J/kg;  $E_A$ , exponent in the Arrhenius formula for calculation of the ignition delay, K;  $E_s^*$ , energy of direct initiation of the silane–air mixture;  $e$ , internal energy, J/kg;  $G$ , weight fraction of particles in the two-phase mixture;  $h_{0,\alpha}$ , enthalpy of formation of the component  $\alpha$ , J/kg;  $k$ , reaction-rate constant;  $l$ , number of reactions;  $m$ , order of reaction;  $M_{\text{exp},1}$ , total mass of silane in the cloud, which is between its ignition concentration limits;  $M_\Sigma$ , total mass of silane in the entire cloud;  $M_\lambda$ , total mass of silane in the regions of the cloud where its mole fraction is smaller than  $\lambda$ ;  $M_\alpha$ , molar weight of the component  $\alpha$ , kg/mole;  $M_{\text{mix}}$ , molar weight of the mixture, kg/mole;  $N$ , number of components in the mixture;  $P$ , pressure, atm;  $p_r$ , pressure behind the reflected SW, atm;  $R$ , universal gas constant, J/K/mole;  $T$ , gas temperature, K;  $T_c$ , temperature at the Chapman–Jouguet point, K;  $T_r$ , temperature behind the reflected SW, K;  $T_{00}$ , standard temperature, K;  $t$ , time, s;  $t_{\text{ign}}$ , ignition delay, s;  $U$ , relative velocity, m/s;  $u$ , velocity, m/s;  $u_f$ , velocity behind the front of the frozen SW, which is determined from the initial velocity, m/s;  $x$ , coordinate, m;  $\beta$ , ordering parameter describing the process of ignition delay;  $\zeta$ , spatial variable, m/s;  $\lambda$ , stoichiometric coefficient of silane;  $\lambda_{\text{min}}$  and  $\lambda_{\text{max}}$ , lower and upper concentration bounds (limits) of ignition of silane;  $\nu_\alpha$ , stoichiometric coefficient of the component  $\alpha$ ;  $\xi_\alpha$ , concentration of the mixture's component  $\alpha$ ;  $\xi_{\alpha,0}$ , concentration of the mixture's component  $\alpha$  behind the SW;  $\rho$ , gas density, kg/m<sup>3</sup>. Subscripts: 0, initial state; SW, state behind the SW; P, gas without particles; s corresponds to the silane–air mixture; g refers to the gas whose parameters and direct-initiation energy are known; the superscript ' corresponds to the back reaction.

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