MODIFIED MODEL OF ALUMINIUM PARTICLE IGNITION

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Abstract: A simple model of aluminum particle ignition in gaseous oxidizer has been developed. The model takes into account transient heat transfer to particle surface and nonuniform temperature distribution inside the particle. Particle ignition delays predicted by the model were compared with available experimental data. The effects of transient heat transfer and temperature nonuniformity in the particle were shown to be important for the improved modeling of aluminum particle ignition.

Introduction

Ignition and combustion of aluminum particles are the issues of interest for many industrial and aerospace applications. When simulating particle ignition and combustion, various models based on the classical Newton heat transfer law and overall metal oxidation kinetics are usually used [1, 2].

As shown in [3, 4] based on the solution of the transient conjugate heat transfer problem between gas and particle, the most widely used standard model applying the Newton law overestimates considerably the mean particle temperature during the preignition period. As for the overall kinetic law of particle ignition, relevant effective kinetic parameters are usually calculated by fitting the standard particle ignition model with experimental data. As a result, the values of the effective kinetic parameters can differ considerably from the actual values. In [5], a simple model of magnesium particle ignition which takes into account transient heat transfer between gas and particle as well as nonuniform temperature distribution inside the particle has been developed. The results presented in [5] do indicate that the ignition delay time is sensitive to these effects. Moreover, the effective kinetic parameters obtained from the solution of the inverse kinetic problem were found to differ significantly from the values available in the literature. In this paper, the model of Ref. [5] is applied to aluminum particle, whose oxidation rate is known to depend on the oxide film thickness at particle surface.

1. Problem Formulation for Aluminum Particle Ignition

1.1. STANDARD MODEL

The most widely used ("standard") model of spherical aluminum particle ignition in oxygen containing atmosphere is written in the following form [1]:

$$cm\frac{d\overline{T}}{dt} = \alpha S(T_{g\infty} - \overline{T}) + Sq\rho_{ox}\frac{K\overline{c}_{ox}}{h}e^{\left(-E/R_{Al}\overline{T}\right)}; \qquad \overline{T}(0) = T_0 \qquad (1)$$

$$\frac{dh}{dt} = \frac{K\overline{c}_{ox}}{h}e^{\left(-E/R_{Al}\overline{T}\right)}; \qquad h(0) = h_0 \qquad (2)$$

where t is time, $\overline{T}$ is the mean particle temperature, $T_{g\infty}$ is the ambient gas temperature, α is the heat transfer coefficient, $S = 4\pi R^2$ is the particle surface area, $m = (4/3)\pi\rho R^3$ is the particle mass, R is the particle radius, c is the specific heat of particle material, ρ is the particle material density, q is the chemical reaction heat related to the metal oxide mass, ρ_{ox} is the oxide density, E and K are the

effective Arrhenius parameters, R_{Al} is the gas constant of aluminum, $\overline{c}_{ox}$ is the mass fraction of oxygen in an environment (for oxygen $\overline{c}_{ox} = 1$), and *h* is the oxide film thickness. From now on, index 0 relates to the initial parameters, and index *g* relates to the gas phase.

1.2. MODIFIED MODEL

The modified model has two differences from the standard model [1], namely

(*i*) heat transfer coefficient α is replaced by the time-dependent effective heat transfer coefficient α_{eff} and

(*ii*) mean particle temperature $\overline{T}$ in the Newton law in Eq. (1) and in the kinetic law in Eq. (2) is replaced by the particle surface temperature T_i.

In addition, instead of the linear oxidation law used for magnesium particles in [5], the modified model applies

(*iii*) the parabolic kinetic law which is more appropriate for simulating aluminum particle ignition.

Thus, the modified model is formulated as

$$cm\frac{d\overline{T}}{dt} = \alpha_{eff} S(T_{g\infty} - T_i) + Sq\rho_{ox}\frac{K\overline{c}_{ox}}{h}e^{\left(-E/R_{Al}T_i\right)}; \qquad \overline{T}(0) = T_0 \qquad (3)$$

$$\frac{dh}{dt} = \frac{K\overline{c}_{ox}}{h}e^{\left(-E/R_{Al}T_i\right)}; \qquad h(0) = h_0 \qquad (4)$$

$$\Theta_i = \sum_{j=0}^{n} b_j \overline{\Theta}^{\,j}; \qquad (5)$$

where $\alpha_{eff} = \lambda_g\left(1 + \sqrt{R^2/\pi a_g t}\right)R^{-1}$ [3, 4], a_g is the gas thermal diffusivity, $\Theta_i = T_i/T_0$ is the dimensionless particle surface temperature, $\overline{\Theta} = \overline{T}/T_0$ is the dimensionless mean particle temperature, *n* is the polynomial order and b_j are the polynomial coefficients ($b_0 = 0.0469521$, $b_1 = 0.931$, $b_2 = 0.03682$, $b_3 = -6.129\text{e-}3$) [3, 4].

1.3. INVERSE KINETIC PROBLEM

The effective kinetic parameters *E* and *K* entering Eq. (4) can be obtained from the solution of the inverse kinetic problem using the following procedure.

a) The activation energy E_{ij} (in J/kg) characteristic for the conditions relevant to any two experimental points *i* and *j* is calculated based on available experimental data using the approach reported in [6]:

$$E_{ij} = \frac{R_{Al}\cdot T_{g\infty i}\cdot T_{g\infty j}}{T_{g\infty i} - T_{g\infty j}}\cdot \ln\left(\frac{R_i\cdot \alpha_{eff\,i}\left(T_{si}, t(T_{si})\right)}{R_j\cdot \alpha_{eff\,j}\left(T_{sj}, t(T_{sj})\right)}\cdot\frac{T_{g\infty i}^{\,4}}{T_{g\infty j}^{\,4}}\right); \qquad (6)$$

$$t(T_s) = \left(\Theta_s - 1\right)\left(\sum_{k=0}^{l} p_k \Theta_g^{\,k}\right); \qquad (6a)$$

where *i* and *j* are the numbers of experimental points, $T_s = T_{g\infty} + \left(R_{Al}T_{g\infty}^{\,2}/E_{ij}\right)$ is the particle surface temperature before ignition, $t(T_s)$ is time, *l* is the polynomial order (*l* = 2), p_k are the polynomial coefficients ($p_0 = 0.045$, $p_1 = -0.012$, $p_2 = 0.0009134$), $\Theta_s = T_s/T_0$ is the

dimensionless particle surface temperature before ignition, $\Theta_g = T_{g\infty} / T_0$ is the dimensionless ambient gas temperature;

b) The mean activation energy E is then calculated as the mathematical expectation of the set E_{ij};

c) The standard deviation $\pm \Delta E$ of the mean activation energy is then readily found;

d) The preexponential factor K_i (in m^2/s) relevant to the ith experimental point is then calculated from the formula [6]:

$$K_i = \frac{\Omega_{kp}}{3} \cdot \left(\frac{R_{Al} \cdot T_{g\infty i}^{\ 2}}{E} \right)^2 \cdot \frac{c \cdot \rho \cdot \alpha_{eff\ i} \cdot R_i}{(q \cdot \rho_{ox})^2 \cdot \bar{c}_{ox}} e^{\left(\frac{E}{R_{Al} \cdot T_{g\infty i}} \right)}. \qquad (7)$$

e) Finally, the mean preexponential factor K is obtained as the mathematical expectation of the set K_i.

2. Results

To evaluate the comparative performance of the modified model (3)-(5) and standard model (1), (2) a set of calculations was made for aluminum particles with R = 3 μm. In all cases, the thermophysical properties of aluminum and its oxide as well as the effective kinetic parameters in the metal oxidation law were taken identical as in [1, 7]. Table 1 shows the values of all parameters used in the calculations.

Table 1: Thermophysical properties and effective kinetic parameters [1, 7].

Material	λ, W/(m·K)	c, J/(kg·K)	ρ, kg/m^3	Reaction	q, MJ/kg	K, m^2/s	E, MJ/kg
Al	225.0	1010.0	2689	$2Al+1.5O_2$	35.6	1.9E-9	2.638
Al_2O_3	40.0	775.0	3970				

The main parameter of interest in the calculations was the ignition delay of aluminum particle placed in the quiescent oxygen gas of different initial temperature (from 975 to 1900 K) at fixed pressure of 1 bar. The ignition delay was defined as time taken for the particle mean temperature to attend the value of 2300 K [8].

Figure 1 compares the predicted ignition delays with the experimental data reported in [8]. The modified model is seen to predict the ignition delays which are by a factor of 1.3 to 1.8 longer than the prediction provided by the standard model and agree somewhat better with experiments [9].

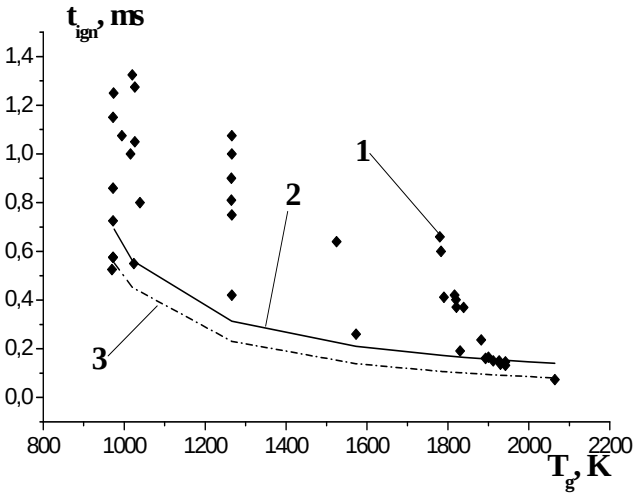

Figure 1: Ignition delays of aluminum particles: 1 - experiments [8], 2 - modified model, 3 - standard model

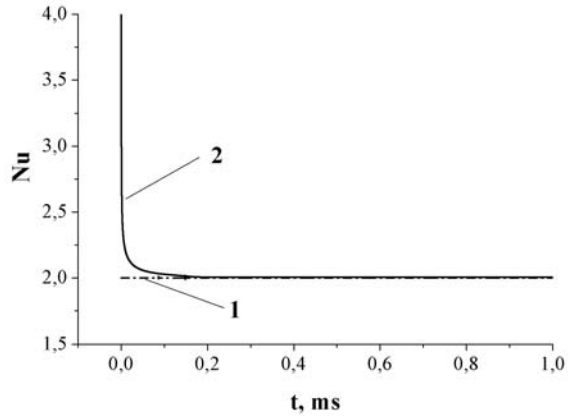

Figure 2: Time history of Nusselt number for aluminum particle (R = 3 μm) at $T_{g\infty}$=1922 K: 1 - standard model; 2 - modified model

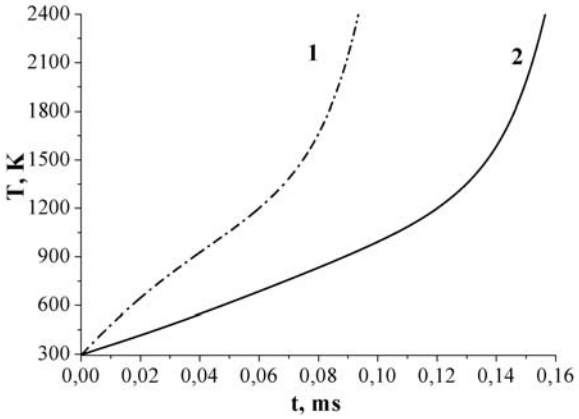

Figure. 3: Time history of the mean particle temperature (R =3 μm) at $T_{g\infty}$=1922 K: 1 - standard model; 2 - modified model

Figure 2 shows the reason for the observed differences in the performances of the modified and standard models. Clearly, the Nusselt number $Nu = 2\alpha R / \lambda_g$ in the modified model (curve 2) is time dependent rather than constant and is always higher than in the standard model (curve 1). The difference in the heat transfer coefficients in the models causes the difference in the particle temperature histories shown in Fig. 3. Note that at high gas temperatures the duration of the transient heating period is comparable with the ignition delay.

In the calculations related to Figs. 1 to 3, the values of the effective kinetic parameters obtained by fitting the standard model with experiments [6] on ignition of aluminum filaments 30-50 μm in diameter were used. As was mentioned above, no account for transient heat transfer and nonuniform temperature distribution inside particles in the standard model can become a reason for inadequate values of the effective kinetic parameters. In view of it, the inverse kinetic problem was solved with the modified model, using the procedure outlined above. For this purpose, the experimental data of [8, 10] (see Table 2) on aluminum particle ignition in oxygen were used.

The resultant set of effective kinetic parameters in the aluminum oxidation law of Eq. (4) is presented in Table 3.

Table 2: Experimental data [8, 10].

№	Diameter, μ m	Temperature of oxygen, K
1	6	689
2	20	1509
3	30	1827
4	40	2003
5	50	2107
6	60	2171
7	70	2212
8	80	2239
9	90	2257
10	100	2269

Table 3: Effective kinetic parameters in the aluminum oxidation law.

Reference	K, m^2/s	E, MJ/kg	$\pm \Delta E$, MJ/kg
[1], [6]	1.90E-9	2.638	-
This paper	0.25E-9	2.660	0.089

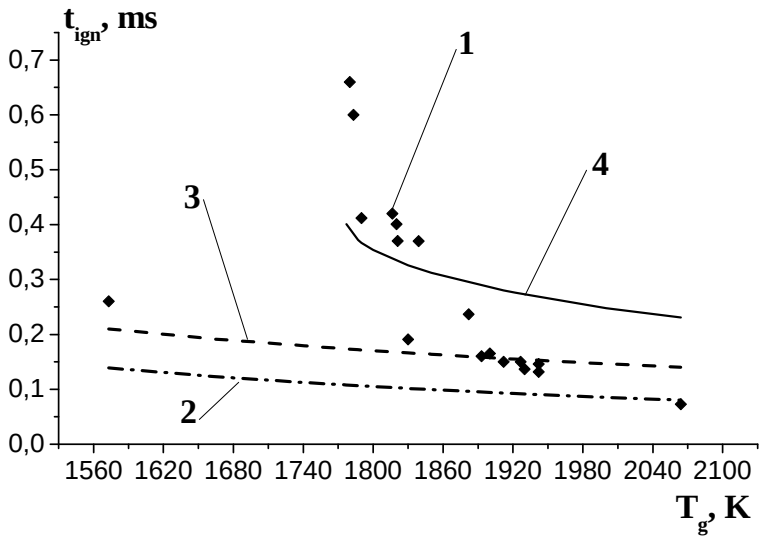

Figure 4: Ignition delay of aluminum particles: 1 - experimental data [9], 2 - modified model with the standard set (E, K), 3 - standard model with the standard set (E, K), 4 – modified model with the new set (E, K)

To show the influence of the effective kinetic parameters on the aluminum particle ignition delay, a set of calculations was made using the modified model with the sets of parameters presented in Table 3. The results of calculations are compared in Fig. 4. Clearly, the new set of effective kinetic parameters provides better agreement of predicted ignition delays and measurements at high temperatures exceeding 1777 K (curve 4). However the use of the new set of effective kinetic parameters at lower temperatures (below 1777 K) results in no ignition.

3. Discussion

The results of calculations provide important implications for the problem of aluminum particle ignition in hot oxygen gas.

First, the modified model provides better particle ignition timing. For example, at oxygen temperatures from 975 to 2064 K for aluminum particles with R = 3 μm the modified model predicts ignition delays which are by a factor of 1.3 to 1.8 longer than the standard model does. These ignition delays agree better with available experimental data.

Second, the modified model provides more adequate values of the effective kinetic parameters.

Third, the difference in the heat transfer conditions and in the effective kinetic parameters exerts a significant effect on the particle ignition timing. For example, for aluminum particles the modified model with the new set of effective kinetic parameters predicts ignition delays which are by a factor of 2.4 to 2.7 longer than those predicted by the standard model at oxygen temperatures ranging from 1777 to 2064 K.

At lower temperatures (below 1777 K) the modified model predicts no ignition. This finding seems to be in line with current understanding of aluminum particle ignition. According to [11] there exist two ignition modes of aluminum particles. At high temperatures (above 1800 K), they ignite in accordance with the parabolic law of Eq. (4). However at low temperatures (below 1800 K) aluminum particles ignite because of structural changes in the solid oxide film which are not taken into account in Eq. (4). Therefore further work will be concentrated on modeling low-temperature ignition by introducing proper changes in the metal oxidation law.

Conclusions

The modified model of aluminum particle ignition has been developed and validated. The model takes into account transient heat transfer between oxidizer gas and particle as well as nonuniform temperature distribution in particle interior. The new set of effective kinetic parameters in the metal oxidation law has been obtained based on fitting the predictions of the modified model with available experimental data. The difference in the ignition delay times predicted by the modified and standard models attains 190% to 250%. The modified model provides better qualitative and quantitative agreement with the high-temperature (above 1800 K) experimental data on aluminum particle ignition. The failure of particle ignition at low temperatures (below 1800 K) predicted by the modified model is explained by no account for structural changes in the solid oxide film of particle surface.

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REFERENCES

1. Fedorov A.V., Kharlamova Yu.V. Ignition of an aluminum particle. Combustion, Explosion, Shock Waves, 2003, No 5, pp. 65-68.
2. Ozerov E.S. Foundations of the theory of gas-dispersed system ignition. Leningrad, LPI Publ., 1978, 69 p.
3. Avdeev K.A., Frolov S.M., Frolov F.S. Rus. J. Chemical Physics, 2006, Vol. 25, No. 11, pp. 17-24.
4. Frolov S.M., Avdeev K.A., Frolov F. S. Effect of transient heat transfer on ignition of solid particles. J. Loss Prevention, 2007, Vol. 20, Issues 4-6, pp. 310-316.
5. Avdeev K.A., Frolov F.S., Borisov A.A., Frolov S.M. Modified model of magnesium particle ignition. In: Nonequilibrium processes: Plasma, Combustion, and Atmospheric Phenomena. Ed. By G.D. Roy, S.M. Frolov, A.M. Starik. Moscow, Torus Press, 2007, p.41.
6. Khaikin B.I., Bloshenko B.N., Merzhanov A.G. On ignition of metal particles. Combustion, Explosion, Shock Waves, 1970, No. 4, pp. 474-488.
7. Grigor'ev Yu.M., Gal'chenko Yu.A., Merzhanov A.G. Investigation of kinetics of high-temperature interaction of aluminum with oxygen by ignition method. Combustion, Explosion, Shock Waves, 1973, No. 2, pp. 191-199.
8. Medvedev A.E., Fedorov A.V., Fomin V.M. Ignition of metal particles in high-temperature flow behind a shock wave. ITAM Preprint No. 33, Novosibirsk, ITAM Publ., 1981.
9. Boyko V.M., Lotov V.V., Papyrin A.N. Ignition of gas suspension of metal powders in reflected shock waves. Combustion, Explosion, Shock Waves, 1989, No. 2, pp. 67-74.
10. Gurevich M.A., Lapkina K.I., Ozerov E.S. Ignition limits of aluminum particles. Combustion, Explosion, Shock Waves, 1970, No. 2, pp. 172-176.
11. Boyko V.M., Poplavski S.V. Ignition of aluminum powders in shock waves. Proc. 4th International conference on aerospace science and technology, Taiwan, 2001. pp. 365-369.4.