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Critical Condition for the Mass Burning Rate Constant of a Carbon Particle Activated; Comparisons with Experimental Results

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Abstract

Relevant to the activation of solid fuel particles, critical condition for the mass burning rate constant, above which particle combustion can successfully be accomplished has been obtained. Use has been made of the asymptotics, with focusing on the temporal variation of the particle temperature in the plateau stage. It has been confirmed that there exists a useful parameter, consisting of particle diameter and combustion rate, which mainly depends on the ambient temperature. It is seen that the critical condition separates regions, upper half of which corresponds to that for particle combustion with surface reactions activated. In addition, existence of the cut-off temperature, with respect to the ambient temperature, has been confirmed, above which the particle combustion can only be accomplished. Appropriateness and/or usefulness of this critical condition has further been examined, by use of such experimental data in the literature as are reported to burn in the quiescent environment. Experimental data used are those from semi-anthracite to low-rank coal char, as well as petroleum coke. A fair degree of agreement has been demonstrated, indicating that excessively smaller particles are unfavorable to the particle combustion. This fair agreement further suggests that the formulation, from which the critical condition has been derived, has captured the essential feature of the particle combustion that has not been elucidated. It has been confirmed again that the reduction in particle size does not necessarily favor the particle combustion, which is inconsistent with the premise prevailed.

Keywords: heterogeneous combustion; char combustion; critical condition for particle activation; critical particle size; cut-off temperature.

Nomenclature

A	reduced surface Damköhler number
B	frequency factor
c	specific heat
c_p	specific heat of gas at constant pressure
D	diffusion coefficient
E	activation energy

m mass consumption rate

$\dot{m}$ mass burning (or combustion) rate in dimensional form

p pressure

q heat of combustion

r particle radius

T temperature

Ta activation temperature

t time

W molecular weight

Y mass fraction

Greek Symbols

α heat loss parameter

β conventional transfer number

δ product(CO_2)-to-carbon mass ratio

ε emissivity

κ combustion rate in the plateau stage

θ perturbed temperature

λ thermal conductivity

ν stoichiometric coefficient

ρ density

σ_{SB} Stefan-Boltzmann constant

τ nondimensional time

Subscripts

C carbon

cut-off cut-off temperature

cr critical condition

O oxygen or $C\text{-}O_2$ surface reaction

P carbon dioxide or $C\text{-}CO_2$ surface reaction

s surface

∞ ambience

0 reference

Superscripts

~ nondimensional or stoichiometrically weighted

1. Introduction

Because of the practical importance, extensive research has been conducted for the carbon combustion, not only experimentally but also theoretically/numerically, relevant to coal/char utilization and/or aerospace applications with respect to the propulsion aiming at utilizing high-energy-density fuels. Although some of the comprehensive reviews [1-7] have already summarized accomplishment hitherto obtained, there still remain several problems indispensable for understanding basic nature of the combustion, because of complexities involved, with commanding fundamental interests.

A problem relevant to sizes of solid fuel particles is one of them. Nonetheless, much of the understanding has been based on the d^2-law, so that there prevails a premise that the burning time would be reduced with decreasing particle size. To the contrary, as has been confirmed [4, 6, 8] not only experimentally [9] but also numerically [10], particles with excessively smaller sizes are unfavorable to the ignition, because of the heat loss, enhanced as the particle size drops. As for the study from the analytical point of view, no one had applied the Semenov thermal-explosion theory to this system, as pointed out [4], until there appeared a work, relevant to the particle activation.

In that work [11], an attempt has been conducted to obtain the critical condition for a particle to be activated, by use of the asymptotics, with focusing on the plateau stage, characterized by a slight temperature-rise just after the initial, rapid heating [2, 4, 6, 8]. It has succeeded in obtaining the critical particle size, below which surface reaction ceases to be activated, due to the heat loss, with confirming the fact that the temporal variation of the particle temperature can be determined by the heat-release and -loss rates, just like that for the spontaneous ignition of the gaseous reactive mixtures, studied in the 1930's [say, 12, 13]. A comprehensive parameter that can fairly correlate dominant parameters with the ambient temperature has also been obtained. Furthermore, a fair degree of agreement has been demonstrated between experimental and theoretical results for graphite [11] and/or coal char [14, 15] particles, for which kinetic parameters are relatively well-known and/or easily evaluated by use of the experimental results for the combustion rate [16-28]. Besides, an attempt to obtain another critical condition with respect to the combustion rate [29] has further been conducted, which results in presenting another comprehensive parameter.

The present study is intended to shed more light on the combustion of carbonaceous solid fuels, by examining other critical condition that can be obtained by combining these two comprehensive parameters. Importance of the cut-off temperature, only above which the particle combustion can successfully be activated, has further been examined.

In the following, formulation relevant to the critical condition is first presented, based on the conventional assumptions for the combustion of small carbon particles (<100 μm). Then, by using representative kinetic parameters for the surface reactions evaluated in the literature, results of numerical calculations are to be presented, for understanding general trend. Experimental comparisons are then conducted, presenting that the particle activation can only be accomplished when the parameter newly introduced is higher than the critical value, as well as satisfying another restriction with respect to the cut-off temperature. Finally, concluding remarks are made.

2. Formulation

2.1 Model Definition

The problem of interest is the combustion of a spherical carbon particle (radius r_s, surface temperature T_s, density ρ_C, and specific heat c_C) in a quiescent environment (temperature T_∞, oxygen mass-fraction $Y_{O,\infty}$, carbon dioxide mass-fraction $Y_{P,\infty}$, density ρ_∞, and specific heat c_p). The major reactions considered are the surface C-O_2 and C-CO_2 reactions and the gas-phase CO-O_2 reaction. The surface $C+O_2 \rightarrow CO_2$ reaction is excluded [30] because our primary concern is the combustion at high surface temperatures, say, higher than 1000 K. The choice of this set of chemical reactions has already been justified by conducting experimental comparisons, in which a fair degree of agreement has been demonstrated between predicted and experimental results, not only in the quiescent environment [11, 14, 15] but also in the stagnation flowfield [31, 32]. Crucial assumptions introduced are conventional, constant property assumptions with unity Lewis number, constant average molecular weight throughout the combustion field, constant value of the product of density ρ and diffusivity *D*, one-step overall irreversible gas-phase reaction, and first-order surface reactions. Surface characteristics, such as porosity and internal surface area, are grouped into the frequency factors for the surface reactions, in order not to probe into specific details of the physical processes.

Since the solid in general possesses great inertia, because of the significant disparity between solid and gas, such properties at the surface as are the regression rate, species concentrations, and temperature, can change at rates much slower than those of the gas-phase transport processes. Therefore, under an assumption of this quasi-steadiness in the gas phase, formulation has been conducted, as described [33-39], so that only the final solution is presented herein.

2.2 Governing Equations

Temporal variations of the particle temperature and size are governed by [34-37, 40]

$$\left(\rho_C \frac{4}{3}\pi r_s^3\right) c_C \frac{dT_s}{dt} = -4\pi r_s \lambda \frac{\tilde{m}}{\beta}(T_s - T_\infty) + m(c_C - c_p)(T_s - T_0)$$
$$+ m\, q_{s,0} - 4\pi r_s^2 \varepsilon \sigma_{SB}(T_s^4 - T_\infty^4) \;, \qquad (1)$$

$$\frac{d}{dt}\left(\rho_C \frac{4}{3}\pi r_s^3\right) = -m. \qquad (2)$$

In the above, m is the mass consumption rate (kg/s), β the Spalding transfer number, λ the thermal conductivity, and q_s the heat of combustion for the surface reaction. Here, use has been made of the assumption that there prevails the frozen gas-phase chemistry for such small particles as are less than ca. 100 μm [39]. Note here that the combustion rate, nondimensional, defined as $\tilde{m} = m/(4\pi\rho_\infty D_\infty r_s)$, can also be expressed as $\tilde{m} = \ln(1+\beta)$. As for the combustion rate, dimensional, defined as $\dot{m} = m/(4\pi r_s^2)$, it has been presented as

$$\dot{m} = \frac{\rho_\infty D_\infty}{r_s} \ln(1+\beta) \quad , \tag{3}$$

with the transfer number, explicitly expressed as [47]

$$\beta \approx \left(\frac{A_{s,O}}{1+A_{s,O}}\right)\left(\frac{2W_C}{W_O} Y_{O,\infty}\right) + \left(\frac{A_{s,P}}{1+A_{s,P}}\right)\left(\frac{W_C}{W_P} Y_{P,\infty}\right) \ ; \tag{4}$$

$$A_{s,i} = \frac{B_{s,i}\, r_s}{D_\infty}\left(\frac{T_\infty}{T_s}\right)\exp\left(-\frac{Ta_{s,i}}{T_s}\right) \qquad (i = \mathrm{O, P})\ , \tag{5}$$

in the ***Frozen mode***, the most plausible situation for small particles, albeit approximate. As for the kinetic parameters for the surface reactions, several sets of them listed in Table 1 have been reported [14, 15, 29] to be appropriate in evaluating combustion rates of representative coals and/or coal chars [19-24], in spite of the conjecture that the combustion rates would strongly depend on types of coals.

Table 1 Kinetic parameters for representative coals and coal chars [11,14, 15].

Type	C-O_2 reaction		C-CO_2 reaction		Remarks
	$B_{s,O}$ (m/s)	$E_{s,O}$ (kJ/mol)	$B_{s,P}$ (m/s)	$E_{s,P}$ (kJ/mol)	
P	4.1×10^6	179	1.1×10^8	270	Called with reference to Petroleum cokes
A	2.2×10^6	180	6.0×10^7	269	Called with reference to Anthracite
L	2.2×10^7	180	6.0×10^8	269	Called with reference to Low-rank coals
S	6.6×10^5	180	1.8×10^7	269	Called with reference to coal chars with

					suppressed reactivity

2.3 Critical Condition with Respect to the Particle Diameter [11]

There exists the critical condition [11, 14, 15] above which particle combustion can successfully be accomplished. As well-recognized [4, 6, 8, 10, 40] in the particle combustion, a particle experiences rapid heating, which is followed by a gradual heating, prior to the activation of surface reactions. With focusing on this temporal variation in the plateau stage, the critical condition below which surface reactions cease to be activated has been obtained [11] as,

$$(2r_{\mathrm{s}}) \geq (2r_{\mathrm{s}})_{\mathrm{cr}} = \frac{(\tilde{m}/\beta)}{\frac{e\,B_{\mathrm{s},0}Y_{0,\infty}}{D_\infty}\frac{W_{\mathrm{C}}q_{\mathrm{s},0}}{W_{\mathrm{O}}c_pT_\infty}K\frac{Ta_{\mathrm{s},0}}{T_\infty}\exp\left(-\frac{Ta_{\mathrm{s},0}}{T_\infty}\right) - \frac{2\varepsilon\sigma_{\mathrm{SB}}T_\infty^3}{\rho_\infty D_\infty c_p}} \quad , \tag{6}$$

which is yielded from the perturbed energy equation

$$\frac{d\theta}{d\tau} = e^\theta - \alpha\theta, \tag{7}$$

derived with introducing a perturbed temperature θ, defined as $T_{\mathrm{s}} = T_\infty\{1 + \epsilon\theta + O(\epsilon^2)\}$ with $\epsilon = T_\infty/Ta_{\mathrm{s},0}$, and the nondimensional time τ. In the above, we have

$$\alpha = \frac{12}{(2r_s)^2}\frac{1}{\Delta}\frac{\rho_\infty}{\rho_{\mathrm{C}}}D_\infty\left(\frac{\tilde{m}}{\beta} + \frac{2r_s}{\rho_\infty D_\infty c_p}2\varepsilon\sigma_{\mathrm{SB}}T_\infty^3\right)\frac{1}{c_{\mathrm{C}}/c_p} \quad , \tag{8}$$

$$\tau = t\cdot\Delta\,, \tag{9}$$

$$\frac{1}{\Delta} = \frac{2r_{\mathrm{s}}}{12B_{\mathrm{s},0}Y_{0,\infty}}\frac{\rho_{\mathrm{C}}}{\rho_\infty}\frac{W_{\mathrm{O}}c_pT_\infty}{W_{\mathrm{C}}q_{\mathrm{s},0}}\frac{c_{\mathrm{C}}/c_p}{K}\frac{T_\infty}{Ta_{\mathrm{s},0}}\exp\left(\frac{Ta_{\mathrm{s},0}}{T_\infty}\right) \quad , \tag{10}$$

$$K = 1 + \left(\frac{c_{\mathrm{C}}}{c_p} - 1\right)\frac{c_pT_\infty}{q_{\mathrm{s},0}}\left(1 - \frac{T_0}{T_\infty}\right) \quad . \tag{11}$$

Note in Eq. (7) that the temporal rise is governed by the heat-release and -loss rates and that the particle temperature only rises when $\alpha < e$, as explained [12, 13]. We also have the comprehensive parameter, $(2r_{\mathrm{s}})Y_{0,\infty}(p_\infty/p_0)$, useful in correlating dominant parameters with the ambient temperature.

2.4 Critical Condition with Respect to the Combustion Rate [29]

As for the combustion rate, in the same manner, we have

$$\dot{m} \geq (\dot{m})_{\mathrm{cr}} = e\, B_{\mathrm{s,0}} Y_{\mathrm{O},\infty} \rho_\infty \frac{2W_{\mathrm{C}}}{W_{\mathrm{O}}} \exp\left(-\frac{Ta_{\mathrm{s,0}}}{T_\infty}\right) \ , \tag{12}$$

which is yielded from its temporal variation in the plateau stage, expressed as

$$\frac{d\dot{m}}{d\tau} = \left[B_{\mathrm{s,0}} Y_{\mathrm{O},\infty} \rho_\infty \frac{2W_{\mathrm{C}}}{W_{\mathrm{O}}} \exp\left(-\frac{Ta_{\mathrm{s,0}}}{T_\infty}\right)\right] e^{2\theta}\left(1 - \alpha\theta e^{-\theta}\right) \ . \tag{13}$$

Since $\theta e^{-\theta}$ has the maximum at $\theta = 1$, we always have a monotonic increase in the combustion rate, when $\alpha < e$, because $d\dot{m}/d\tau > 0$. That is, it is suggested that particles anticipated to burn are required to have the combustion rate that exceeds this critical value, in the course of the particle combustion. Otherwise, no more particle activation is anticipated. Another comprehensive parameter, $\dot{m}/\left[Y_{\mathrm{O},\infty}(p_\infty/p_0)\right]$ has been found, which also depends on the ambient temperature T_∞.

2.5 Critical Condition with Respect to the Mass Burning Rate Constant

Thus far, two kinds of comprehensive parameters have been obtained. By combining these two, we have the other, as expressed,

$$(2r_{\mathrm{s}})\dot{m} \geq [(2r_{\mathrm{s}})\dot{m}\,]_{\mathrm{cr}} = \frac{(\tilde{m}/\beta)\rho_0 D_0 (T_\infty/T_0)^{0.75}}{\frac{1}{2}\frac{q_{\mathrm{s,0}}}{c_p T_\infty} K \frac{Ta_{\mathrm{s,0}}}{T_\infty} - \frac{2\varepsilon\sigma_{\mathrm{SB}} T_\infty^3}{\rho_\infty Y_{\mathrm{O},\infty} c_p} \frac{1}{e\, B_{\mathrm{s,0}}} \frac{1}{2W_{\mathrm{C}}/W_{\mathrm{O}}} \exp\left(\frac{Ta_{\mathrm{s,0}}}{T_\infty}\right)} \ , \tag{14}$$

which also depends on the ambient temperature.

Relevant to this comprehensive parameter, we should note the relation as

$$(2r_{\mathrm{s}})\dot{m} = -\frac{\rho_{\mathrm{C}}}{4}\frac{d}{dt}(2r_{\mathrm{s}})^2 \ , \tag{15}$$

yielded from Eq. (2), by use of the definition of $\dot{m}$. It is suggested that this comprehensive parameter is nothing but the mass burning rate constant in the diffusionally controlled regime. By use of this comprehensive parameter, we can easily determine whether the combustion rate measured is that in or near the diffusion controlled regime or not.

In addition, it may be informative to note that the critical condition of this parameter, $[(2r_\mathrm{s})\dot{m}]_\mathrm{cr}$, increases in proportional to $T_\infty^{2.75}$ at high temperatures, at which dependence on the activation temperature $Ta_{\mathrm{s},0}$ disappears completely. To the contrary, as T_∞ decreases, we encounter the so-called cut-off temperature, below which particle combustion ceases to be accomplished, because of the vanishment of the denominator in Eq. (14). The existence of the cut-off temperature further suggests that this can be of great use in evaluating kinetic parameters for the surface reaction, which will be discussed in the next section.

2.6 Cut-off Temperature

As for the cut-off temperature, obtained from the condition that the denominator in Eq. (14) vanishes, we have the following relation as

$$\left[Y_{\mathrm{O},\infty}(p_\infty/p_0)\right]_{\mathrm{cut-off}} = \left[e\, B_{\mathrm{s},0}\frac{W_\mathrm{C}q_{\mathrm{s},0}}{W_\mathrm{O}c_pT_\infty}K\frac{Ta_{\mathrm{s},0}}{T_\infty}\frac{\rho_0c_p}{2\varepsilon\sigma_\mathrm{SB}T_\infty^3}\frac{T_0}{T_\infty}\exp\left(-\frac{Ta_{\mathrm{s},0}}{T_\infty}\right)\right]^{-1}. \quad (16)$$

It is seen that the parameter $\left[Y_{\mathrm{O},\infty}(p_\infty/p_0)\right]_{\mathrm{cut-off}}$ which mainly depends on the ambient temperature T_∞, as well as the kinetic parameters for the surface reaction, can be a useful parameter in examining critical conditions, relevant to the particle combustion.

3. Results and Discussion

3.1 Critical Condition for the Particle Combustion

Figure 1 shows the Arrhenius plot of the critical conditions of $[(2r_\mathrm{s})\dot{m}]_\mathrm{cr}$ for the petroleum coke in air, with (p_∞/p_0) taken as a parameter. It is seen that the critical condition separates regions, the upper half of which corresponds to that for the particle combustion. In addition, up to a certain temperature, because of the existence of cut-off temperature, particle combustion cannot be activated, due to the radiative heat loss, which is superior to the heat released by the reaction. Once the ambient temperature exceeds the cut-off temperature, the particle activation comes to proceed, resulting in a steep drop in

$[(2r_s)\dot{m}]_{cr}$, which is followed by a gradual increase, proportional to $T_\infty^{2.75}$ at high temperatures, as mentioned. As for the cut-off temperature, it decreases as (p_∞/p_0) increases, because of the surface reaction enhanced.

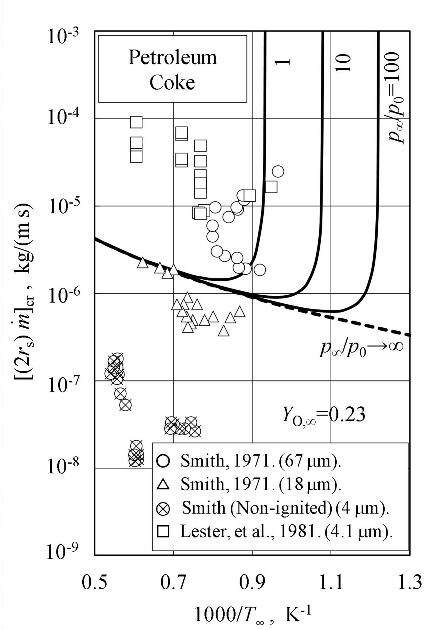

Fig. 1 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$, with respect to the critical condition for the particle combustion in air, with the pressure ratio (p_∞/p_0) taken as a parameter. Solid and dashed curves are theoretical. Data points are experimental in the literature; those for petroleum coke with p_∞/p_0=1, measured by Smith [21]; those for bituminous coal with p_∞/p_0=5~10, measured by Lester, et al. [41].

3.2 Experimental Comparisons

3.2.1 Petroleum Coke in Air Environment

Experimental data presented in Fig. 1 are those reported by Smith [21], who measured the combustion rate of petroleum coke, narrowly sized, by suspending them in a preheated airflow that passed through a reactor at an atmospheric pressure. While data for the larger particles with 67 μm diameter locate in the upper half region, separated by the critical condition, those with 4 μm diameter, designated by a symbol (⊗), locate far below the critical condition, suggesting that the reactions have failed to be activated. It is seen that the reduction in particle diameter does not necessarily facilitate particle combustion, as shown analytically in the previous sections. As for the larger particles, to the contrary, their data are fairly bounded by the critical condition, including the cut-off temperature, suggesting that particles are to be activated in the course of combustion.

As for the results with 18 μm diameter, they are intermediate, so that they locate around or slightly below the critical condition. As pointed out [29], it is confirmed again that many of these particles would not be activated in their combustion. As for the combustion of 4 μm petroleum coke, reported to be that in the diffusionally controlled regime in its original work, because of an experimental observation of its internal surface area, it is strongly required to change explanations from the viewpoint of particle combustion, whether it is activated or not.

In Fig. 1, other experimental results are also plotted, reported by Lester and his co-workers [41], who measured the combustion rates of bituminous coals, called “Illinois No. 6” with 4.1 μm diameter, at high temperatures (>1700 K) and pressures (0.5-1.0 MPa) in air, by use of the conventional shock tube. Note that the combustion rate of coal char from “Illinois No. 6” can fairly be evaluated [15] by use of the “P type” kinetic parameters, listed in Table 1, the name of which has been called with reference to the petroleum coke. Again, experimental results are in the region, bounded by the critical condition predicted.

As for the cut-off temperature, fair agreement has been demonstrated between the lower limit of the experimental data and predicted results, as shown in Fig. 1. This fair agreement further suggests that evaluation of kinetic parameters for the surface reaction, conducted in the previous work [42], has been appropriate.

3.2.2 In the Environment Oxygen Suppressed

Further comparisons are to be conducted, in order to confirm the validity and viability of the theory and/or formulation. Experimental results used are those obtained in an oxidizer-flow with $Y_{O,\infty}$ reduced to 0.11, reported by Field [20], who measured combustion rates of eleven coals and/or coal chars, from anthracite to bituminous coal in the UK, by use of the experimental method described, under an atmospheric pressure.

Figure 2 shows the similar plot of $[(2r_s)\dot{m}]_{cr}$. Experimental data examined are those for coal char from bituminous coal called "Bagworth" with 23 μm diameter, designated by a symbol (○), the combustion rate of which has already been found to be fairly evaluated [15] by use of the "P type" kinetic parameters (*cf.* Table 1). Experimental results are mainly in the region, bounded by the critical condition, suggesting that many of the particles examined are considered to be activated in the course of the combustion. In Fig. 2, results [20] for "Prince of Wales" and "Ferrymoor" with 34 μm and 38 μm diameters, respectively, are also shown, both of which have also turned out to be evaluated by use of the "P type" kinetic parameters. The same trend as that for "Bagworth" can be seen, while some results at relatively lower temperatures suggest that they could not be activated.

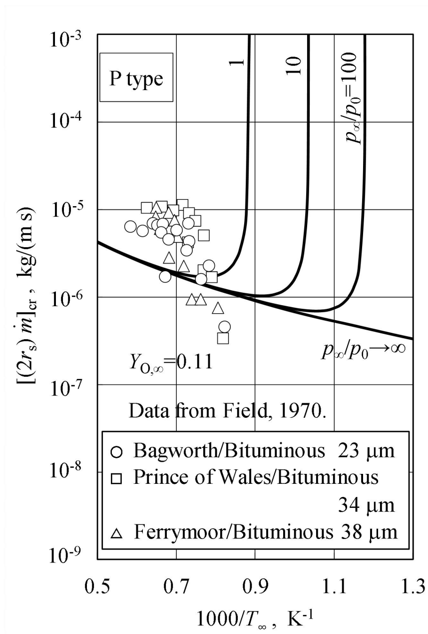

Fig. 2 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$ for the particle combustion in the oxidizer ($Y_{0,\infty}$=0.11). Data points are experimental [20] with p∞/p0=1.

Experimental results measured in much reduced oxygen environment ($Y_{0,\infty}$ =0.03) have been reported by Goetz and his co-workers [26], who measured combustion rates of representative coal chars in the United States. Figure 3 shows the similar plot of $[(2r_s)\dot{m}]_{cr}$. Experimental data examined are those for coal char from bituminous coal called "Pittsburgh No. 8" with 37-74 μm diameter, the combustion rate of which has fairly be evaluated [15] by use of the "P type" kinetic parameters (*cf.* Table 1). Because of the reduced particle diameter, as well as the suppressed oxygen concentration, it is seen that particles have failed to be activated in the course of particle combustioon.

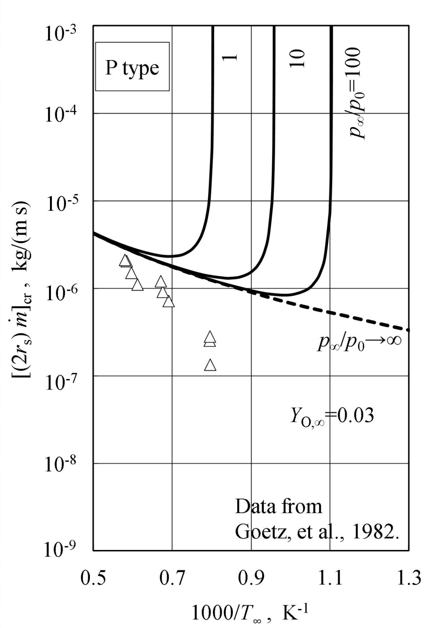

Fig. 3 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$ for the particle combustion in oxidizer ($Y_{0,\infty}$=0.03). Data points are experimental [26] with p∞/p0=1.

3.3 Cut-off Temperature

3.3.1 Results of "P type" Coal Char

Since conditions for the particle activation has been confirmed to exist by examining critical conditions, with respect to $[(2r_s)\dot{m}]_{cr}$ and T_∞, let us now demonstrate appropriateness and/or usefulness of the relation, expressed in Eq. (16), by conducting experimental comparisons. Experimental results used are those, shown in Figs. 1-3 as listed in Table 2.

Here, use has been made of an extended parameter, consisting of $Y_{0,\infty}(p_\infty/p_0)$ and the frequency factor $B_{s,O}$, since it is intended to compare predicted results with experimental data for various solid fuels, from anthracite to low-rank coal char, as well as petroleum coke. Figure 4 shows the Arrhenius plot of the extended parameter, $B_{s,0}Y_{0,\infty}(p_\infty/p_0)$. It is seen that the critical condition for the cut-off temperature separates regions, the upper half of which corresponds to that for the particle combustion, with the surface reaction activated. Experimental results in Refs. [20, 21, 41] are also shown in Fig. 4, by use of symbols black in color.

Table 2 List of symbols used in experimental comparisons, with respect to the cut-off temperature, shown in Fig. 4.

Color	Symbol	Investigator	Year	Size (μm)	Coal Type	Kinetic	$B_{s,O}$
Black	○	Field	1970	34	Prince of Wales/	P type	4.1×10^6
	◇	Smith	1971	67	Petroleum Coke	P type	
	△	Lester, et al.	1981	4.1	Illinois No. 6/	P type	
Blue	○	Field	1969	82, 105	Coal Char	A type	2.2×10^6
	◇	Smith	1971	78	Semi-Anthracite	A type	
Red	○	Field	1970	27	Whitwick/	L type	2.2×10^7
	◇	Hamor, et al.	1973	89	Brown Coal Char	L type	

As far as the trend and the approximate magnitude are concerned, a fair degree of agreement has been demonstrated between the lower limit of the experimental results and the predicted curve for the cut-off temperature. Therefore, further comparisons by use of other experimental data in reliable sources are strongly required, in order to elucidate characteristics, relevant to the cut-off temperature in the particle combustion.

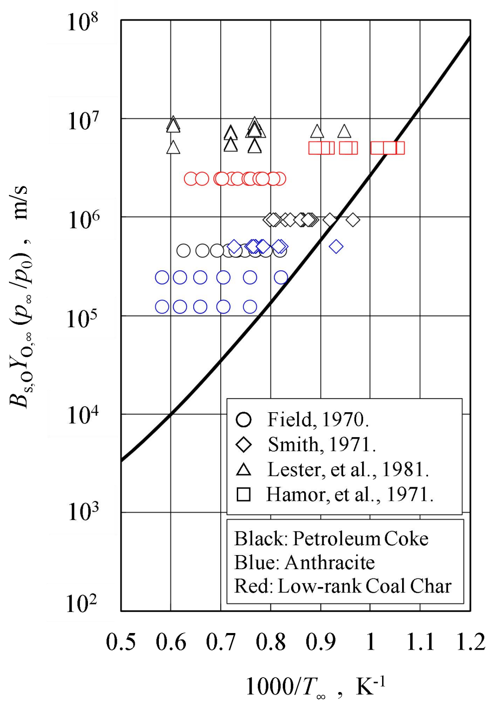

Fig. 4 Arrhenius plot of the parameter $B_{s,0}Y_{0,\infty}(p_\infty/p_0)$, relevant to the cut-off temperature. Data points are experimental; [20, 21, 41] for the petroleum coke; [19, 22] for the anthracite; [20, 23] for the low-rank coal char. While p_∞/p_0=5~10 for [41], p_∞/p_0=1 for other experimental results. A sold curve is predicted by the theory.

3.3.2 Results of Other Coal Chars

Experimental data from other sources, as listed in Table 2, have also been used for this comparison, after having examined the critical conditions (cf. Appendix A). Since fair evaluation of the combustion rates by use of the “A type” kinetic parameters has been confirmed [15], those results [19, 22] are also plotted in Fig. 4, by use of symbols blue in color. As for results [20, 23], since fair evaluation [15] by use of the “L type” kinetic parameters has been shown, they are also plotted in Fig. 4, by use of symbols red in color, after having examined the critical conditions (cf. Appendix B). It is seen that these results further confirm the lower limit with respect to the cut-off temperature, presenting fair degree of agreement between those, examined in the experiments and predicted in the theory.

3.4 Discussion

Thus far, comparisons relevant to the critical condition have been conducted by use of the parameter, $[(2r_\mathrm{s})\dot{m}]_\mathrm{cr}$, derived by combining two kinds of comprehensive parameters, reported in the previous works. It has been shown that the region for the particle combustion activated is bounded by the critical condition and that it expands as the oxygen mass-fraction $Y_{0,\infty}$ and the pressure ratio (p_∞/p_0) increase. In addition, it is seen that there exists the lower limit of the ambient temperature, called the cut-off temperature.

In comparisons, use has been made of various experimental results of coals and/or coal chars, from anthracite to low-rank coal char, as well as petroleum coke. Note that appropriateness of the surface kinetic parameters for those solid fuels, indispensable in evaluating combustion rate, has already been confirmed in a series of studies. Through experimental comparisons, the validity of the theory and/or formulation for the particle combustion in the quiescent environment, especially for the critical condition relevant to the particle activation, has been demonstrated, suggesting that they are applicable to practical combustion in manipulating particle combustion, as well as academic works to be done not only from the qualitative but also from the quantitative point of view.

As for the cut-off temperature, albeit it has been predicted by theoretical/numerical works, its existence has further been shown by use of experimental results reported in the literature, through conducting the present study. Although experimental results for individual coal char with different reactivity merely indicate that they locate in the upper half region, it is seemed that once gathered together, such various experimental results with the lowest values can indicate the cut-off temperature, relevant to the ambient temperature, oxygen concentration, pressure, and surface reactivity. In addition, as far as the trend and approximate magnitude are concerned, a fair degree of agreement is demonstrated between the lower limit of the experimental results and the predicted curve for the critical condition, including the cut-off temperature. Therefore, further studies by use of experimental data in other reliable sources are strongly required, in order to elucidate the critical conditions, relevant to the particle combustion, with putting an emphasis on the problem whether it is to be activated or not, in the course of combustion.

4. Concluding Remarks

A useful parameter consisting of the particle diameter and the combustion rate has newly been obtained, by combining two kinds of comprehensive parameters, derived from the asymptotics, with focusing on the temporal variation of the particle temperature in the plateau stage, in order to elucidate the critical condition for the particle combustion that can be activated in the course of combustion. It has been shown that the critical condition separates regions, upper half of which corresponds to that for particle combustion, with the surface reaction activated, that experimental results for burning certainly locate in the upper half region, and that excessively smaller particles are unfavorable to the particle combustion. As for the ambient temperature, it is closely related to the cut-off temperature, only above which particle combustion can be accomplished. In addition, fair agreement between experimental and predicted results suggests that the formulation, including that for the critical condition, has been constructed with capturing essential feature. Since this kind of feature has not been examined in the previous studies, the present results are expected to contribute much not only to academic works but also to practical applications in manipulating particle combustion.

Acknowledgment

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Appendix A. Critical Condition for the "A type" Coal Char

A.1 In Air Environment

Results of semi-anthracite in air measured by Smith [22] are plotted in Fig. A1. Similarly, data for the larger particle (78 μm) locate in the upper half region, bounded by the critical condition, in general, while those for the smaller particles (6 μm) are below the critical condition. The same comments done in Figs. 1-3 are also to be made. Note here that the combustion rate of semi-anthracite, as reported [14], can fairly be evaluated, by use of the "A type" kinetic parameters, the frequency factor of which is about a half of that for the "P type" kinetic parameters, as listed in Table 1.

In addition, it may be informative to note that the combustion of 6 μm semi-anthracite, in contrast with that of the 4 μm petroleum coke discussed, has been categorized to that in the kinetically controlled regime in its original work [22], because of the much reduced combustion rate, compared to those for the larger particles, due to a change of regimes from pore-diffusionally controlled to kinetically controlled. Nonetheless, since very much suppressed combustion rate has been reported, even much smaller, compared to that in the kinetically controlled regime, another explanation is anxiously required even for this case, to be conducted from the view-point of particle activation.

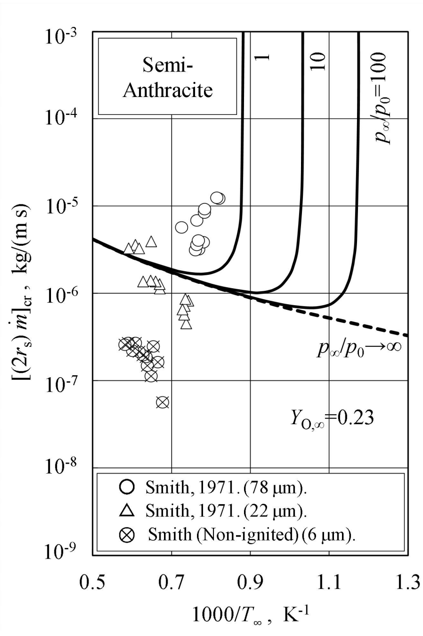

Fig. A1 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$ for the particle combustion in air, with (p_∞/p_0) taken as a parameter. Data points are experimental [22] for the semi-anthracite.

A.2 In the Environment Oxygen Suppressed

As for the particle combustion in the environment oxygen suppressed, it has also been reported by Field [19], so that let us now examine those results from the viewpoint whether particles are to be burned out or not. Figure A2 shows the similar plot of $[(2r_s)\dot{m}]_{cr}$ for coal chars, parent coal of which are bituminous [5]. It has even been reported [14, 15] that these combustion rates can fairly be evaluated by use of the "A type" kinetic parameters. It is seen that results for $2r_s$=105 μm and 82 μm in diameters are in the region, bounded by the critical condition, suggesting that the particle activation would be anticipated during combustion. As for the smaller particles with ca. 30 μm diameter, we see that some are in the region bounded by the critical condition when T_∞ is higher than about 1400 K, while others are around or below the critical condition, suggesting that those would fail to burn, in the course of combustion. Again, it is suggested that excessively smaller particles are unfavorable to particle activation to burn.

Although it is not shown here, experimental results for particles, from bituminous coals called “Brodswarth” and “Britannia” with ca. 30 μm diameter, measured by Field [20], present the same trend. In addition, in a much reduced oxygen environment ($Y_{0,\infty}$=0.06), results measured by Field [19] present that the particle activation becomes much harder, which has also been confirmed through conducting experimental comparisons by use of the similar plot.

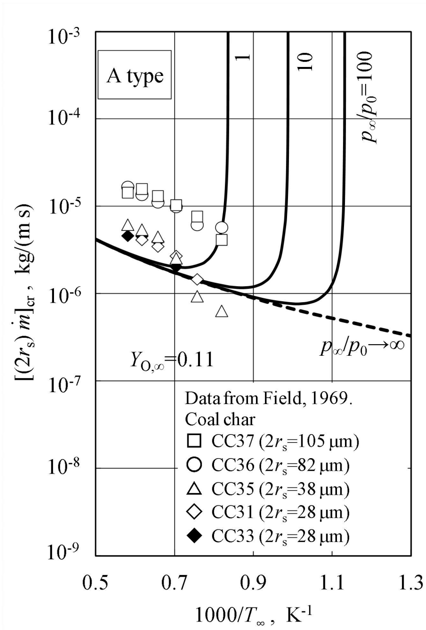

Fig. A2 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$ for the particle combustion in the oxidizer ($Y_{0,\infty}$=0.11), with (p_∞/p_0) taken as a parameter. Data points are experimental for the coal char (bituminous) [19].

Appendix B. Critical Condition for the “L type” Coal Char

B.1 In Air Environment

Results of brown coal char in air measured by Hamor, et al. [23] are plotted in Fig. B1.

Similarly, data for the larger particles (89 μm) locate in the upper half region, bounded by the critical condition, in general, while those for the smaller particles (22 μm) are below or around the critical condition. For the intermediate particles, some are to be activated when T_∞ is high, while others are not, in the course of combustion. The same comments done in Figs. 1-3 are also to be made. Note here that the combustion rate of the brown coal char, as reported [14], can fairly be evaluated, by use of the "L type" kinetic parameters (*cf.* Table 1), which is named with reference to the low-rank coal char.

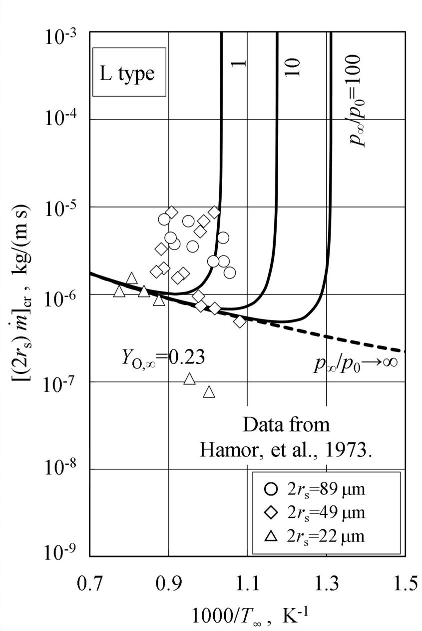

Fig. B1 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$, for the particle combustion in air, with (p_∞/p_0) taken as a parameter. Data points are experimental [23] for the brown coal char.

B.2 In the Environment Oxygen Suppressed

Figure B2 shows the similar plot with respect to $[(2r_s)\dot{m}]_{cr}$ and T_∞ for coal chars [20], from bituminous coal called "Whitwick" with 27 μm diameter. In the oxidizing-flow, $Y_{O,\infty}$ is reduced to 0.11. Note that this combustion rate has also been reported [14] to fairly be evaluated by use of the "L type" kinetic parameters. It is seen that the results are in the upper half region, bounded by the critical condition.

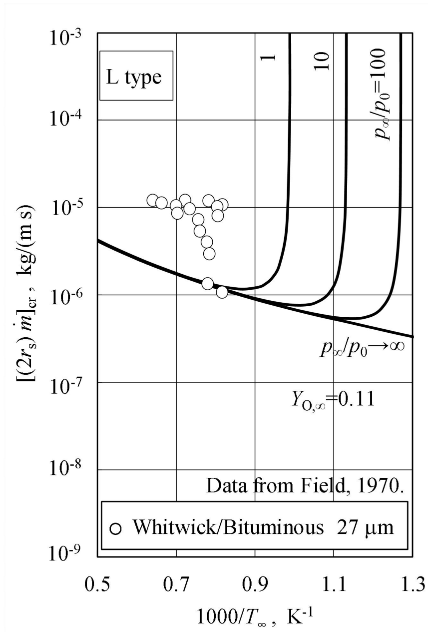

Fig. B2 Arrhenius plot of $[(2r_s)\dot{m}]_{cr}$ for the particle combustion in the oxidizer ($Y_{O,\infty}$=0.11), with (p_∞/p_0) taken as a parameter. Solid curves are theoretical. Data points are experimental [20] for the bituminous coal char.

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