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INFLUENCE OF INTERNAL RESPONSE ON THE CRITICAL CONDITIONS OF HEAT AND MASS TRANSFER OF CARBON PARTICLES

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Critical conditions of heterogeneous ignition and extinction of solid and porous carbon particles are determined in a gaseous oxidizer with allowance for concurrent chemical reactions and heat losses due to radiation in relation to inside diffusion of the oxidizer. The influence of the internal response on high- and low-temperature heat and mass transfer processes and the conditions of their implementation is investigated.

Optimum implementation of heat and mass transfer (HMT) processes in combustion of disperse carbon fuels in power plants is a complicated and multifaceted problem. To simplify it, we employ the fact that the concentrations of carbon dust in consumed gas suspensions are rather low and therefore thermal interactions between particles can be neglected [1].

The critical parameters of HMT of an individual spherical carbon particle determine its spontaneous and induced transitions from high-temperature to low-temperature steady-state conditions and vice versa. Due to a decrease in the diameter of a burning particle the heat and mass transfer coefficients of the reactions increase. When they reach critical values, a rapid transition to the low-temperature state occurs, which is characterized by heterogeneous oxidation of the particle. It is established [2] that at a temperature of the reactor walls T_w lower than the gas temperature T_2 heterogeneous combustion is accomplished in a size range limited by the critical diameters of the particle d_{cr} . Its lower limit is determined by d_{ex1} at which its spontaneous or forced extinction occurs, and the upper limit, by the critical diameter d_{ex2} when only forced extinction can take place.

The influence of pore response on the critical parameters of heat and mass transfer is still only partially understood [1, 3]. The main gap here is insufficient understanding of the mechanisms determining the contribution of the internal response in HMT processes and heterogeneous chemical reactions of carbon particles with the surrounding gas, which is manifested in an arbitrary choice of the temperature dependence of the internal diffusion coefficients of the oxidizer [3]. Thus, in [4] without qualitative substantiation of the character of the internal response its influence on the critical conditions of spontaneous and forced ignition and extinction of a porous carbon particle in the case of two concurrent chemical reactions is considered. It is noted that the value of the limiting gas temperature T_2 below which it is impossible to ignite a particle by changing its initial temperature $T_2 \ge 1100$ K although according to experimental results of [5] this occurs at $T_2 = 1000$ K. Such discrepancies are due to neglect of the effect of pore response and can lead to significant errors in analysis of experimental data on the times of reaching a high-temperature regime and other characteristics of combustion of carbon particles in turbulent furnaces.

In the present work it is assumed that on the internal and external particle surfaces two concurrent reactions proceed with formation of gaseous products:

$$C + O_2 \xrightarrow{k_1} CO_2$$
, (I)

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$$2C + O_2 \xrightarrow{k_2} 2CO$$
. (II)

For our investigations we have chosen anthracite of grade ASh, for which sufficiently reliable values of thermokinetic characteristics are available [1]. The assumption of the implicit form of the time dependences of the heat and mass transfer coefficients is based on the fact that the characteristic times of establishing temperature and concentration fields in a gaseous medium are considerably smaller than the time of change of the particle temperature T_1 . Neglect of the gradient of T_1 for the particle is valid since its thermal conductivity is considerably higher than for the gas and evaluation of the Biot number yields a value smaller than 0.2 [6]. Therefore the equations of unsteady-state heat and mass transfer for the particle can be represented in the form

$$\frac{c_{1}\rho_{1}d}{6}\frac{dT_{1}}{dt} = q_{ch} - q_{h}, \quad T_{1} (t=0) = T_{ini};$$

$$\frac{d(d)}{dt} = -\frac{2\rho_{2}}{\rho_{1}} \left[\Omega_{1}k_{1} + \Omega_{2}k_{2}\right] \left[1 + \frac{k_{eff}}{\beta}\right]^{-1} n_{ox\infty}, \quad d(t=0) = d_{ini},$$
(1)

where T_{ini} , d_{ini} are its initial temperature and diameter.

System (1) is valid for describing the HMT processes and the kinetics of chemical transformations of a spherical carbon particle that diminishes with time.

The rate of change of T_1 is determined by the ratio of the rates of heat accumulation due to chemical reactions on the external and internal surfaces of the particle

$$q_{\rm ch} = q_{\rm ch,s} + q_{\rm ch,v} \tag{2}$$

and molecular-convective and radiative heat transfer

$$q_{\rm h} = q_{\rm m.c} + q_{\rm r} \,. \tag{3}$$

The terms in (2) are

$$q_{\rm ch.s} = n_{\rm ox.s} \rho_2 \sum_{i=1}^2 q_i k_i, \ q_{\rm ch.v} = \frac{S_{\rm in}}{S} \langle n_{\rm ox} \rangle_v \rho_2 \sum_{i=1}^2 q_i k_i,$$

 $k_i = k_{0i} \exp(-E_i/R/T_1)$ are the preexponentials (i = 1, 2) for reactions (I) and (II).

The mean oxygen concentration $\langle n_{ox} \rangle_v$ is determined from the solution of the internal-diffusion equation of the particle [1] $\langle n_{ox} \rangle_v = 6n_{ox.s}k_{in}/(k_1 + k_2)F_{in}d$, where

$$k_{\rm in} = \frac{2D_{\rm in}}{d} \left(\text{Se cth Se} - 1 \right), \tag{4}$$

Se =
$$\frac{d}{2} \left[\frac{(k_1 + k_2) F_{\text{in}}}{D_{\text{in}}} \right]^{1/2}$$
; $F_{\text{in}} = \frac{6S_{\text{in}}}{\pi d^3}$.

From the boundary condition $\beta(n_{ox\infty} - n_{ox.s})\rho_2 = n_{ox.s}\rho_2 k_{eff}$ we find that

$$n_{\rm ox.s} = \frac{n_{\rm ox\infty}}{1 + k_{\rm eff}/\beta},$$
(5)

 $k_{\text{eff}} = k_1 + k_2 + k_{\text{in}}; \beta = \text{Nu}D_2/d$. Since the particle is considered to be fixed, Nu = 2. Therefore the rate of heat release due to external and internal chemical reactions is of the form

(2)



Fig. 1. Time dependences of the temperatures (a), their derivatives (b), particle diameters (c), rate constants of chemical reactions and mass transfer coefficient (d) at $n_{0x\infty} = 0.23$, $T_2 = T_w = 1400$ K, $d_{ini} = 505 \,\mu\text{m}$; 1) solid ($D_{in} = 0$); 2) porous ($D_{in} = 7.5 \cdot 10^{-8}$) carbon particles; 3) experimental result [5].

$$q_{\rm ch} = n_{\rm ox\,\infty} \,\rho_2 \, \frac{q_1 k_1 + q_2 k_2}{k_1 + k_2} \left[\frac{1}{\beta} + \frac{1}{k_{\rm eff}} \right]^{-1} \,. \tag{6}$$

In composing the equation for the change in the carbon particle diameter with time (the second equation in (1)), we have taken into account chemical transformations of carbon only over its external surface [7]; $\Omega_1 = 3/8$, $\Omega_2 = 3/4$ are the stoichiometric coefficients of reactions (I), (II).

Molecular-convective heat transfer between the particle and the surrounding gas is described by the Newton-Reichman law

$$q_{\rm m.c} = \alpha \left(T_1 - T_2 \right),$$
 (7)

 $\alpha = \mathrm{Nu}\lambda_2/d.$

The density of the radiative heat flux is determined as

$$q_{\rm r} = \varepsilon \sigma \left(T_1^4 - T_{\rm w}^4 \right) \,, \tag{8}$$

where $\varepsilon = \text{const} [7]$.

We have also used temperature dependences of the density ρ_2 and the thermal conductivity λ_2 of the medium in the form $\rho_2 = \rho_{20}(T_0/T_*)$, $\lambda_2 = \lambda_{20}(T_*/T_0)^{3/4}$, where T_* is some temperature $T_* = (T_1 + T_2)/2$ [2, 4].

The values of the kinetic and thermophysical parameters are chosen as follows: $q_1 = 10.125 \cdot 10^6$, $q_2 = 6.843 \cdot 10^6$ J/kg O₂; $k_{01} = 4.5 \cdot 10^4$, $k_{02} = 13 \cdot 10^4$ m/sec; $E_1 = 140,030$, $E_2 = 154,000$ J/mole [1]; R = 8.31 J/(mole·K); 1) $D_{in} = 0, 2$) $D_{in} = 7.5 \cdot 10^{-8}$ [3], 3) $D_{in} = D_{in0}(T_*/T_0)^2$ [1] m²/sec; $F_{in} = 3 \cdot 10^7$ m⁻¹ [3]; the constants $D_{in0} = 2.435 \cdot 10^{-4}$ m²/sec, $\lambda_{20} = 0.0819$ W·m/K, and $\rho_{20} = 0.277$ kg/m³ [2] are determined for $T_0 = 1273$ K; $c_2 = 1110$ J/(kg·K); $\varepsilon = 0.78$; $\sigma = 5.67 \cdot 10^{-8}$ W/(m²·K⁴); the density and heat capacity of the particle $\rho_1 = 1440$ kg/m³; $c_1 = 947$ J/(kg·K) correspond to anthracite, grade ASh [1].

The system of differential equations describing the heat and mass transfer processes with a solid particle [7] is similar to (1) but with the exception that $k_{in} = 0$, i.e., in the expressions (5), (6) $k_{eff} = k_1 + k_2$.



Fig. 2. Time dependences of the temperatures (a) and diameters of carbon particles (b) at $n_{0x\infty} = 0.23$, $T_2 = T_w = 1140$ K, $d_{ini} = 100 \,\mu\text{m}$; 1) solid particle $(D_{in} = 0)$; 2, 3) porous particles $(D_{in} = 7.5 \cdot 10^{-8} \text{ and } D_{in} = D_{in0} (T_*/T_0)^2$, respectively).

The characteristics of heterogeneous ignition, combustion, and extinction of the carbon particle can be described by the dependences $T_1(t)$, $dT_1/dt(t)$, and d(t) (Fig. 1a, b, c). An induction period indicating the time of attainment of a high-temperature regime by the HMT process is determined by the first maximum of the function $dT_1/dt(t)$, i.e., fulfillment of the conditions $d^2T_1/dt^2 = 0$, $d^3T_1/dt^3 < 0$ (the point Ig). Then the stage of quasistationary particle combustion proceeds, where $dT_1/dt \approx 0$ (or $q_{ch} \approx q_h$) in a diffusion regime since $k_{eff} >> \beta$ (Fig. 1d). After reaching the maximum value T_{1M} , the combustion temperature decreases drastically and when the particle diameter attains its critical value d_{ex} , the particle undergoes spontaneous extinction (the point Ex), when $d^2T_1/dt^2 = 0$, $d^3T_1/dt^3 > 0$.

In Fig. 2 the conditions for solving system (1) are chosen so as to exclude extinction of a solid particle. Account for the temperature-dependent internal diffusion coefficient D_{in} allows a considerable decrease in the induction period of ignition [8] and an increase in the combustion time [9] of the porous carbon particle (curves 3) due to strong activation of the internal HMT processes.

An analysis of the dependences $q_{ch}[T_1(t)]$ and $q_h[T_1(t)]$ makes it possible to reveal regularities of spontaneous extinction of the particle determined by the mechanism of feedback of the change in the temperature T_1 with the heat-mass transfer processes and the kinetics of the heterogeneous reactions. With allowance for the pore response the value of d_{ex} decreases substantially (see Fig. 1c), which makes it possible to improve the description of experimental data [5], although heat losses due to radiation lead to an increase in the critical diameter in the case of spontaneous extinction.

Expressions for the critical parameters are obtained as a result of an analysis of the condition of steadiness and stability of the HMT regimes:

$$dT_1/dt = 0$$
, $d^2T_1/dt^2 = 0$,

which with a finite value of the particle diameter is equivalent to

$$q_{\rm ch} = q_{\rm h} , \ \frac{\partial q_{\rm ch}}{\partial T_1} = \frac{\partial q_{\rm h}}{\partial T_1} .$$
 (9)

For the model of a solid particle $(k_{eff} = k_1 + k_2)$ the first equality of system (9) corresponds to a quadratic equation for the particle diameter:

$$Ad^2 + Bd + C = 0,$$

where

$$A = q_{\rm r} \sum_{i=1}^{2} k_{i}; \ B = \left(q_{\rm r} + c_{2} \rho_{2} \left(T_{1} - T_{2} \right) \sum_{i=1}^{2} k_{i} - \rho_{2} n_{\rm ox\infty} \sum_{i=1}^{2} q_{i} k_{i} \right) 2D_{2};$$



Fig. 3. Dependences of the diameters d of solid (2) and porous (2, 3) carbon particles on their stationary temperatures T_{1st} (u.s denotes unsteady states) at $n_{0x\infty} = 0.23$, $T_2 = T_w = 1400$ K: 1) $D_{in} = 0$; 2) $7.5 \cdot 10^{-8}$; 3) $D_{in} = D_{in0}(T_*/T_0)^2$.

$$C = \frac{4\lambda_2^2 (T_1 - T_2)}{c_2 \rho_2}$$

Its solutions are

$$d_{1,2} = \frac{\sqrt{\Delta} \pm B}{2A},\tag{10}$$

where the discriminant is $\Delta = B^2 - 4AC$.

In determining the dependence $d(T_{1st})$ in the model of a porous particle $(k_{eff} = k_1 + k_2 + k_{in})$ it is necessary to take into account $k_{in} = k_{in}(d)$, which stipulates the implicit character of the sought function.

An analysis of $d(T_{1st})$ with account for the condition of stability loss of the stationary regime

$$\frac{\partial q_{\rm ch}}{\partial T_1} > \frac{\partial q_{\rm h}}{\partial T_1} \tag{11}$$

allows determination of the dependence of the critical particle diameter d_{cr} characterizing heterogeneous ignition on its temperature T_{ini} and investigation of the influence of the particle size on its combustion temperature T_c . The critical diameter values are determined by the extrema of the curve $d(T_{1st})$ corresponding to (9).

The branch connecting the points of ignition Ig and extinction Ex (the short dashed line in Fig. 3) describes the influence of the initial temperature of the particle on the critical ignition diameter $d_{ig}^*(T_{ini})$. The curve running from larger sizes through the points M and Ex determines the influence of the particle diameter on its combustion temperature $T_c(d)$ in the quasisteady regime.

Heat losses due to radiation at $T_w < T_2$ lead to the appearance of the upper range of the critical diameters, whose boundaries correspond to additional extrema on the curve $d(T_{1st})$. When the wall temperature increases and approaches T_2 , they degenerate into an inflection point, and there a transition to the combustion branch can occur for a particle of any large diameter.

After ignition of a particle of large size, the decrease in the latter in the course of combustion is accompanied by an increase in T_1 (the arrows in Fig. 3 are given for one case) since the heat release q_{ch} increases due to the coefficient β to a larger degree than the heat losses by heat conduction $q_{m.c}$ due to α with a considerable contribution of q_r . The combustion temperature, passing through a maximum (the point M) where $\partial T_{1st}/\partial(d) = 0$ or $\partial q_{ch}/\partial(d)$ $= \partial q_h/\partial(d)|_{T_{1st}}$ is fulfilled, reaches the critical value T_{1ex} to which the particle extinction diameter $d_{ex}(T_1)$ corresponds. This ambiguous course of the heat and mass transfer processes is illustrated by the hysteresis character of $T_{1st}(d)$.



Fig. 4. Critical values of the diameters (a) and temperatures (b) of solid and porous carbon particles versus the medium temperature at $n_{\text{ox}\infty} = 0.23$, $T_2 = T_w$: 1) $D_{\text{in}} = 0$; 2) $7.5 \cdot 10^{-8}$; 3) $D_{\text{in}} = D_{\text{in}0}(T_*/T_0)^2$. d_{cr} , μ m

Although the dependences $d(T_{1st})$ are similar for the three cases, their slight difference lies in some weakening of the ambiguity (approach of the diameters d_{ig} and d_{ex}) in porous particles and the increase in their combustion temperatures caused by additional heat release due to the internal response. The decrease in the critical sizes of the particles to 1-2 mm for ignition and extinction in case 3 is most pronounced, which can be attributable to the high-rate pore diffusion with D_{in} decreasing by a factor of 10^3-10^4 [1].

Figure 4 depicts the critical diameters d_{cr} and temperatures T_{1cr} as a function of the gas temperature T_2 . The ignition and extinction branches correspond to the "displacement trajectory" of the extrema Ig and Ex in Fig. 3 with change in T_2 (together with T_w) in a temperature range of 300-2000 K.

For particles having different internal structures these dependences are of the same form, with the exception of case 3 in Fig. 4. The points γ_1 and γ_2 connect the ignition (Ig) and extinction (Ex₁, Ex₂) branches so that at the given gas temperature the boundary conditions $[d_{ex1}, d_{ex2}]$, $[T_{1ex2}, T_{1ex1}]$ and $[d_{ex1}, d_{ig}]$, $[T_{1ig}, T_{1ex1}]$ are formed for the transitions from low-temperature to high-temperature regimes and vice versa. With increase in the gas temperature the difference in the values of d_{ig} and d_{ex} , T_{1ig} and T_{1ex} decreases, and at the points $\gamma_{1, 2}$ the critical conditions of ignition and extinction degenerate. This corresponds to the approach of the extrema Ig and Ex on the curve $d(T_{1st})$ and their transformation into an inflection point. The parameters of degeneracy of the critical conditions are determined by the equalities

$$\frac{\frac{\partial d}{\partial T_{1st}} = 0}{\frac{\partial^2 d}{\partial T_{1st}^2} = 0} \quad \text{or} \quad \frac{\frac{\partial^n q_{ch}}{\partial T_1^n}}{\frac{\partial T_1^n}{\partial T_1^n}} = \frac{\frac{\partial^n q_{h}}{\partial T_1^n}}{\frac{\partial T_1^n}{\partial T_1^n}}, \quad n = 0, 1, 2.$$

The functions $d_{cr}(T_2)$ and $T_{1cr}(T_2)$ restrict the regions of thermodynamic states where particles can ignite if their initial temperature acquires a value within the limits $T_{1ig} < T_{ini} < T_{1ex1}$ and $T_{1ex2} < T_{ini} < T_{1ex1}$. With the initial diameters $d_{ini} < d_{ex1}$ and $d_{ini} > d_{ex2}$ only the low-temperature regime is implemented, while in the region to the right of the curve $d_{ig}(T_2)$ (Fig. 4a) the transition to the high-temperature state occurs independently of the initial temperature of the particle.

The upper parts of the curve $d_{cr}(T_2)$ are determined by the ratios of the heat loss due to radiation and the heat release due to chemical reactions. If $T_{1ig} < T_{ini} < T_2$, then $q_r > q_{ch}$, while at $T_{ini} > T_2$, $q_{ch} > q_r$. At the point γ_2 the gas temperature is determined mainly by q_r . For small diameters (the lower part of the curve $d_{cr}(T_2)$) $q_r < q_{ch}$ and the critical values of the initial temperature are higher than the gas temperatures, $T_2 < T_{1ig} < T_{ini} < T_{1ex1}$.

To determine the limiting critical gas temperature T_{2C} , we use the steadiness and stability conditions of the stationary regime

$$q_{\rm ch}(d, T_1, T_2) = q_{\rm h}(d, T_1, T_2),$$

$$\frac{\partial q_{\rm ch}}{\partial d} + \frac{\partial q_{\rm ch}}{\partial T_1} \frac{\partial T_1}{\partial d} + \frac{\partial q_{\rm ch}}{\partial T_2} \frac{\partial T_2}{\partial d} = \frac{\partial q_{\rm h}}{\partial d} + \frac{\partial q_{\rm h}}{\partial T_1} \frac{\partial T_1}{\partial d} + \frac{\partial q_{\rm h}}{\partial T_2} + \frac{\partial T_2}{\partial d} \cdot$$

The point C is determined by the equality $\partial T_{2cr}/\partial d = 0$, for the curves $d_{cr}(T_2)$ and $T_{1cr}(T_2)$ the equality $\partial q_{ch}/\partial T_1 = \partial q_h/\partial T_1$ is fulfilled, and therefore at $\partial T_1/\partial (d) \neq \infty$ we obtain $\partial q_{ch}/\partial d = \partial q_h/\partial d$. Then, substituting the expressions for q_{ch} and q_h , we write

$$\frac{q_{\rm r}}{q_{\rm m.con}} = \frac{1 + \frac{\beta}{k_{\rm eff}}}{\Pi} - 1$$

where $\Pi = 1 + (k_{in}D_2/k_{eff}^2)(k_{in}/D_{in} + 4/d - F_{in}(k_1 + k_2)/k_{in}$ is the correction for porosity. For a solid particle $\Pi = 1$ since $k_{in} = 0$, and the ratio of heat losses due to radiation and the molecular-convective mechanism acquires the simpler form [2]

$$\frac{q_{\rm r}}{q_{\rm m.con}} = \frac{\beta}{k_1 + k_2}.$$
(12)

For porous particle 2 the limiting temperature T_{2C} is considerably lower than for the solid particle. The temperature difference $|T_{2C} - T_{1C}|$ in case 2 decreases, as compared to case 1, from three to two distinguishing intervals of RT_2^2/E_1 , which is in better agreement with experimental data [5]. Among the distinctive features of the curves in Fig. 4 we would like to point out that the point C for porous particles 3 and its corresponding temperature T_{2C} are absent under real conditions. In actuality this implies that extinction of such a particle is impossible just by decreasing the gas temperature. Such a decrease in T_{2C} is obviously explained by enhancement of the processes of pore response with increase in the internal-diffusion coefficient since in a first approximation k_{in} is proportional to $\sqrt{D_{in}}$ (4).

Thus, account for the internal diffusion of oxygen and the chemical response inside a carbon particle makes it possible to reveal a whole series of new distinctive features of its heterogeneous combustion and to evaluate more correctly their influence on the basic parameters of stable and critical regimes of heat and mass transfer processes.

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NOTATION

 c_1 , specific heat capacity of carbon (anthracite ASh), J/(kg·K); c_2 , specific heat capacity of the medium (air), J/(kg·K); d, diameter of the carbon particle, m; D_2 , external-diffusion coefficient of oxygen, m²/sec; D_{in} , internal-diffusion coefficient of oxygen, m^2/sec ; D_{in0} , internal-diffusion coefficient of oxygen at $T_* = 1273$ K, m^2 /sec; E_1 , E_2 , activation energies of the chemical reactions, J/mole; F_{in} , specific area of the inner surface of the particle, m^{-1} ; k_1 , k_2 , rate constants of the chemical reactions, m/sec; k_{01} , k_{02} , preexponentials of the rate constants of the chemical reactions, m/sec; kin, rate constant of the internal chemical reactions, m/sec; keff, effective rate constant of the chemical reactions, m/sec; $n_{0x\infty}$, relative oxidizer concentration in the medium; $n_{0x.s}$, relative oxidizer concentration at the outer surface of the particle; Nu, Nusselt number; q_1 , q_2 , mass thermal effects of the reactions, J/kgO₂; q_r , specific radiative heat flux, W/m²; $q_{m.c}$ specific flux of molecular-convective heat transfer, W/m^2 ; q_h , total heat flux due to radiative and molecular-convective heat transfer, W/m^2 ; q_{ch} , specific heat flux due to the chemical response, W/m^2 ; $q_{ch.s}$, specific heat flux due to the chemical response on the outer surface of the particle; q_{chv} , specific heat flux due to the chemical response on the inner surface of the particle, W/m²; R, universal gas constant, J/(mole \cdot K); S, outer surface area of the particle, m²; S_{in}, inner surface area of the particle, m^2 ; Se, Semenov number; t, time, sec; T_1 , particle temperature, K; T_2 , gas temperature, K; T_w , wall temperature, K; T_{1st}, stationary temperature of the particle, K; α , heat transfer coefficient, W/K; β , mass transfer coefficient, m/sec; ε , integrated emissivity of the carbon particle (black emissivity); λ_2 , thermal conductivity of the medium

(air), $W \cdot m/K$; λ_{20} , thermal conductivity of the medium (air) at $T_* = 1273$ K, $W \cdot m/K$; ρ_1 , density of carbon (anthracite ASh), kg/m³; ρ_2 , density of the medium (air), kg/m³; ρ_{20} , density of the medium (air) at $T_* = 1273$ K, kg/m³; σ , Stefan-Boltzmann constant, $W/(m^2 \cdot K^4)$. Subscripts: int, internal; r, radiative; cr, critical; m.con, molecular-convective; in, initial; ox, oxidizer; ex, extinction; w, reactor walls; h, heat; eff, effective; st, stationary; ∞ , at an infinite distance.

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