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Physical and Mathematical Modeling of One-Dimensional and Two-Dimensional Structures of Detonation Waves in a Silane-Air Mixture

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Abstract. The paper deals with the problem of the 1D and 2D structures of detonation waves (DW) in the silane-air mixture within the framework of mathematical model of a nonequilibrium gas dynamics. Detailed kinetics scheme of silane oxidation as well as the newly developed reduced kinetic model of detonation combustion of silane are used. On its basis the DW structure in stoichiometric silane - air mixture and dependences of Chapman-Jouguet parameters of mixture on stoichiometric ratio between the fuel (silane) and an oxidizer (air) were obtained. It was revealed that in the reaction zone, in the structure of DW, the heat release in the model with detailed kinetics is non-monotonic.

INTRODUCTION

Silane (silicon tetrahydride) is widely used in semiconductor and photoelectric industries as a source of silicon. Silane is a self-igniting gas, which ignites due to its contact with air even under standard conditions. For this reason, it is rather hazardous in terms of fire. Attention of many researchers is riveted to modeling chemical transformations of silane, in particular, because of various issues associated with explosion and fire safety. In the paper calculations of parameters and DW structure on the basis of detailed and reduced chemical kinetics will be made.

DETAILED KINETIC MODEL OF DETONATION COMBUSTION IN SILANE MIXTURES

Let us consider the problem of the DW structures in the silane-air mixture. To describe the kinetics of ignition and combustion of this reacting mixture, we use the detailed kinetic scheme\textsuperscript{[1-2]}, which takes into account 140 forward and backward reactions of 25 species (H\textsubscript{2}, O\textsubscript{2}, H\textsubscript{2}O, OH, O, H, HO\textsubscript{2}, H\textsubscript{2}O\textsubscript{2}, SiH\textsubscript{4}, SiH\textsubscript{3}, SiH\textsubscript{2}, HSiO, SiH\textsubscript{2}O, SiH\textsubscript{3}O, SiH\textsubscript{3}O\textsubscript{2}, xSiH\textsubscript{3}O\textsubscript{2}, SiH\textsubscript{3}OH, SiH\textsubscript{3}O\textsubscript{2}H, SiH\textsubscript{2}OH, HSiOOH, SiOOH, SiO, SiO\textsubscript{2}, N\textsubscript{2}, Ar). Note that earlier\textsuperscript{[2-5]}, using this kinetic model, we have been able to describe: - ignition delay times of silane/hydrogen/air mixtures and give an analytical approximation dependence for its description; - the ignition limits of silane-oxygen and silane-air mixtures. In this paper, we will use this model and the reduced kinetics to calculate the DW structure in silane-air mixture. We also give a comparison of the DW structures obtained by the mentioned models of chemical kinetics.
APPROXIMATE KINETIC MODEL OF DETONATION COMBUSTION IN SILANE MIXTURES

Earlier we used the detailed kinetics models to calculate: – the DW propagation, attenuation and suppression in reactive gas mixtures with inert particles [6-9]; – geometrical limits of detonation [10-11]; – ignition delay times of reactive gas mixtures [3, 6] and others. In these papers a lot of advantages of detailed kinetics models were presented. But, the detailed chemistry has one disadvantage. The detailed system of equations of chemical kinetics for reactive gas mixtures, especially for silane-air mixtures, is rather cumbersome. This significantly complicates the numerical (especially, multi-dimensional) analysis of detonation processes in silane-air mixtures. Because of this difficulty in the detailed description of the chemical reaction, it is very useful to develop an approximate kinetics model of detonation combustion of silane-air mixtures.

One of the most suitable approximate models of chemical kinetics in mixtures of hydrogen with oxygen, water vapor, and inert diluents is the NFZ (i.e., Nikolaev, Fomin, Zak) model suggested in [12-15]. This 2-step model allows one to describe energy release and changes in thermodynamic parameters of the gas after the induction period with the help of one differential equation and several algebraic formulas. Comprehensive summary of NFZ model can be found in [16]. The model includes two stages: induction stage and subsequent stage of heat release. During the induction period the molar mass of a mixture is assumed to be constant and the heat release of chemical reaction is assumed to be zero.

CALCULATION RESULTS

The DW structure in a stoichiometric silane - air mixture was obtained on the basis of both models. It was found that DW velocity in Chapman-Jouget regime in the frames of the detailed kinetics model is 1844 m/s, in reduced kinetic model – 1748 m/s. The difference in these values is about 5%.

One-Dimensional Chapman-Jouguet Detonation

Figure 1 shows the DW structure in stoichiometric silane-air mixture in Chapman - Jouguet regime calculated using both kinetic models. The DW front is located at the initial zero point. The wave is moving from right to left. Behind the front the induction zone is located followed by the main heat release zone (burning zone) and at the endpoint - the equilibrium state, i.e. Chapman - Jouguet point, in which performed the condition \( u = c_e \) or \( M_e = 1 \) (Fig. 1c). Figure 1a presents the velocity distribution of the gas mixture. It is evident that these velocities are different both on the frozen shock wave (for the reduced kinetics \( u_f = 298 \) m/s, for detailed kinetics \( u_f = 365 \) m/s) and at the equilibrium point (for the reduced kinetics \( u_e = 857 \) m/s, for detailed kinetics \( u_e = 998 \) m/s). Figure 1b shows the distributions of normalized mixture pressure and temperature in the DW structure. Normalization was performed by the values of the parameters on the DW front (for reduced kinetics: \( p_f = 30.6 \) atm, \( T_f = 1567 \) K; for detailed kinetics: \( p_f = 32.85 \) atm, \( T_f = 1943 \) K). However, it turned out that the normalized pressure at equilibrium point calculated using both models are equal. Besides, the mixture temperature growth in the case of the reduced kinetics model larger compared with the detailed kinetics model. Obviously, reduced kinetics model gives more heat release in the reaction zone in comparison with the detailed kinetics model. The absolute values of pressure and temperature in the equilibrium point are follows: for the reduced kinetics – \( p_e = 19.19 \) atm, \( T_e = 2806 \) K; for detailed kinetics – \( p_e = 19.22 \) atm, \( T_e = 3060 \) K. Thus, it is seen that the absolute values of the thermodynamic parameters at equilibrium point are close to each other. Fig. 1c shows the distributions of the flow Mach number in the DW structure: for the reduced kinetics – frozen \( M_f \) and equilibrium \( M_e \) Mach numbers, for detailed kinetics – frozen Mach number \( M_f \). Note that in the model with reduced kinetics the value of equilibrium Mach number at the equilibrium point approaches the unity that corresponds to Chapman - Jouguet regime. At the same time, the values of frozen Mach number in equilibrium point for both kinetic models coincides with each other and are equal to \( M_f = 0.85 \). Therefore, it can be argued that in the case of detailed kinetics we also have the Chapman - Jouguet regime. Figure 1d presents the gas mixture molar mass distribution. It is evident that the differences in the
parameters calculated for both models, is not greater than 3%. Obviously, all these differences in the data caused by the slight differences in DW velocities in Chapman-Jouget regimes in the calculation by these two models.

FIGURE 1. Distribution of flow parameters in DW structure: (a) gas velocity, (b) gas pressure and temperature, (c) Mach numbers, (d) gas molar mass

Non-Monotonic Heat Release

Furthermore, it should be noted that in calculations with detailed kinetics model distribution of flow parameters in the DW structure is a non-monotonic (Fig. 1) as it was in [3]. At the same time, the reduced kinetics model provides monotonous distribution of flow parameters, which means that the heat release in the reaction zone is monotonous. Figure 2 shows the distribution of heat release in the reaction zone, calculated by the model with the detailed kinetics. It is seen that near the DW front we have a small peak of heat release, followed by a small heat absorption area, which replaced by small area of the second heat release, and at the equilibrium point there is no heat release. Thus, in silane-air mixture in calculations with the detailed kinetics model nonmonotonic heat release in the reaction zone is observed.
Calculation of Detonation Cell Size in a Stoichiometric Silane-Air Gaseous Mixture

The real structure of the detonation wave differs significantly from the one-dimensional Chapman-Jouguet wave structure. It is essentially non-uniform [17, 18]. At the leading front of the detonation wave the transverse waves are presented, the motion and periodic collision of which leads to the formation of the so-called cellular (multi-front) structure of the detonation wave. Despite the strong difference in the wave structures, the average velocity, pressure and temperature of the multi-front wave are close to the corresponding quantities in Chapman-Jouguet one-dimensional wave.

As noted above, along with such parameters as velocity, pressure and temperature, the cell size of the gas detonation is one of the main characteristics of the detonation hazard of gas mixtures [17-18]. In particular, the geometric limits of the DW propagation, the energy of its direct initiation and the conditions of deflagration to detonation transition are determined by the cell size. The dimensions of the detonation cells were measured experimentally and were calculated theoretically for a fairly wide range of gas mixtures [17-18]. At the same time, cell sizes in silane-air gas mixtures are still not known, which makes it difficult to assess the detonation hazard of silane-air mixtures.

To estimate the cell size of the detonation wave in silane-air mixtures, we use the conventional formula given in [18]:

\[ b = 4 \frac{E_a}{RT_{SW}} u_D \tau_{SW}, \quad a = (0.6\div0.8)b. \]

Here \( b \) and \( a \) are the longitudinal and transverse cell size, \( u_D \) and \( \tau_{SW} \) are the detonation velocity and the induction period of the chemical reaction behind the front of leading shock of the Chapman-Jouguet wave. Thus, to calculate the cell size, it is sufficient to know the formula for calculating the induction period of the chemical reaction and the parameters of the one-dimensional wave.

A formula for calculating the induction period of chemical reaction in a silane-air mixture was proposed in [3]:

\[ \tau = 1.37 \cdot 10^{-10} \left( \frac{P}{P_0} \right)^{-0.47} e^{-0.84} \xi_{SiH_4} \xi_D_2 \cdot \exp(9695 / T). \]

Induction period \( \tau \) - in seconds, \( P \) - gas pressure (atm), \( \xi_{SiH_4} \) and \( \xi_D_2 \) - silane and oxygen mass concentrations, \( P_0 \) are equal to 1 atm.

Thus, we calculated the parameters of the Chapman-Jouguet wave: its velocity and also the pressure and temperature behind the front of leading shock. So, it is possible to calculate the cell size of a detonation wave in...
silane-containing mixtures. The calculation results of the transverse detonation cell size in a silane-air mixture in dependence of stoichiometric ratio - $\alpha$ in different scales are shown in Fig. 3.

From the calculation results it follows that the cell size in the silane-air mixture are much lower than the corresponding values in the acetylene-air mixture characterized by high detonation hazard. For example, in a stoichiometric mixture, the cell size is 2.6 and 8 mm, respectively. This indicates a high detonation hazard of silane.

It is known that a regular cellular structure of the detonation wave, which is characterized by a symmetrical transverse waves and small cell size dispersion, corresponds to the low activation energy in the formula of the induction period. Among the mixtures with a regular cellular structure are, for example, hydrogen-oxygen mixtures. In contrast, the irregular structures of detonation wave correspond to large values of the activation energy. Irregular structure of detonation wave (for example, in methane-oxygen mixture) includes asymmetric main transverse waves, the hierarchy of secondary transverse waves and big dispersion of detonation cell sizes. As can be seen from the above formula for calculating the induction period in silane-air mixtures, the activation energy is comparable with the corresponding value for oxygen-air mixtures. Therefore, we should expect that the cellular structure of a detonation wave in silane-air mixtures will be regular.
CONCLUSIONS

The physical and mathematical model of non-equilibrium gas dynamics describing the oxidation of silane-air mixture in a detonation wave based on detailed kinetics of silane oxidation was proposed. On its basis DW structure in a stoichiometric silane-air mixture were defined. It was revealed that in the reaction zone, in the DW structure, the heat release in calculations with detailed kinetics model is non-monotonic.

The reduced two-stage kinetic model of detonation combustion of stoichiometric silane-air mixture is suggested. The model was used in calculation of parameters and structure of stationary one-dimensional DW in stoichiometric silane-air mixture. It is shown that the reduced model can be successfully used for calculation velocity of DW, DW parameters in C.-J. point and estimation of spatial scales of induction zone and the main heat release zone. At the same time, it is problematic to use the approximate model for calculation of wave profiles inside the induction zone. Because in the frames of the approximate kinetic model the heat release during induction period is monotonous, but according to the detail kinetic scheme the heat release during induction period is non-monotonous.

The detonation cell size in stoichiometric silane-air mixture has been estimated. It is shown that this size is of the same order of magnitude as the cell size in stoichiometric acetylene-air mixtures. This clearly indicates a high detonation hazard of silane-air mixtures. It is shown that detonation cell structure in silane-air mixtures is regular.

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