

Linear Stability and Pressure-Driven Response Function of Solid Propellants with Phase Transition

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An extension of the Zel'dovich–Novozhilov approach to adiabatic burning of solid energetic materials subjected to a concentrated phase transition is presented. The pressure-driven frequency response function and intrinsic stability boundary are obtained in the linear approximation of the problem. The intrinsic stability boundary is portrayed as a parametric representation of oscillatory burning frequency. The corresponding previous results are recovered as a special case for no phase transition. The surface-temperature sensitivity parameter r is deduced by assuming the Arrhenius surface pyrolysis law. It is shown that phase transition may strongly affect the frequency response function, notwithstanding its limited thermal effect, if the operating point moves closer to the stability boundary. Some typical results are discussed. To validate these theoretical expectations, accurate error estimates of experimental results are needed.

Nomenclature

A	= nondimensional function used in linear frequency response analysis, Eq. (19)	Q	= heat release, cal/g (positive if exothermic)
A_1, A_2, A_3	= coefficients used in stability matrix; Sec. III.C	R_p	= mass burning rate response to pressure fluctuations, defined in Eq. (85)
\tilde{A}_s	= multiplicative factor of Arrhenius pyrolysis, (cm/s)/(atm) ^{n_s} , Eq. (21)	\Re	= universal gas constant, 1.987 cal/mol · K
a	= flame modeling (FM) nondimensional stability parameter, Eq. (8)	r	= ZN initial-temperature steady sensitivity parameter, nondimensional, defined in Eq. (4)
B	= nondimensional function used in linear frequency response analysis, Eq. (15)	r_b	= burning rate, cm/s
b	= FM nondimensional stability parameter, Eq. (9)	T	= temperature, K
C_1, C_2	= constants used in the unsteady condensed phase thermal profiles; Sec. III.B	T_0	= initial propellant temperature, K
C_3, C_4		t	= time coordinate, s
c	= specific heat, cal/g · K	x	= space coordinate, cm
\tilde{E}	= activation energy, cal/mol	Z, Z_k, Z_r	= coefficients used in Sec. III.C
f	= thermal gradient at condensed-phase side of burning surface, K/cm	$z_{1,2}$	= $[1 \pm \sqrt{(1 + 4i\Omega)}]/2$, complex characteristic roots of fluctuating thermal profile; Eq. (57)
i	= imaginary unit	α	= thermal diffusivity, cm ² /s; condensed-phase parameter defined in Eq. (40)
k	= Zel'dovich–Novozhilov (ZN) initial-temperature steady sensitivity parameter, nondimensional, defined in Eq. (3)	β	= condensed-phase parameter defined in Eq. (41)
$k_{(\dots)}$	= thermal conductivity, cal/cm · s · K	γ	= condensed-phase parameter defined in Eq. (42)
ℓ_{tra}	= transition position, cm	Δ	= condensed-phase parameter defined in Eq. (43)
m	= mass burning rate, g/cm ² · s	δ	= ZN Jacobian, nondimensional, defined in Eq. (5)
n	= pressure exponent of ballistic steady burning rate, nondimensional, Eq. (17)	μ	= ZN pressure sensitivity of steady surface temperature, nondimensional, defined in Eq. (2)
n_s	= pressure exponent of pyrolysis law, nondimensional, Eq. (21) (Arrhenius)	ν	= ZN pressure sensitivity of steady burning rate, nondimensional, defined in Eq. (1)
n_{T_s}	= pressure sensitivity of steady surface temperature, nondimensional, Eq. (20)	ρ	= density, g/cm ³
p	= pressure, atm	σ_p	= $[\partial \ln \tilde{m} / \partial T_0]_{p=\text{const}}$, temperature sensitivity of steady burning rate, 1/K
p_{ref}	= reference pressure (68 atm)	σ_{T_s}	= $[\partial \ln \tilde{T}_s / \partial T_0]_{p=\text{const}}$, temperature sensitivity of steady surface temperature, 1/K
		Ω	= nondimensional circular frequency, $\omega \cdot \alpha_c / \tilde{r}_b^2$
		ω	= circular frequency, rad/s

Subscripts

bif	= bifurcation
c	= condensed phase
i	= initial
im	= imaginary part
p	= pressure
real	= real part
ref	= reference
s	= burning surface
tra	= transition

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Superscripts

$\overline{(\dots)}$	= steady-state value
(\dots)	= dimensional value
$(\dots)'$	= fluctuating value
$(\dots)^*$	= complex conjugate value

I. Introduction

THE current intrinsic stability analyses of solid energetic materials assume chemical reactions concentrated at the burning surface (assumed infinitely thin), and no other chemical activity whatsoever in the condensed phase. However, comparison with experimental results shows discrepancies from the theoretical expectations (e.g., see Ref. 1). Although other factors also may play a role, as a first step to bridge this possible gap an extension of the theory is presented to include concentrated phase transitions somewhere in depth in the condensed phase. Moreover, in several instances, phase transitions—solid to solid or solid to liquid—are known to occur in practical applications. This may affect components of energetic materials, for example, ammonium nitrate (AN)^{2,3}; ammonium perchlorate (AP)^{4,5}; cyclotetramethylene-tetranitramine (Ref. 6, p. 142, and Ref. 7, p. 203), cyclotrimethylene-trinitramine (RDX; Ref. 6, p. 142, Ref. 7, p. 203, and Ref. 8); additives to energetic materials, e.g., KCl and LiF (Ref. 9); or just the overall mixture making the energetic material, e.g., AN-based energetic compositions,¹⁰ RDX-based composite propellants,¹¹ the foam zone in double-base propellants,¹² and so on. The modification of condensed-phase steady thermal profiles due to AP phase transition is clearly shown in Figs. 7–9 of Ref. 13. The extension is conducted within the Zel'dovich–Novozhilov (ZN) framework, but connections with the corresponding flame modeling (FM) framework also are discussed. Both approaches, ZN and FM, share the basic assumptions of quasi-steady gas-phase, homogeneous condensed-phase, and one-dimensional (QSHOD) burning-strand framework. The intrinsic stability boundary of QSHOD burning was found first by Zel'dovich^{14,15} in his pioneering work started in 1942, but assuming variable burning rate with a constant surface temperature. This assumption was relaxed in the successive investigations. Novozhilov (e.g.; Refs. 16–19) in 1965 first obtained the QSHOD ZN stability boundary and frequency response function including both variable burning rate and surface temperature.

The corresponding QSHOD FM result for a premixed flame had been obtained previously by Denison and Baum²⁰ in 1961. A successive investigation conducted by Krier et al.²¹ (KTSS) at Princeton University in 1968 extended the FM approach to diffusive flames. Systematic investigations were carried out by Culick for a variety of flame configurations, e.g., Refs. 22–24. All of these investigations were conducted for the linear approximation of pressure-driven burning; in general, except the chemical reactions concentrated at the burning surface, a chemically inert condensed phase was otherwise assumed.

Few papers allowed for chemical reactions distributed in the condensed phase: Results were presented in Refs. 22, 25–30 for pressure-driven burning examined by a variety of techniques. A further generalization of the FM approach was carried out in 1995 1) for the full nonlinear problem with arbitrary flames but limited to chemically inert condensed phase, except the chemical reactions concentrated at the burning surface, and 2) for the linear approximation of the problem but considering also distributed chemical reactions.³¹

The objective of this paper is to determine the intrinsic stability boundary and frequency response function, including in-depth phase transition effects, for the linear approximation of pressure-driven burning, within the ZN framework. Adiabatic burning is assumed.

The problem is formulated in Sec. III. The solution obtained with phase transition is discussed in Sec. III.C (intrinsic stability) and Sec. III.D (frequency response function); the standard solution with no phase transition, recalled in Sec. II, is recovered as a particular case. Typical results are illustrated for the test case shown in Table 1. Conclusions and future work are discussed in Sec. V.

Table 1 Properties of test-case baseline and modifications

Assumed or measured properties	
Transition temperature, T_{tra}	$= 513 \text{ K}$
Transition heat, Q_{tra}	$= -18.3 \text{ cal/g}$
Initial temperature of sample, T_0	$= 298 \text{ K}$
Condensed-phase specific heat, c_c	$= 0.33 \text{ cal/g K}$
Burning-rate initial-temperature sensitivity, σ_p	$= 0.003 \text{ K}^{-1}$
Condensed-phase thermal diffusivity, α_c	$= 0.0018 \text{ cm}^2/\text{s}$
Steady burning rate, \bar{r}_b	$= 1.132 \cdot (\bar{p}/p_{\text{ref}})^{0.526} \text{ cm/s}$
Steady burning surface temperature, \bar{T}_s	$= 947 \cdot (\bar{p}/p_{\text{ref}})^{0.045} \text{ K}$
Surface Arrhenius activation energy, E_s	$= 16,000 \text{ cal/mol}$
Pyrolysis law pressure power	
(Arrhenius simple), n_s	$= 0$
(Arrhenius generalized), n_s	$= 0.526 - 0.045 \cdot (16,000/1.987 \cdot 947) \cdot (\bar{p}/p_{\text{ref}})^{-0.045}$
Condensed-phase parameter $\gamma \equiv \beta/\alpha$	$= -[c_c(T_{\text{tra}} - T_0)/Q_{\text{tra}}] = 3.877$
Operating pressure, p	$= 10 \text{ atm}$
Modified test-case baseline	
Operating pressure, p	$= 1 \text{ or } 68 \text{ atm}$
Burning-rate initial-temperature sensitivity, σ_p	$= 0.002 \text{ or } 0.004 \text{ K}^{-1}$
Transition temperature, T_{tra}	$= 400 \text{ or } 600 \text{ K}$
Transition heat, Q_{tra}	$= -80 \text{ or } +40 \text{ cal/g}$

II. Standard Solution

In the standard ZN formulation proposed by Novozhilov^{16–19} in 1965, four nondimensional steady-state parameters are introduced to describe the dependence of ballistic properties on pressure and ambient temperature

$$\nu \equiv \left[\frac{\partial \ln \bar{m}}{\partial \ln \bar{p}} \right]_{T_0 = \text{const}} \quad (1)$$

$$\mu \equiv \frac{1}{(\bar{T}_s - T_0)} \left[\frac{\partial \bar{T}_s}{\partial \ln \bar{p}} \right]_{T_0 = \text{const}} \quad (2)$$

$$k \equiv (\bar{T}_s - T_0) \left[\frac{\partial \ln \bar{m}}{\partial T_0} \right]_{\bar{p} = \text{const}} \quad (3)$$

$$r \equiv \left[\frac{\partial \bar{T}_s}{\partial T_0} \right]_{\bar{p} = \text{const}} \quad (4)$$

where $\bar{m}_s = \rho_c \bar{r}_b = \bar{m}$ is the steady surface mass burning rate. A possible correlation among the four sensitivity parameters is revealed by the pressure Jacobian defined as

$$\delta \equiv \frac{\partial(\ln \bar{m}, \bar{T}_s)}{\partial(\ln \bar{p}, T_0)} = \nu r - \mu k \quad (5)$$

Should $\delta = 0$, then one of the four sensitivity parameters can be evaluated from the remaining three. A finite value of the Jacobian, although suspected for different compositions, cannot be shown experimentally in a convincing way (e.g., see Ref. 18, pp. 21–22, Ref. 19, pp. 617–618) because of inherent difficulties in measuring surface temperatures even under steady conditions.³² At any rate, for ZN, one finds in general that

$$\frac{\delta}{r} \equiv \frac{\partial(\ln \bar{m}, \bar{T}_s)/\partial(\ln \bar{p}, T_0)}{[\partial \bar{T}_s/\partial T_0]_{\bar{p} = \text{const}}} = \left[\frac{\partial \ln \bar{m}}{\partial \ln \bar{p}} \right]_{\bar{T}_s = \text{const}} = \nu - \mu \frac{k}{r} \quad (6)$$

yielding different expressions for different pyrolysis laws and burning regimes.

For the reader's convenience, the following well-established results, when chemical reactions are concentrated at the burning surface and no phase transition occurs, are recalled:

1) The (intrinsic) stability condition in the ZN formulation^{16,18,19} can be written as

$$\begin{cases} k < 1 & \text{always stable} \\ k > 1 & \text{stable if } r > \frac{(k-1)^2}{k+1} \end{cases}$$

Table 2 Data matrix of baseline and modifications

p , atm	T_s , K	Q_{tra} , cal/g	T_{tra} , K	σ_p , K ⁻¹	$\Delta \equiv \bar{T}_s - T_0$ $-(Q_{\text{tra}}/c_c)$, K	$\alpha \equiv$ $-(Q_{\text{tra}}/c_c)/\Delta$	$\beta \equiv$ $(T_{\text{tra}} - T_0)/\Delta$	$\gamma \equiv -[c_c(T_{\text{tra}}$ $- T_0)/Q_{\text{tra}}]$	r	k	$A =$ k/r	$B =$ $1/k$	$n_s =$ δ/r
1	783.23	-18.3	513	0.003	540.68	0.103	0.398	3.877	0.229	1.456	6.358	0.687	0.063
10	868.73	-18.3	513	0.003	626.18	0.089	0.343	3.877	0.281	1.712	6.093	0.584	0.109
68	947	-18.3	513	0.003	704.45	0.079	0.305	3.877	0.334	1.947	5.829	0.514	0.143
1	783.23	-18.3	513	0.002	540.68	0.103	0.398	3.877	0.152	0.971	6.388	1.030	0.063
10	868.73	-18.3	513	0.002	626.18	0.089	0.343	3.877	0.188	1.142	6.075	0.876	0.109
68	947	-18.3	513	0.002	704.45	0.079	0.305	3.877	0.223	1.298	5.821	0.770	0.143
1	783.23	-18.3	513	0.004	540.68	0.103	0.398	3.877	0.305	1.941	6.364	0.515	0.063
10	868.73	-18.3	513	0.004	626.18	0.089	0.343	3.877	0.375	2.283	6.088	0.438	0.109
68	947	-18.3	513	0.004	704.45	0.079	0.305	3.877	0.446	2.596	5.821	0.385	0.143
1	783.23	-18.3	400	0.003	540.68	0.103	0.189	1.839	0.229	1.456	6.358	0.687	0.063
10	868.73	-18.3	400	0.003	626.18	0.089	0.163	1.839	0.281	1.712	6.093	0.584	0.109
68	947	-18.3	400	0.003	704.45	0.079	0.145	1.839	0.334	1.947	5.829	0.514	0.143
1	783.23	-18.3	600	0.003	540.68	0.103	0.559	5.446	0.229	1.456	6.358	0.687	0.063
10	868.73	-18.3	600	0.003	626.18	0.089	0.482	5.446	0.281	1.712	6.093	0.584	0.109
68	947	-18.3	600	0.003	704.45	0.079	0.429	5.446	0.334	1.947	5.829	0.514	0.143
1	783.23	-80	513	0.003	727.65	0.333	0.296	0.887	0.229	1.456	6.358	0.687	0.063
10	868.73	-80	513	0.003	813.15	0.298	0.264	0.887	0.281	1.712	6.093	0.584	0.109
68	947	-80	513	0.003	891.42	0.272	0.241	0.887	0.334	1.947	5.829	0.514	0.143
1	783.23	+40	513	0.003	364.02	-0.333	0.591	-1.774	0.152	0.971	6.388	1.030	0.063
10	868.73	+40	513	0.003	449.52	-0.270	0.478	-1.774	0.188	1.142	6.075	0.876	0.109
68	947	+40	513	0.003	527.79	-0.230	0.407	-1.774	0.223	1.298	5.821	0.770	0.143

and the equivalent FM formulation³³ of the parabolic boundary is

$$a = \frac{b(b-1)}{2} \quad (7)$$

being³¹

$$a = k/r \quad (8)$$

$$b = 1 + k/r - 1/r \quad (9)$$

2) The natural oscillatory frequency just at the stability boundary in the ZN formulation^{16,18,19} can be written as

$$\Omega_{\text{bif}} = \frac{\sqrt{k}}{r} = \sqrt{k} \frac{(k+1)}{(k-1)^2} \quad (10)$$

and the equivalent FM formulation³³ is

$$\Omega_{\text{bif}} = [(b-1)/2]\sqrt{b(b-2)} \quad (11)$$

requiring $b > 2$ for real Ω .

3) The adiabatic pressure-driven frequency response function in the ZN formulation¹⁷⁻¹⁹ can be written as

$$R_p(\Omega) = \frac{\nu + \delta(z_1 - 1)}{r(z_1 - 1) + k[(1/z_1) - 1] + 1} \quad (12)$$

and the equivalent Arrhenius FM formulation²³ is

$$R_p(\Omega) = \frac{n + (n_s/AB)(z_1 - 1)}{(1/AB)(z_1 - 1) + (1/B)[(1/z_1) - 1] + 1} \quad (13)$$

where³⁴

$$A \equiv k/r, \quad B \equiv 1/k, \quad n \equiv \nu, \quad n_s \equiv \delta/r \quad (14)$$

In both versions, the static limit $R_p(\Omega \rightarrow 0) = n$ (FM) or ν (ZN) is defined by the experimental steady-burning-ratelaw.

4) The ZN parameters can be converted into FM parameters by putting

$$k = \sigma_p(\bar{T}_s - T_0) = \frac{1}{B} = \frac{a}{1+a-b} \quad (15)$$

$$r = \sigma_{T_s} \bar{T}_s = \frac{1}{AB} = \frac{1}{1+a-b} \quad (16)$$

$$\nu = n \quad (17)$$

$$\mu = n_{T_s} \frac{\bar{T}_s}{\bar{T}_s - T_0} \quad (18)$$

Thus³²

$$\frac{k}{r} = \frac{\sigma_p}{\sigma_{T_s}} \frac{\bar{T}_s - T_0}{\bar{T}_s} = A = a \quad (19)$$

and the ballistic Jacobian can be written as

$$\delta/r = n - n_{T_s} \cdot \sigma_p/\sigma_{T_s} \quad (20)$$

If one assumes a general form of the Arrhenius pyrolysis law (e.g., see Refs. 22-24, 35-38)

$$r_b = \tilde{A}_s p^{n_s} \exp(-\tilde{E}_s/\Re T_s) \quad (21)$$

then one can model the surface temperature sensitivity parameter r as

$$r = \sigma_p(\Re \bar{T}_s^2 / \tilde{E}_s) \quad (22)$$

and $\delta/r = n_s$ (for discrete pyrolysis functions, see Ref. 32; see also the Appendix, of Ref. 39). Note that if one assumes a simple ($n_s = 0$) Arrhenius pyrolysis law

$$r_b = \tilde{A}_s \exp(-\tilde{E}_s/\Re T_s) \quad (23)$$

then the sensitivity parameter r is not modified [see Eq. (22)] but necessarily $\delta/r = n_s = 0$.

Under all circumstances, the accurate knowledge of σ_p is important but still a challenge; data collections can be found in Refs. 40-42.

III. Formulation of the Phase Transition Problem

The physical problem is sketched in Fig. 1. A one-dimensional strand of homogeneous material is assumed to be burning with a quasi-steady gas phase subjected to pressure changes in time only. Thermophysical properties are assumed to be, at most, pressure dependent. The strand is burning in a vessel at uniform pressure and is subjected to no radiation, no velocity coupling, and no external forces. Assume no condensed-phase chemical activity, except the chemical reactions concentrated at the burning surface and at the phase transition plane located somewhere in depth. Define a

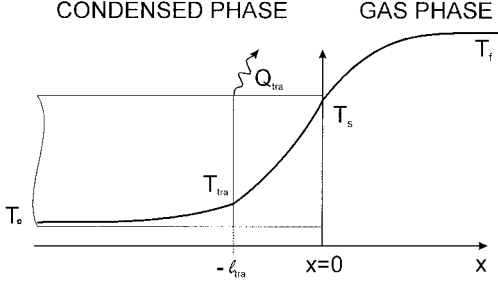


Fig. 1 Schematic of the physical problem.

Cartesian axis with its origin anchored at the burning surface and positive in the gas-phase direction. The phase transition, be it solid to solid or solid to liquid, is assumed to be concentrated at the position $x = -\ell_{\text{tra}}(t)$, where the transition temperature T_{tra} is observed. In general, the phase transition will take place with a (net) heat release Q_{tra} (positive if exothermic). Both parameters T_{tra} and Q_{tra} are considered to be known and fixed quantities. For the sake of simplicity, thermophysical properties are assumed to be unaffected by the phase transition.

Let the condensed phase ($x < 0$) be a semi-infinite slab of uniform and isotropic composition, and T_0 its initial temperature. Only the particular case of pressure-driven burning is considered. Under these circumstances, the energy conservation equation is simply

$$\frac{\partial T}{\partial t} + r_b \frac{\partial T}{\partial x} = \alpha_c \frac{\partial^2 T}{\partial x^2} \quad (24)$$

The boundary conditions can be written over the thickness $-\infty < x < -\ell_{\text{tra}}$ as

$$T(x \rightarrow -\infty, t) = T_0 \quad (25)$$

$$T(x \rightarrow -\ell_{\text{tra}}, t) = T_{\text{tra}} \quad (26)$$

and over the thickness $-\ell_{\text{tra}} < x < 0$ as

$$T(x \rightarrow -\ell_{\text{tra}}, t) = T_{\text{tra}} \quad (27)$$

$$T(x = 0, t) = T_s \quad (28)$$

At the position $x = -\ell_{\text{tra}}(t)$, energy conservation requires matching of the two thermal profiles

$$k_c \left[\frac{\partial T}{\partial x} \right]_{-\ell_{\text{tra}}} = k_c \left[\frac{\partial T}{\partial x} \right]_{-\ell_{\text{tra}}} + \rho_c \left(r_b + \frac{d\ell_{\text{tra}}}{dt} \right) Q_{\text{tra}} \quad (29)$$

If pressure and initial temperature are taken as independent variables, then, under steady state,

$$\bar{r}_b = \bar{r}_b(\bar{p}, T_0) \quad (30)$$

$$\bar{T}_s = \bar{T}_s(\bar{p}, T_0) \quad (31)$$

Likewise, under steady state, the thermal gradient at the condensed-phase side of the burning surface is

$$\bar{f} \equiv \left[\frac{dT}{dx} \right]_{x=0^-} = \frac{\bar{r}_b}{\alpha_c} \left(\bar{T}_s - T_0 - \frac{Q_{\text{tra}}}{c_c} \right) \quad (32)$$

taking explicitly into account the heat release associated with phase transition. Under both steady and nonsteady operations, introducing pressure and thermal gradient as independent variables, one can write

$$r_b = r_b(p, f) \quad (33)$$

$$T_s = T_s(p, f) \quad (34)$$

A. Steady-State Solution

For the steady-state part of the problem, one finds for the thermal profile

$$1) -\infty < x < -\ell_{\text{tra}}$$

$$\bar{T}(x) = T_0 + (T_{\text{tra}} - T_0) \exp[\bar{r}_b/\alpha_c(x + \ell_{\text{tra}})] \quad (35)$$

$$2) -\ell_{\text{tra}} < x < 0$$

$$\bar{T}(x) = \frac{T_{\text{tra}} - \bar{T}_s \exp(-\bar{r}_b/\alpha_c \ell_{\text{tra}}) + (\bar{T}_s - T_{\text{tra}}) \exp(\bar{r}_b/\alpha_c x)}{1 - \exp(-\bar{r}_b/\alpha_c \ell_{\text{tra}})} \quad (36)$$

Matching at the transition location yields

$$3) x = -\ell_{\text{tra}}$$

$$k_c \left[\frac{d\bar{T}}{dx} \right]_{-\ell_{\text{tra}}} = k_c \left[\frac{d\bar{T}}{dx} \right]_{-\ell_{\text{tra}}} + \rho_c \bar{r}_b Q_{\text{tra}} \quad (37)$$

which, by replacing the proper thermal gradients, in turn gives

$$\frac{\bar{r}_b}{\alpha_c} (T_{\text{tra}} - T_0) = \frac{\bar{r}_b}{\alpha_c} \frac{\bar{T}_s - T_{\text{tra}}}{1 - \exp(-\bar{r}_b/\alpha_c \ell_{\text{tra}})} \exp(-\bar{r}_b/\alpha_c \ell_{\text{tra}}) + \frac{\bar{r}_b}{\alpha_c} \frac{Q_{\text{tra}}}{c_c} \quad (38)$$

Thus, the transition location inside the condensed phase is defined by

$$\ell_{\text{tra}} = \frac{\alpha_c}{\bar{r}_b} \ln \frac{\bar{T}_s - T_0 - Q_{\text{tra}}/c_c}{T_{\text{tra}} - T_0 - Q_{\text{tra}}/c_c} = \frac{\alpha_c}{\bar{r}_b} \ln \left(\frac{1}{\alpha + \beta} \right) \quad (39)$$

where (cf. the corresponding definitions in Ref. 29)

$$\alpha \equiv -\frac{Q_{\text{tra}}/c_c}{\bar{T}_s - T_0 - Q_{\text{tra}}/c_c} = -\frac{Q_{\text{tra}}/c_c}{\Delta} \quad (40)$$

$$\beta \equiv \frac{T_{\text{tra}} - T_0}{\bar{T}_s - T_0 - Q_{\text{tra}}/c_c} = \frac{T_{\text{tra}} - T_0}{\Delta} \quad (41)$$

$$\gamma \equiv \frac{\beta}{\alpha} = -\frac{c_c(T_{\text{tra}} - T_0)}{Q_{\text{tra}}} \quad (42)$$

$$\Delta \equiv \bar{T}_s - T_0 - \frac{Q_{\text{tra}}}{c_c} \quad (43)$$

For the selected test case (see Table 1), the pressure dependence of the relevant thermophysical parameters is illustrated in Fig. 2. Parameter $\Delta \equiv \bar{T}_s - T_0 - Q_{\text{tra}}/c_c$ shows a fair increase with pressure due to increasing surface temperature; both parameters $\alpha \equiv -(Q_{\text{tra}}/c_c)/\Delta$ and $\beta \equiv (T_{\text{tra}} - T_0)/\Delta$ show a slight decrease for increasing pressure, again due to increasing surface temperature. For the assumptions made, pressure effects cannot exist for $\gamma \equiv \beta/\alpha = -c_c(T_{\text{tra}} - T_0)/Q_{\text{tra}}$. Typical trends of the nondimensional transition depth $\ell_{\text{tra}} \bar{r}_b/\alpha_c$ are illustrated in Fig. 3: $\ell_{\text{tra}} \bar{r}_b/\alpha_c$

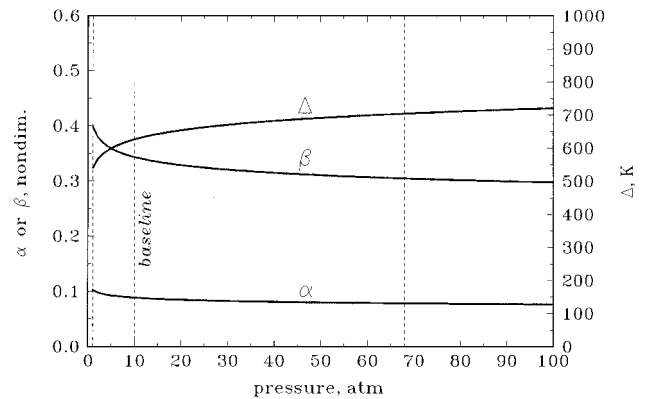


Fig. 2 Dependence of thermophysical parameters α , β , and Δ on pressure in the test case for the indicated set of operating parameters.

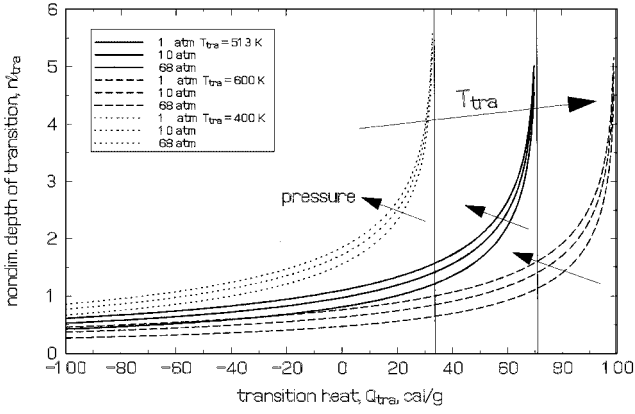


Fig. 3 Dependence of nondimensional transition depth on operating conditions in the test case for the indicated set of operating parameters.

is seen to increase with increasing Q_{tra} , increasing pressure, and decreasing T_{tra} . Notice that a limiting value of $Q_{tra} \leq c_c(T_{tra} - T_0)$ exists for which the denominator of the logarithmic factor in Eq. (39) vanishes. This bound concerns only exothermic processes and is more severe for smaller T_{tra} , as manifest in Fig. 3.

B. Unsteady Solution

To solve the nonsteady part of the problem, first the linear approximation of the problem is considered¹⁹:

$$r_b(t) = \bar{r}_b + r'_b \exp(i\omega t) \quad r'_b \ll \bar{r}_b \quad (44)$$

$$T_s(t) = \bar{T}_s + T'_s \exp(i\omega t) \quad T'_s \ll \bar{T}_s \quad (45)$$

$$p(t) = \bar{p} + p' \exp(i\omega t) \quad p' \ll \bar{p} \quad (46)$$

$$f(t) = \bar{f} + f' \exp(i\omega t) \quad f' \ll \bar{f} \quad (47)$$

$$\ell_{tra}(t) = \bar{\ell}_{tra} + \ell'_{tra} \exp(i\omega t) \quad \ell'_{tra} \ll \bar{\ell}_{tra} \quad (48)$$

$$T(x, t) = \bar{T}(x) + T'(x) \exp(i\omega t) \quad T'(x) \ll \bar{T}(x) \quad (49)$$

Then, one finds for the thermal profile:

$$1) -\infty < x < -\ell_{tra}$$

$$\frac{\partial}{\partial t}[T'(x) \exp(i\omega t)] + [\bar{r}_b + r'_b \exp(i\omega t)] \frac{\partial}{\partial x}[\bar{T}(x) + T'(x) \exp(i\omega t)] \quad (50)$$

$$= \alpha_c \frac{\partial^2}{\partial x^2}[\bar{T}(x) + T'(x) \exp(i\omega t)] \quad (51)$$

By variable separation, one gets the steady portion whose solution is again Eq. (35) and the unsteady portion

$$i\omega T'(x) + \bar{r}_b \frac{\partial}{\partial x} T'(x) - \alpha_c \frac{\partial^2}{\partial x^2} T'(x) = -r'_b \frac{\partial}{\partial x} \bar{T}(x) \quad (52)$$

whose general solution is

$$T'(-\infty < x < -\ell_{tra}) = C_3 \exp\left(\frac{\bar{r}_b}{\alpha_c} z_1 x\right) + C_4 \exp\left(\frac{\bar{r}_b}{\alpha_c} z_2 x\right) - \frac{\bar{r}_b}{\alpha_c} \frac{r'_b}{i\omega} \frac{T_{tra} - T_0}{\alpha + \beta} \exp\left(\frac{\bar{r}_b}{\alpha_c} x\right) \quad (53)$$

$$2) -\ell_{tra} < x < 0$$

$$\frac{\partial}{\partial t}[T'(x) \exp(i\omega t)] + [\bar{r}_b + r'_b \exp(i\omega t)] \frac{\partial}{\partial x}[\bar{T}(x) + T'(x) \exp(i\omega t)] = \alpha_c \frac{\partial^2}{\partial x^2}[\bar{T}(x) + T'(x) \exp(i\omega t)] \quad (54)$$

By variable separation, one gets the steady portion whose solution is again Eq. (36) and the unsteady portion

$$i\omega T'(x) + \bar{r}_b \frac{\partial}{\partial x} T'(x) - \alpha_c \frac{\partial^2}{\partial x^2} T'(x) = -r'_b \frac{\partial}{\partial x} \bar{T}(x) \quad (55)$$

whose general solution is

$$T'(-\ell_{tra} < x < 0) = C_1 \exp[(\bar{r}_b/\alpha_c) z_1 x] + C_2 \exp[(\bar{r}_b/\alpha_c) z_2 x] - (\bar{r}_b/\alpha_c)(r'_b/i\omega) \Delta \exp[(\bar{r}_b/\alpha_c) x] \quad (56)$$

where

$$z_{1,2} \equiv \frac{1 \pm \sqrt{1 + 4i\omega\alpha_c/\bar{r}_b^2}}{2} \quad (57)$$

The constants C_1 , C_2 , C_3 , and C_4 are to be evaluated by satisfying the following constraints:

$$T'(x \rightarrow -\infty) = 0 \quad (58)$$

$$-\left[\frac{\partial \bar{T}(x)}{\partial x}\right]_{x=-\bar{\ell}_{tra}} \ell'_{tra} + T'(x = -\bar{\ell}_{tra}^-) = 0 \quad (59)$$

$$-\left[\frac{\partial \bar{T}(x)}{\partial x}\right]_{x=-\bar{\ell}_{tra}^+} \ell'_{tra} + T'(x = -\bar{\ell}_{tra}^+) = 0 \quad (60)$$

$$T'(x = 0) = T'_s \quad (61)$$

In addition, energy conservation is required both at the position $x = -\ell_{tra}(t)$, yielding matching of the fluctuating portions of the thermal profile

$$\left[-\frac{\partial^2 \bar{T}}{\partial x^2} \ell'_{tra} + \frac{\partial T'}{\partial x}\right]_{x=-\bar{\ell}_{tra}} = \left[-\frac{\partial^2 \bar{T}}{\partial x^2} \ell'_{tra} + \frac{\partial T'}{\partial x}\right]_{x=-\bar{\ell}_{tra}^+} - \frac{Q_{tra}}{c_c} \frac{r'_b}{\bar{r}_b} \frac{i\omega \ell'_{tra}}{\alpha_c} \bar{r}_b$$

and at the burning surface yielding the fluctuating portion of the thermal gradient

$$\left[\frac{\partial T'}{\partial x}\right]_{x=0^-} = f'$$

By replacing the appropriate thermal profiles, one obtains

$$T'(x \rightarrow -\infty) = 0 \quad (62)$$

$$C_3 \exp[-(\bar{r}_b/\alpha_c) z_1 \bar{\ell}_{tra}] - (\bar{r}_b/\alpha_c)(r'_b/i\omega)(T_{tra} - T_0) = (\bar{r}_b/\alpha_c)(T_{tra} - T_0) \ell'_{tra} \quad (63)$$

$$C_1 \exp[-(\bar{r}_b/\alpha_c) z_1 \bar{\ell}_{tra}] + C_2 \exp[-(\bar{r}_b/\alpha_c) z_2 \bar{\ell}_{tra}] - (\bar{r}_b/\alpha_c)(r'_b/i\omega) \Delta \exp[-(\bar{r}_b/\alpha_c) \bar{\ell}_{tra}] = (\bar{r}_b/\alpha_c)[T_{tra} - T_0 - (Q_{tra}/c_c)] \ell'_{tra} \quad (64)$$

$$T'(x = 0) = C_1 + C_2 - (\bar{r}_b/\alpha_c)(r'_b/i\omega) \Delta = T'_s \quad (65)$$

$$z_1 C_1 \exp\left(-\frac{\bar{r}_b}{\alpha_c} z_1 \bar{\ell}_{tra}\right) + z_2 C_2 \exp\left(-\frac{\bar{r}_b}{\alpha_c} z_2 \bar{\ell}_{tra}\right) - z_1 C_3 \exp\left(-\frac{\bar{r}_b}{\alpha_c} z_1 \bar{\ell}_{tra}\right) - \frac{\bar{r}_b}{\alpha_c} \frac{r'_b}{i\omega} \Delta \exp\left(-\frac{\bar{r}_b}{\alpha_c} \bar{\ell}_{tra}\right) + \frac{\bar{r}_b}{\alpha_c} \frac{r'_b}{i\omega} (T_{tra} - T_0) = \frac{\bar{r}_b}{\alpha_c} \frac{Q_{tra}}{c_c} \ell'_{tra} - \frac{\bar{r}_b}{\alpha_c} \frac{Q_{tra}}{c_c} \frac{r'_b + i\omega \ell'_{tra}}{\bar{r}_b} \quad (66)$$

$$\left[\frac{\partial T'(x)}{\partial x} \right]_{0-} = \frac{\bar{r}_b}{\alpha_c} z_1 C_1 + \frac{\bar{r}_b}{\alpha_c} z_2 C_2 - \left(\frac{\bar{r}_b}{\alpha_c} \right)^2 \frac{r'_b}{i\omega} \Delta = f' \quad (67)$$

By enforcing the boundary condition of Eq. (62), one finds that $C_4 = 0$, whereas Eq. (64) gives $\ell'_{\text{tra}} = \ell'_{\text{tra}}(C_1, C_2)$ and Eq. (63) gives $C_3 = C_3(\ell'_{\text{tra}}) = C_3(C_1, C_2)$. By replacing ℓ'_{tra} and C_3 in Eq. (66) and taking into account Eq. (65), one can evaluate C_1, C_2 . At this point, Eq. (67) is fully determined, which connects the three fluctuating values of burning rate, surface temperature, and thermal gradient. This equation is used to define the intrinsic stability boundary, as discussed next.

C. Intrinsic Stability

The condition of solvability of the homogeneous system comprising the 1) energy boundary condition at the condensed-phaseside of the burning surface, 2) fluctuating burning-rate dependence on the fluctuating thermal gradient, and 3) fluctuating surface temperature on the fluctuating thermal gradient (see Ref. 18, p. 82) is

$$\begin{vmatrix} A_1 & A_2 & A_3 \\ 1 & 0 & -\frac{k/(1-\alpha)}{k/(1-\alpha) + r - 1} \\ 0 & 1 & -\frac{r}{k/(1-\alpha) + r - 1} \end{vmatrix} = 0$$

where

$$A_1 = \frac{(\alpha + \beta)^{1-z_1}}{z_1} + \frac{(z_1 - 1)(\alpha + \beta)^{z_1}}{(z_1 + 2\gamma)z_1 - \gamma} \quad (68)$$

$$A_2 = z_1(\alpha + \beta)^{1-z_1} + \frac{(\alpha + \beta)^{z_1}(z_1 - 1)^3}{(z_1 + 2\gamma)z_1 - \gamma} \quad (69)$$

$$A_3 = -(\alpha + \beta)^{1-z_1} + \frac{(\alpha + \beta)^{z_1}(z_1 - 1)^2}{(z_1 + 2\gamma)z_1 - \gamma} \quad (70)$$

This condition leads to a complex equation describing the linear stability boundary and associated oscillatory burning frequency, at constant pressure, of the stated problem

$$rZ_r + k[Z_k/(1-\alpha)] + Z = 0 \quad (71)$$

Here r and k are real while Z_r, Z_k, Z are coefficients depending on the nondimensional frequency Ω and thermophysical properties of the energetic material (α, β, γ)

$$Z_r = z_1 - 1 + z_1 \frac{(z_1 - 1)^2(\alpha + \beta)^{2z_1 - 1}}{z_1^2 + \gamma(2z_1 - 1)} \quad (72)$$

$$Z_k = \frac{1}{z_1} - 1 + z_1 \frac{(z_1 - 1)(\alpha + \beta)^{2z_1 - 1}}{z_1^2 + \gamma(2z_1 - 1)} \quad (73)$$

$$Z = 1 - \frac{(z_1 - 1)^2(\alpha + \beta)^{2z_1 - 1}}{z_1^2 + \gamma(2z_1 - 1)} \quad (74)$$

Notice that the former (with no transition) μ and k parameters now are defined more generally as

$$\mu' \equiv \frac{1}{(\bar{T}_s - T_0 - Q_{\text{tra}}/c_c)} \left[\frac{\partial \bar{T}_s}{\partial \ell_n \bar{p}} \right]_{\bar{p} = \text{const}} \quad (75)$$

$$k' \equiv (\bar{T}_s - T_0 - Q_{\text{tra}}/c_c) \left[\frac{\partial \ell_n \bar{m}}{\partial T_0} \right]_{\bar{p} = \text{const}} \quad (76)$$

but the pressure Jacobian [see Eq. (5)] is not affected; for $Q_{\text{tra}} \rightarrow 0$, i.e., phase transition with no heat release, the former definitions are recovered. Notice also that for $Q_{\text{tra}} \rightarrow 0$, i.e., $\alpha \rightarrow 0$, one finds

$$Z_r \rightarrow z_1 - 1 \quad (77)$$

$$Z_k \rightarrow (1/z_1) - 1 \quad (78)$$

$$Z \rightarrow 1 \quad (79)$$

thus recovering the previous result (see Ref. 19, p. 619)

$$r(z_1 - 1) + k[(1/z_1) - 1] + 1 = 0 \quad (80)$$

In turn, this leads to the explicit solution (cf. Sec. II)

$$r = \frac{(k-1)^2}{k+1} \quad \text{with } k > 1 \quad (81)$$

$$\Omega_{\text{bif}} = \sqrt{k} \frac{(k+1)}{(k-1)^2} \quad (82)$$

Unfortunately, in this instance, an explicit solution of Eq. (71) is no longer possible, but a parametric representation in the nondimensional frequency Ω can be implemented. Multiplying Eq. (71) separately by Z_k^* and Z_r^* , the following complex relationships are obtained:

$$rZ_r Z_k^* + [k/(1-\alpha)]Z_k Z_k^* + Z Z_k^* = 0$$

$$rZ_r Z_r^* + [k/(1-\alpha)]Z_k Z_r^* + Z Z_r^* = 0 \quad (83)$$

Imaginary parts of Eqs. (83) give

$$r = \frac{\text{Im}(-Z \cdot Z_k^*)}{\text{Im}(Z_r \cdot Z_k^*)}, \quad k = (1-\alpha) \frac{\text{Im}(-Z \cdot Z_r^*)}{\text{Im}(Z_k \cdot Z_r^*)} \quad (84)$$

Thus, Eqs. (84) are parametric representations of the stability boundary in the parameter Ω . The values k and r given by Eqs. (84), for Ω spanning from 0 to infinity, describe a line that is just the wanted stability boundary in the k, r plane. Notice that parameter k without transition is replaced by $k/(1-\alpha)$ when transition occurs.

Representative applications are discussed in Sec. IV.A.

D. Linear Frequency Response Function

The linear frequency response function is defined as

$$R_p(\Omega) \equiv (m'/\bar{m})/(p'/\bar{p}) \quad (85)$$

and, for adiabatic burning, is found to be

$$R_p(\Omega) = \frac{\nu Z + \delta Z_r}{r Z_r + k[Z_k/(1-\alpha)] + Z} \quad (86)$$

Notice that, for $\alpha \rightarrow 0$, i.e., phase transition with no heat release,

$$Z_r \rightarrow z_1 - 1 \quad (87)$$

$$Z_k \rightarrow (1/z_1) - 1 \quad (88)$$

$$Z \rightarrow 1 \quad (89)$$

thus recovering the previous ZN result (cf. Sec. II)

$$R_p(\Omega) = \frac{\nu + \delta(z_1 - 1)}{r(z_1 - 1) + k[(1/z_1) - 1] + 1} \quad (90)$$

Notice that the denominator of Eq. (86) coincides with Eq. (71); as already noticed by Culick,²³ this boundary corresponds to the condition of unbounded response of the burning propellant even for vanishing fluctuations of the forcing term; i.e., it is the intrinsic stability boundary.

Representative applications are discussed in Sec. IV.B.

IV. Representative Results

For the sake of completeness, the pressure dependence of the sensitivity parameters k and r is illustrated in Fig. 4: In both cases, the increase with pressure is due to increasing surface temperature while σ_p is kept constant. A more realistic σ_p , decreasing with pressure (see Fig. 5), contrasts this trend by opposing the increase of surface temperature and thus favoring stability (see discussion in Sec. IV.A). Unfortunately, an exact definition of the σ_p value requires an accuracy of ballistic property measurements that cannot be obtained at this time.

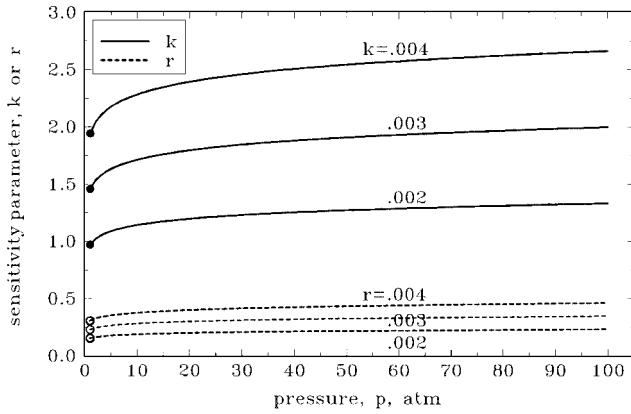


Fig. 4 Dependence of ZN parameters k and r on operating pressure for constant σ_p .

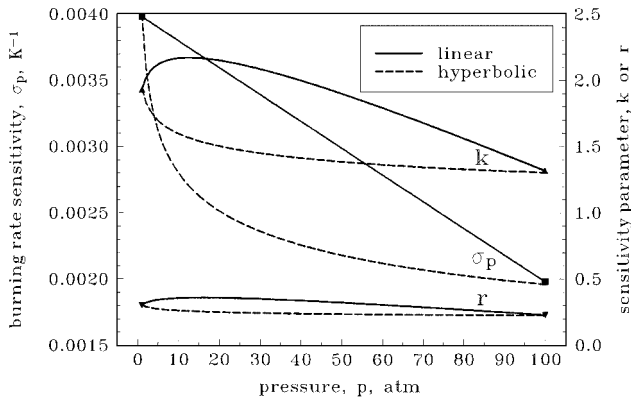


Fig. 5 Dependence of ZN parameters k and r on operating pressure for σ_p decreasing in pressure.

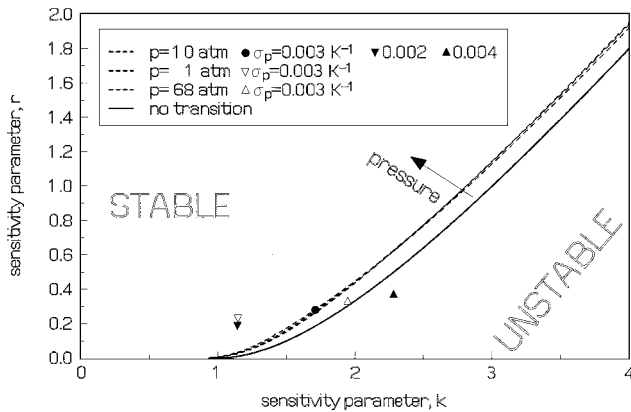


Fig. 6 QSHOD intrinsic stability map for pressure-driven burning, showing the selected test case and several modifications.

A. Intrinsic Stability

The intrinsic linear stability boundary is affected by the phase transition inclusion, as shown in Figs. 6–10. For the enforced set of operating conditions (a typical endothermic transition), the stability region for a given set of k, r parameters apparently is decreased with respect to the no-transition boundary, with little sensitivity to pressure and more sensitivity to $\gamma \equiv \beta/\alpha = -c_c(T_{\text{tra}} - T_0)/Q_{\text{tra}}$. However, this observation might be misleading if one neglects to reexamine simultaneously the possible change of the corresponding operating point on the stability plot.

Several cases are illustrated in Fig. 6. For the selected test case (see Table 1), the corresponding operating point ($\sigma_p = 0.003 \text{ K}^{-1}$ at 10 atm) accidentally falls very close to the new stability boundary (cf. frequency response functions as portrayed in Figs. 11–18).

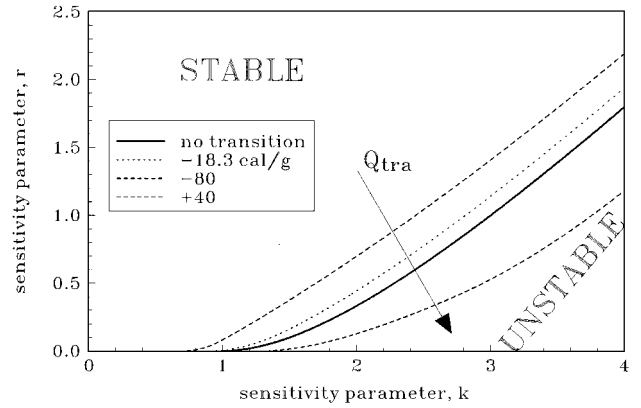


Fig. 7 QSHOD intrinsic stability map for pressure-driven burning, showing dependence of stability boundary on the transition heat release (Q_{tra} parametrically varying from -80 to -18.3 to $+40 \text{ cal/g}$) for the indicated set of operating parameters.

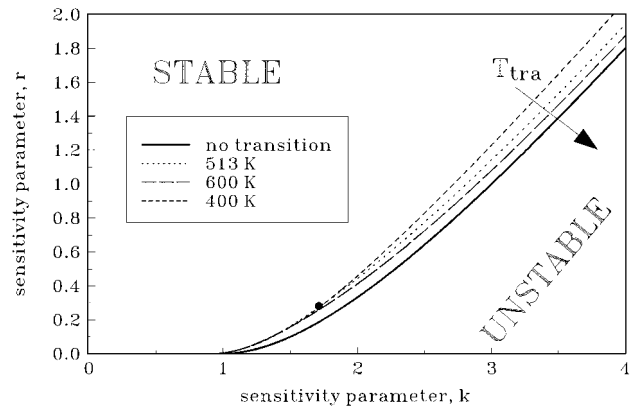


Fig. 8 QSHOD intrinsic stability map for pressure-driven burning, showing dependence of stability boundary on the transition temperature (T_{tra} parametrically varying from 400 to 600 K) for the indicated set of operating parameters.

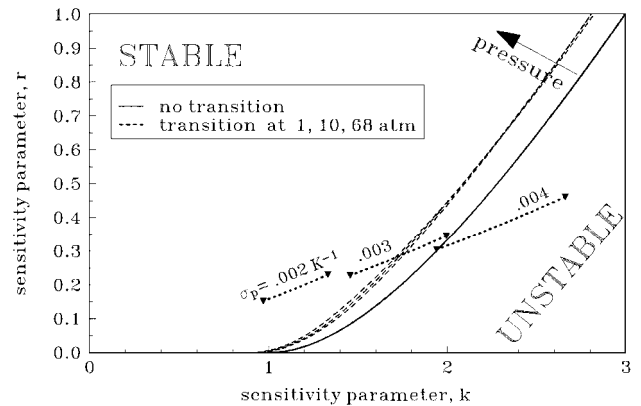


Fig. 9 QSHOD intrinsic stability map for pressure-driven burning, showing loss of stability of the operating point, ranging from 1 to 100 atm, when pressure and/or σ_p are parametrically increased for the indicated set of operating parameters.

Changing either pressure or σ_p (see Table 2) strongly affects the stability features. An increase of pressure (from 10 to 68 atm) at constant σ_p or an increase of σ_p (from 0.003 to 0.004 K^{-1}) at constant pressure is destabilizing; vice versa, a decrease of pressure (from 10 to 1 atm) at constant σ_p or a decrease of σ_p (from 0.003 to 0.002 K^{-1}) at constant pressure is stabilizing. By parametrically varying the phase transition heat release (see Fig. 7), the stability region apparently is decreased for endothermic (likely) transition but increased for exothermic (unlikely) transition with respect to the no-transition boundary; these effects are more evident for larger

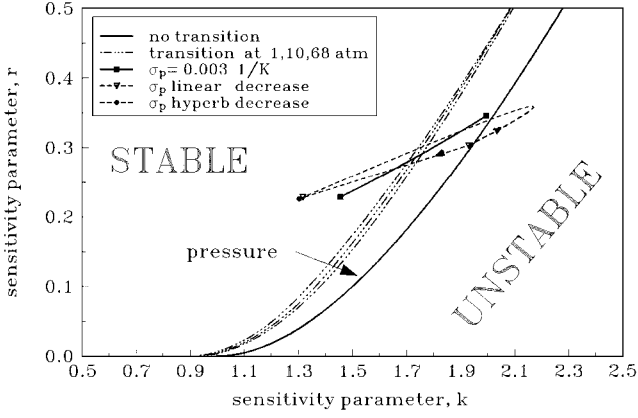


Fig. 10 QSHOD intrinsic stability map for pressure-driven burning, showing gain of stability of the operating point for σ_p decreasing in pressure for the indicated set of operating parameters.

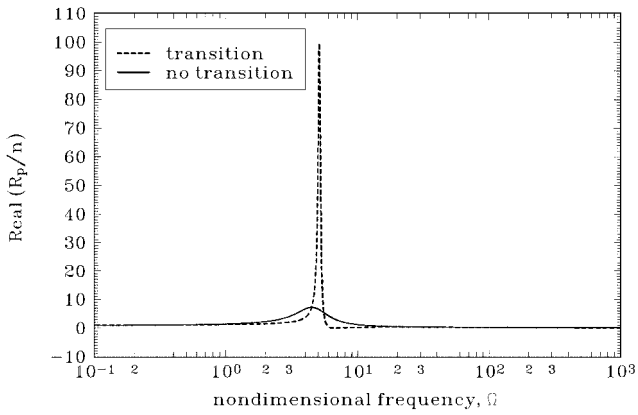


Fig. 11 Frequency response (real part) for pressure-driven burning, showing the effect of phase transition for the test case.

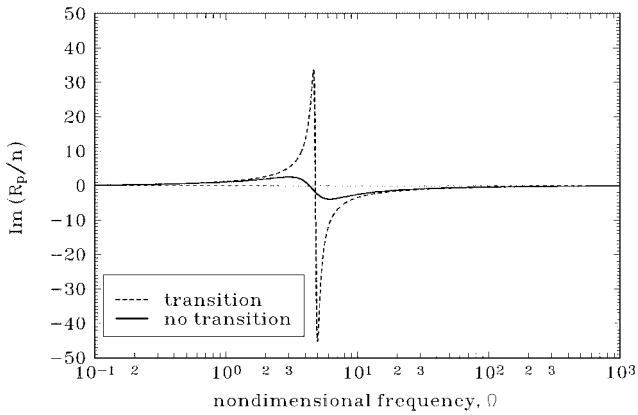


Fig. 12 Frequency response (imaginary part) for pressure-driven burning, showing the effect of phase transition for the test case.

values of Q_{tra} , but, anyway, exothermicity is upper bounded (see Sec. III.A). By parametrically varying the phase transition temperature (see Fig. 8), the main effect seems again to be an apparent shrinking of the stability region; the effect is stronger for decreasing T_{tra} but overall less pronounced than for Q_{tra} . The effect of σ_p assumed to be constant is shown parametrically in Fig. 9 as loss of stability for increasing σ_p , while the operating point ranges from 1 to 100 atm. A more complex behavior is shown in Fig. 10 for σ_p assumed to be decreasing in pressure according to the laws implemented in Fig. 5: For increasing pressure, a linearly decreasing σ_p yields instability and then stability; a faster σ_p decrease yields stability.

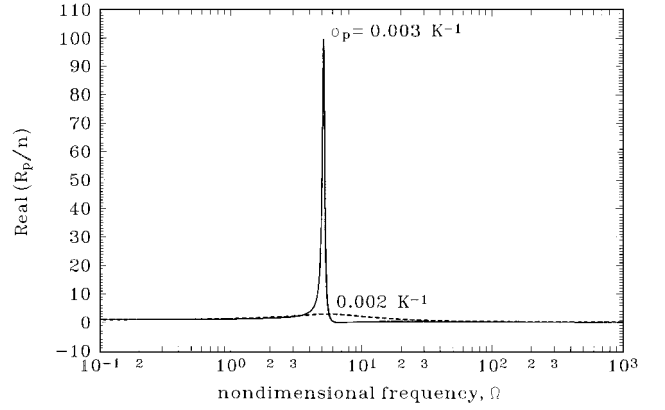


Fig. 13 Frequency response (real part) for pressure-driven burning showing the effect of a σ_p decrease from 0.003 to 0.002 K^{-1} for $p = 10$ atm, $T_{tra} = 513$ K, and $Q_{tra} = -18.3$ cal/g.

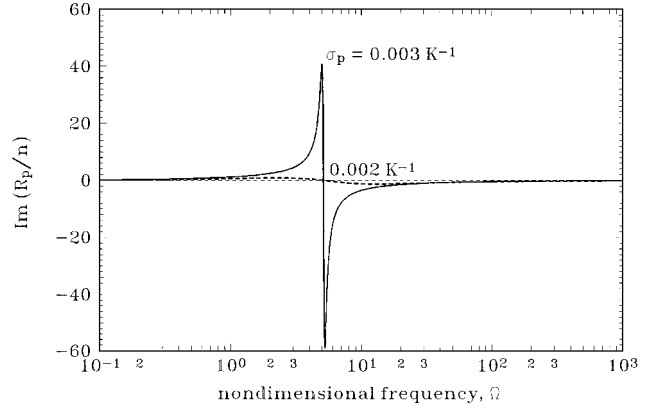


Fig. 14 Frequency response (imaginary part) for pressure-driven burning, showing the effect of a σ_p decrease from 0.003 to 0.002 K^{-1} for $p = 10$ atm, $T_{tra} = 513$ K, and $Q_{tra} = -18.3$ cal/g.

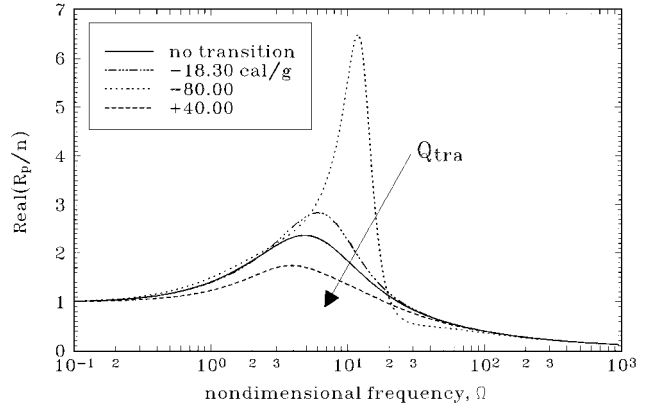


Fig. 15 Frequency response (real part) for pressure-driven burning, showing the effect of the transition heat release (Q_{tra} parametrically varying from -80 to -18.3 to $+40$ cal/g) for $\sigma_p = 0.002$ K^{-1} , $p = 10$ atm, and $T_{tra} = 513$ K.

B. Linear Frequency Response Function

Typical trends for the test case and selected modifications (see Table 2) are illustrated in Figs. 11–18. A direct comparison between frequency response functions computed for the test case with or without phase transition, for the particular operating point selected in Fig. 8, is shown in Figs. 11 and 12 (respectively, real and imaginary parts). Taking into account phase transition brings about a much stronger peak response, although the peak frequency is affected only slightly; this is a consequence of the selected operating point falling almost on the stability boundary (cf. Fig. 6). As a matter of fact, for the selected test case, a decrease of σ_p (from 0.003 to 0.002

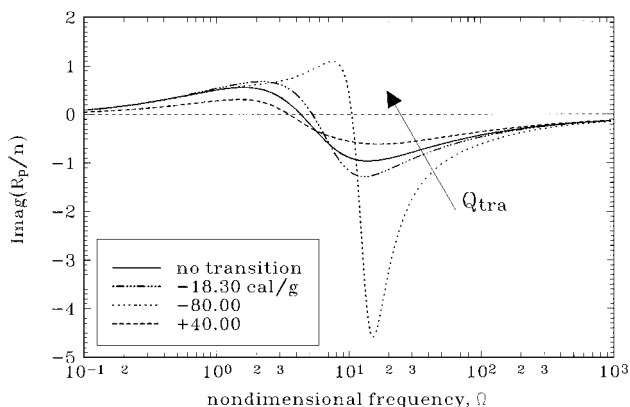


Fig. 16 Frequency response (imaginary part) for pressure-driven burning, showing the effect of the transition heat release (Q_{tra} parametrically varying from -80 to -18.3 to $+40$ cal/g) for $\sigma_p = 0.002$ K $^{-1}$, $p = 10$ atm, and $T_{tra} = 513$ K.

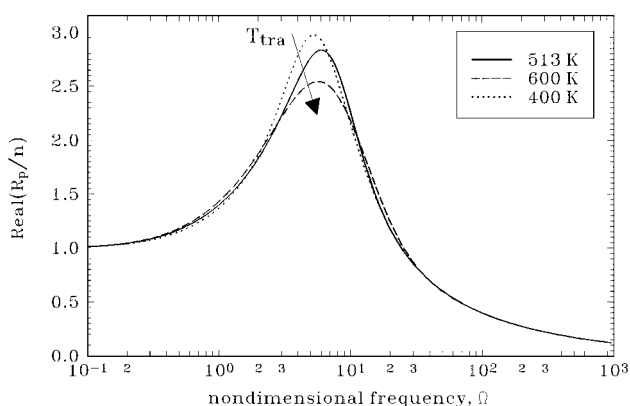


Fig. 17 Frequency response (real part) for pressure-driven burning, showing the effect of the transition temperature (T_{tra} parametrically varying from 400 to 600 K) for $\sigma_p = 0.002$ K $^{-1}$, $p = 10$ atm, and $Q_{tra} = -18.3$ cal/g.

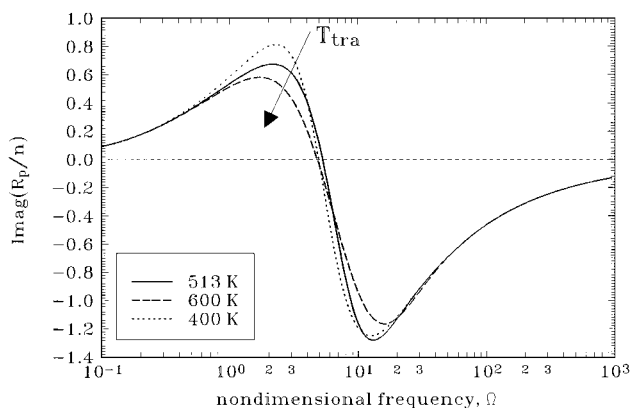


Fig. 18 Frequency response (imaginary part) for pressure-driven burning, showing the effect of the transition temperature (T_{tra} parametrically varying from 400 to 600 K) for $\sigma_p = 0.002$ K $^{-1}$, $p = 10$ atm, and $Q_{tra} = -18.3$ cal/g.

K $^{-1}$) implies a strong reduction of the response, as clearly shown in Figs. 13 and 14. The effect of Q_{tra} is illustrated in Figs. 15 and 16 for $\sigma_p = 0.002$ K $^{-1}$: For this case, increasing or decreasing Q_{tra} yields a much weaker peak response. The effect of T_{tra} is illustrated in Figs. 17 and 18 for $\sigma_p = 0.002$ K $^{-1}$: For this case, increasing or decreasing T_{tra} yields a less pronounced peak response.

The fact that the heat release associated with the phase transition for the test case is a small fraction of the overall condensed-phase heat sink ($\alpha \ll 1$) should not be misunderstood. The unifying interpretation is that phase transition, as any other effect, affects the

response function more when the operating point is closer to the intrinsic stability boundary. Parameters favoring the displacement of the operating point far away from the stability boundary (low σ_p , low pressures, etc.) will manifest few consequences on the frequency response function, and vice versa.

V. Conclusions

Frequency response functions and the intrinsic stability boundary were obtained for a concentrated phase transition in the condensed phase with constant thermophysical properties. Both endothermic and exothermic phase transitions are allowed. The results of the classical no-transition configuration are recovered as a limiting case. The destabilizing effects of large σ_p , or σ_p not decreasing with pressure, are manifest. Thus, the importance of accurate experimental determination of the sensitivity parameters, especially σ_p , is stressed.

An extension including the simultaneous change of thermophysical properties is in progress.

Acknowledgments

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