

Revisiting the Modeling of Ammonium-Perchlorate Combustion: Development of an Unsteady Model

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This paper examines afresh features of ammonium-perchlorate (AP) combustion. Here an AP model, which predicts most experimental observations, is proposed. The one-dimensional aero-thermo-chemical field is captured through the solution of mass, energy, and species conservation equations. The unsteady one-dimensional phase heat transfer accounting for regression is solved for in the condensed phase. The uncertain parameters for AP pyrolysis, gas-phase reaction, and extent of surface exothermicity are chosen so that the most certain of the measured parameters of AP combustion, namely, pressure index of stable combustion (0.77 between 2 and 7 MPa), initial temperature sensitivity of burn rate, σ_p (about 0.0021 to 0.0015 K⁻¹), range of surface temperatures measured and suggested in several studies (~850 to 875 K), and low-pressure deflagration limit (LPDL) at 300 K (~2 MPa), are correctly predicted. The results obtained show the pressure index of AP combustion to be 0.77 and σ_p to be 0.0024–0.0023 K⁻¹. The LPDL is caused by a combination of loss of liquid layer and transient conduction into the condensed phase and not by heat loss.

Nomenclature

A_{DB}	= stability parameter $E_s(T_s - T_i)/RT_s^2$
A_g, A_s	= preexponential factor for gas-phase and surface reactions
C_{pc}, C_{pg}	= specific heats for condensed phase and gas phase
D_i	= diffusivity of the i th species
d	= strand hydraulic diameter, cross-sectional area/strand perimeter
E_g, E_s	= activation energy for gas-phase and surface reactions
F	= factor to account for multidimensional effects in a one-dimensional framework
f	= fraction of AP decomposing to APD at the surface
$H_{fAP}, H_{fAPP}, H_{fAPD}$	= heat of formation of AP, APP, and APD, respectively
H_L	= heat loss per unit volume for individual condensed phase cell
H_r	= Heat of reaction
h	= convective heat-transfer coefficient is obtained from a conservative estimate of $Nu = 10$ based on diameter of strand
k_c, k_g	= thermal conductivity for condensed phase and gas phase
Nu	= Nusselt number
n	= pressure index of burn rate
Q_s	= heat of pyrolysis
R	= universal gas constant
r	= burn rate
S_j	= stoichiometric coefficient of species j
$T_c, T_f, T_g, T_i, T_m, T_s$	= condensed phase, flame, gas phase, initial, melt and regressing surface temperature
u, u_0	= velocity of gases, velocity of the gas at the regressing surface

Y_i	= mass fraction of the i th species
Y_{iv}	= mass fraction of the i th species in the pyrolysis products
Y_{i0}	= mass fraction of the i th species at the surface
α	= stability parameter $C_{pg}T_f/C_{pc}(T_s - T_i)\epsilon$
ϵ	= $[(\eta + 2)/2] + (E_g/2RT_f)$
η	= reaction order
ρ_c, ρ_g	= density of condensed phase and gas phase
σ_p	= initial temperature sensitivity of burn rate
$\dot{\omega}''', \dot{\omega}_j'''$	= reaction rate, reaction rate of j th species

Introduction

ALTHOUGH ammonium perchlorate (AP) has been extensively used as a standard oxidizer in composite propellants for a long time, certain issues related to AP combustion are yet to be resolved. Most models^{1–5} of AP combustion, except those presented by Price et al.,⁶ Sohn,⁷ Manelis and Strunin,⁸ predict correctly the experimentally^{9–11} observed burn-rate pressure index of 0.77. Although some models^{1,5,6,8} have predicted the burn-rate dependence on initial temperature, most^{1,4,5,8} with the exception of Price et al.⁶ predict a higher σ_p than those experimentally obtained by Boggs and Zurn.⁹

Some models^{1,2,3,5} have discussed the low-pressure deflagration limit (LPDL) of AP. These models^{1,2,3,5} with a quasi-steady condensed-phase approximation attribute LPDL of AP to loss of melt layer alone¹ (no heat loss), loss of melt layer accompanied by a heat loss to the surroundings,^{2,5} and to heat loss to the surroundings alone.³ A good review of the literature on LPDL of AP monopropellant combustion can be found in Bruno et al.¹²

Detailed experimental investigation of the LPDL of AP has been carried out by Nir.¹³ He has demonstrated that the LPDL of AP increases with a decrease in the size of the strand, indicating some kind of heat-loss phenomenon to be responsible. The low-pressure deflagration limit of AP reported in literature along with the strand size and initial temperature are presented in Table 1. Apart from Levy and Friedman's¹⁴ data, the rest of the data are within the experimental scatter. It would be difficult to attribute the slightly higher value of LPDL reported by Levy and Friedman¹⁴ to strand size effects, as there are doubts regarding the purity of AP utilized by them. The studies indicated in Table 1 have all determined the variation of burn rate for a particular strand size with pressure and thus determined the LPDL, while Nir¹³ has looked at LPDL alone of different strand sizes. Nir's¹³ experiments do not deal with the determining of burn-rate pressure index. Besides, unlike the studies reported in Table 1 that report a burn rate of around 3 mm/s at LPDL, Nir¹³ reports of

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Table 1 Low-pressure deflagration limit reported in literature

Investigators	T_i , K	LPDL, MPa	Strand size
Levy and Friedmann ¹⁴	298	2.22	4 mm × 4 mm × 38 mm
Hightower and Price ¹⁰	Possibly 299	1.92	8 mm × 2 mm × 10 mm
Beckstead and Hightower ¹⁵	Possibly 299	1.92	8 mm × 2 mm × 10 mm
Watt and Petersen ¹⁶	296	1.9	4 mm × 4 mm × 15 mm
Boggs et al. ¹⁷	299	2.07	2 mm × 6 mm × 10 mm

having obtained LPDL at a burn rate of around 1.5–1.8 mm/s. In view of the disagreement in the literature on the causes for LPDL of AP, it is necessary to examine the issue more carefully.

The choice of appropriate activation parameters and the extent of surface exothermicity are crucial to the success of the model. A brief overview of the literature on the same is presented here. The values reported in literature^{18–24} are wide, ranging from 37.7–210 kJ/g-mole. A closer look at the consolidated data of surface pyrolysis for AP presented in Fig. 5.9 of Williams et al.²⁵ reveals that in the high burn-rate region (closer to the regression rates obtained during deflagration) E_s (extracted as a slope of burn rate vs inverse surface temperature plot) is lower than those encountered at low burn-rate regimes. Thus, while studying the deflagration of AP, preference for a lower value of E_s would be more appropriate as compared to a higher value.

The extent of surface exothermicity (indicated by f fraction of AP decomposing at the surface) has been a point of contention in the literature. Bircumshaw and Newman²⁶ experimentally observed that 30% of AP decomposes ($f = 0.3$) at the surface under isothermal conditions (less than 300°C). Hightower and Price¹⁰ report of having observed a thin frothy liquid layer 2–5 μm thick that exists during the combustion of AP. They report of having found no trace of AP decomposition products in the crystalline phase. Based on these observations, they argued that the pathway for condensed-phase reactions during deflagration (10⁵ K/s) to be different from those observed in differential thermal analysis (DTA) (80 K/s). They postulated that during AP deflagration a majority of the decomposition of AP occurs in the melt layer rather than in or on the crystal. Acheson and Jacobs,²⁷ using DTA and thermogravimetric analysis (TGA), have argued that observed acceleration of AP decomposition by addition of small amounts of magnesium perchlorate was caused by the formation of a melt in which perchlorate ions decomposed at an enhanced rate. The fraction of AP decomposing at the surface under combustion-like conditions still remained undetermined. With this as the background, models^{1,2} have been developed with the fraction of AP decomposing at the surface varying between 75 (surface heat release of 502 kJ/kg) and 70% (surface heat release of 420 kJ/kg), respectively. Pellet²⁸ attempted to determine fraction of AP decomposing at the surface using mass spectrometry on CO₂ laser-assisted pyrolysis of pressed AP pellets. His studies have not been well received because of lingering doubts on whether they really observed condensed phase decomposition. He has reported an f of 0.6 during quasi-steady vaporization when the incident heat flux to the surface was in the range of 8.4–16.8 MW/m². Thus, it is seen that experimental evidence is nebulous.

The knowledge that propellant combustion can be unstable under certain combination of physicochemical parameters is old,²⁹ but most of the earlier investigators,^{1–6,8} with the exception of Strahle³⁰ (who carried out a small-perturbation investigation of AP deflagration to determine the intrinsic stability of a one-dimensional configuration) have not verified if the parameters of AP combustion chosen are in the stable regime. A good review of the literature on unsteady combustion of propellants can be found in review papers of Culick,³¹ De Luca,³² Novozhilov,³³ and Brewster.³⁴

This paper aims to take into account the stability of AP combustion (one-dimensional) in arriving at the uncertain parameters for AP combustion such as the activation parameters for pyrolysis and gas-phase reaction and the extent of surface exothermicity.

They are also chosen so that the most certain of the experimentally obtained parameters of AP combustion, namely, n of 0.77 as reported in literature,^{9–11} σ_p of about 0.0021 to 0.0015 K⁻¹ as reported by Boggs and Zurn,⁹ range of surface temperatures measured and suggested in several studies^{1,2,15,18,35} (~800 to 875 K) and LPDL of AP at 300 K (~2 MPa),^{9,10,13–17} are correctly predicted. The model for AP combustion developed here has been adopted in the sandwich propellant model.^{36,37} The sandwich propellant model^{36,37} has been successful in explaining some of the features like quenching of sandwiches with thin binders below the LPDL of AP, the low-pressure and high-pressure features of AP combustion, and the protrusion of AP close to the AP-binder interface region. The features of this model are to be incorporated in a comprehensive model for composite propellant combustion that is currently under development.

Mathematical Formulation

The computational domain is the region above and below the pyrolyzing surface. The geometry is assumed to be one-dimensional. The coordinate system is fixed to the regressing surface of the burning strand with the unsteady equations solved in both the gas and the condensed phase. This feature allows for the propellant to exhibit inherent instability depending on the choice of certain parameters. The conservation equations solved in the gas phase are those of mass, energy, and species, with pressure being assumed to be constant. These equations are solved in the primitive variables using finite volume discretization with all of the variables being evaluated at the cell centers employing Patankar's^{38,39} algorithm. The condensed phase and the gas phase are solved independently after obtaining the surface temperature. The temperature of the regressing surface is obtained by discretizing and numerically solving the surface heat-flux condition given in the section "Initial and Boundary Conditions." The density is obtained from the equation of state after solving for the energy and species conservation equations. The Lewis and Prandtl numbers are assumed to be unity, and the diffusivities of all of the species are assumed to be identical. First-order-accurate schemes are used for discretizing the time derivative term and the convective term, whereas second-order-accurate schemes are used for discretizing the diffusive terms.

Governing Equations

The governing equations with the usual notations are as given here:

Gas-phase continuity equation:

$$\frac{\partial \rho_g}{\partial t} + \frac{\partial \rho_g u}{\partial x} = 0 \quad (1)$$

Gas-phase energy equation:

$$\frac{\partial T_g}{\partial t} + u \frac{\partial T_g}{\partial x} = \frac{k_g}{\rho_g C_{pg}} \left[\frac{\partial^2 T_g}{\partial x^2} \right] - \frac{H_r}{\rho_g C_{pg}} \dot{\omega}''' \quad (2)$$

Gas-phase species conservation:

$$\frac{\partial Y_i}{\partial t} + u \frac{\partial Y_i}{\partial x} = D_i \left[\frac{\partial^2 Y_i}{\partial x^2} \right] + \dot{\omega}_i''' \quad (3)$$

Condensed-phase energy conservation equation:

$$\frac{\partial T_c}{\partial t} + r \frac{\partial T_c}{\partial x} = \frac{k_c}{\rho_c C_{pc}} \left[\frac{\partial^2 T_c}{\partial x^2} \right] - \frac{H_L}{\rho_c C_{pc}} \quad (4)$$

Heat-Loss Model

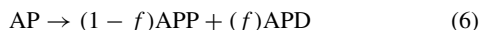
The low-pressure deflagration limit has long been argued to be caused by heat loss in the literature.^{2,3} To understand the role of heat loss in relation to LPDL of AP, an elementary heat-loss model with distributed convective heat loss in the condensed phase has been incorporated. The regressing strand is assumed to lose heat from the side walls and it is modeled as a negative source term.

$$H_L = (4Fh/d)(T_c - T_i) \quad (5)$$

Further discussions on this will be presented in the section titled "Low Pressure Deflagration Limit."

Kinetic Details

The surface decomposition process is taken as



The surface decomposition process is known to occur through a liquid layer (similar to Guirao and Williams² model), which causes the pyrolysis to be exothermic. When the surface temperature goes below 850 K (corresponding^{9,10} to 2.07-MPa pressure and burn rate of 3.3 mm/s), the thickness of the liquid layer begins to decrease and goes to zero when surface temperature becomes equal to the melt temperature¹⁵ of 825 K. As the thickness of the liquid layer decreases, the fraction f of APP pyrolysis product of AP [$\text{NH}_3(\text{gas}) + \text{HClO}_4(\text{gas})$] going to APD (composition obtained from equilibrium decomposition products AP⁴³) at the surface also decreases and goes to zero at $T_s = 825$ K. This is consistent with the surface decomposition being related to the activity in the liquid layer. The fraction f is taken as 0.6 for $T_s > 850$ K, and for $T_s < 850$ K it is assumed to decay linearly in conformity with the liquid layer thickness (justification for the choice of f will be given while discussing the results). A linear relationship connecting surface temperature and f is written as

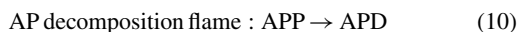
$$f = 0.6 \quad \text{for} \quad T_s > 850 \text{ K} \quad (7)$$

$$f = 0.6 - 0.024(850 - T_s) \quad \text{for} \quad 825 < T_s < 850 \text{ K} \quad (8)$$

$$f = 0 \quad \text{for} \quad T_s < 825 \text{ K} \quad (9)$$

The preceding model intrinsically allows for variation of LPDL with the initial temperature, as variations in the initial temperature do get reflected in T_s .

A single reaction scheme is used for the gas-phase chemical kinetics model.



The present model assumes a single-step gas-phase reaction. The consideration of full chemistry in the gas phase is postponed for a future consideration because it is thought advisable to explore the limits of a single-step chemistry model in predicting the combustion behavior of AP. The kinetic expression is chosen as

$$\dot{w}''' = A_g \rho_g^\eta \exp\left(\frac{-E_g}{RT_g}\right) \prod Y_i \quad (11)$$

The reaction rate of any species j is obtained as

$$\dot{w}_j''' = \sum_j s_j \dot{w}''' \quad (12)$$

Initial and Boundary Conditions

The initial conditions are uniform velocity field (obtained from the surface pyrolysis expression) coupled with the mass fraction of APP and APD being 0.4 and 0.6, respectively, and an ignition

temperature of 950 K in the entire gas phase. The final solution obtained is independent of the initial conditions as a steady-state solution is sought here through an unsteady formulation. The analytical solution corresponding to the surface temperature of 850 K is given as the initial temperature profile in the condensed phase. At the gas-phase exit boundary, the diffusive fluxes are taken as zero while allowing for convective fluxes to cross the boundary. The pyrolysis expression along with continuity of heat and mass flux constitutes the boundary condition at the vaporizing surface. The flux conditions are

$$-D_i \rho_g \left. \frac{\partial Y_i}{\partial x} \right|_0^{0+} = \rho_g u_0 (Y_{iv} - Y_{i0}) \quad (13)$$

$$-k_g \left. \frac{\partial T_g}{\partial x} \right|_0^{0+} = -k_c \left. \frac{\partial T_c}{\partial x} \right|_0^{0-} + \rho_g u_0 Q_s \quad (14)$$

$$Q_s = (C_{pg} - C_{pc})(T_s - T_i) + f H_{fAPD} + (1 - f) H_{fAPP} - H_{fAP} \quad (15)$$

The surface heat release Q_s being positive for endothermic phase change and negative for exothermic phase change. This varies from -205 kJ/kg for $f = 0.6$ at $T_s = 850$ K to 1.85 MJ/kg for $f = 0$ at $T_s = 825$ K. The values of other variables in Eq. (15) are taken from Tables 2 and 3 with $T_i = 299$ K. The pyrolysis expression for the surface is

$$\rho_g u_0 = \rho_c r = A_s \exp(-E_s/RT_s) \quad (16)$$

The condensed-phase thickness is chosen to simulate the infinity condition of ambient temperature.

Choice of Parameters

The thermodynamic data and the kinetic expressions for the various steps are as shown in Tables 2 and 3. The average values of thermodynamic parameters in the condensed phase have been used for the present study in order to keep the model simple. The choice of surface temperature of 850 K at a pressure of 2.07 MPa and 3.3-mm/s burn rate is within the range of measured surface temperature values reported in literature.^{15,18,35} The adiabatic flame temperature is obtained from the equilibrium calculations. This varies with pressure from 1394 K at 2.07 MPa to 1412 K at 13.82 MPa. The value of C_{pg} (see Table 3) is chosen to account for this variation in flame temperature. The experimentally determined value of flame temperature^{35,45} is much lower at around 1200 K. The prediction of experimentally observed temperature profile would need the use of complex chemistry, which is not taken up here. The value of $D_i \rho_g$ at any location is obtained as a function of the temperature of a particular location as given in Table 3. The justification for choosing the values given in Table 3 for the surface pyrolysis and gas-phase reaction activation parameters will be presented while discussing the results.

Table 2 Thermophysical properties of AP

Property	Value	Reference
AP heat of formation	$-2,517,545$ J/kg	JANAF ⁴⁰
Specific heat capacity of condensed phase C_{pc}	$1,602$ J/kg K	Based on average values of JANAF ⁴⁰
Thermal conductivity of condensed phase k_c	0.3 W/m K	Based on average values of Zanotti et al. ⁴¹
Density of condensed phase ρ_c	1957 kg/m ³	Density of orthorhombic phase of Taylor et al. ⁴²
Surface temperature T_s	850 K at a pressure of 2.07 MPa and burn rate 3.3 mm/s	Beckstead and Hightower, ¹⁵ Powling, ¹⁸ Mitani and Niioka ³⁵
Melt temperature T_m	825 K	Beckstead and Hightower ¹⁵

Table 3 Reaction-rate parameters, thermodynamic and transport data

Property	Value	Comment
C_{pg}	$1273.6 - (P - 2.07) \cdot 1.7$ J/kgK	Tuned to obtain an adiabatic flame temperature variation (obtained from equilibrium calculations) of 1394 to 1412 K at an initial temperature of 299 K.
$D_i \rho_g$ (1000 K)	4×10^{-5} kg/ms	In the same range as Guirao and Williams ² $D_i \rho_g \sim T^{0.68}$
Molecular weight of APP	117.5	
Molecular weight of APD	27.8	Obtained from NASA SP-273 ⁴³
Heat of formation of APP	-482,220 J/kg	Calculated with inputs from Narahari ⁴⁴
Heat of formation of APD	-3,898,200 J/kg	Calculated with inputs from Narahari ⁴⁴ and NASA SP-273 ⁴³
A_s	7,864 kg/m ² s	Tuned to get a burn rate of 3.3 mm/s for a T_s of 850 K
E_s	50.24 kJ/g-mole	Obtained through parametric study
A_g	8.55×10^5	Obtained through parametric study
E_g	27.45 kJ/g-mole	Obtained through parametric study
f	0.6	Obtained through parametric study
Order of reaction for AP decomposition flame η	2	At the pressures encountered in AP combustion second-order reaction are important

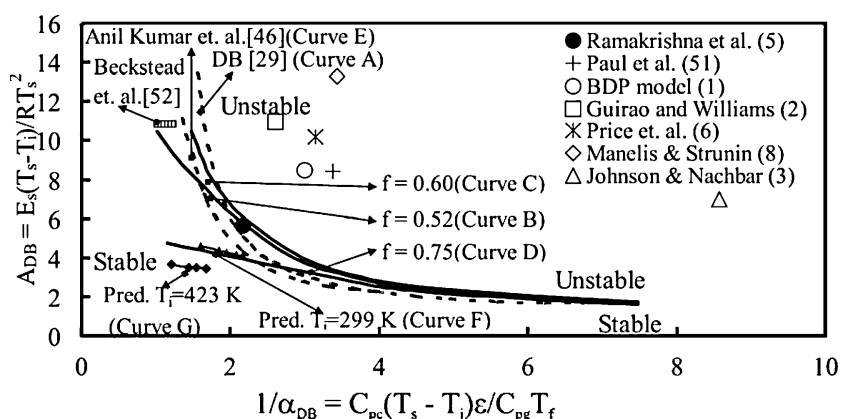


Fig. 1 Predicted results of AP combustion on a Denison and Baum²⁹ stability parameters plot along with the neutral stability curves from Denison and Baum²⁹ and current studies for different values of surface heat release ($f = 0.52$ implies $Q_s = 0$ kJ/kg, $f = 0.60$ implies $Q_s = -205$ kJ/kg and $f = 0.75$ implies $Q_s = -526$ kJ/kg). The results of other models are also plotted.

Grid and Time-Step Details

The cell size in the gas phase is geometrically increased from $0.01 \mu\text{m}$ near the burning surface to $4.35 \mu\text{m}$ at the exit plane for a pressure of 2.07 MPa. The corresponding values for a pressure of 13.82 MPa, where the gradients are steep, are 0.002 and $0.87 \mu\text{m}$, respectively. The number of cells in the gas phase is 250. The cells in the condensed phase are geometrically stretched from $0.01 \mu\text{m}$ near the burning surface to $27.5 \mu\text{m}$ at the exit plane for a pressure of 2.07 MPa. The corresponding values for a pressure of 13.82 MPa are 0.002 and $20.2 \mu\text{m}$, respectively. The number of grids in the condensed phase is 270. The height of the computational domain above the burning surface is $182 \mu\text{m}$ at 2.07 MPa and $36.5 \mu\text{m}$ at 13.82 MPa. The depth of the computational domain below the burning surface is $918 \mu\text{m}$ at 2.07 MPa and $576 \mu\text{m}$ at 13.82 MPa. The time step used in the present calculation is $0.01 \mu\text{s}$. In all of the cases studied, the steady-state solution is obtained with residuals being brought down by at least nine orders of magnitude. All of the solutions obtained are grid and time-step independent.

Results and Discussions

Intrinsic Instability

The stability plot using Denison and Baum²⁹ stability parameters (refer Fig. 1) shows the neutral stability curves obtained for the parameters specified in Tables 2 and 3 for three different surface heat release rates (curves B, C, and D, respectively). Also, shown in Fig. 1 are the neutral stability curves from Denison

and Baum²⁹ and Anil Kumar and Lakshmisha⁴⁶ (curves A and E, respectively).

The neutral stability curves (for the parameters specified in Tables 2 and 3) for a zero surface heat release is obtained as outlined next. For A_{DB} of 10.5 corresponding to a E_s of 116 kJ/g-mole, E_g is varied while holding all other variables in the Denison and Baum²⁹ parameters constant and stability of the solution is verified. The solution obtained is considered stable only if the nondimensional burn rate (nondimensionalized with the burn rate obtained with steady condensed-phase calculations) is invariant (nonzero value) with time as time increases. The values of gas-phase reaction activation energies, $1/\alpha_{DB}$ and the result if the solution is stable or not, are indicated in Table 4. Thus, the neutral stable point in terms of Denison and Baum²⁹ stability parameters for $A_{DB} = 10.5$ is $1/\alpha_{DB} = 1.148$. Similarly, other neutral stable points were determined, and the locus of such points forms the neutral stability line for zero surface heat-release rate (curve B). A similar exercise is repeated for other values of surface heat-release rates and the corresponding neutral stability lines (curves C and D) are as shown in Fig. 1.

The neutral stability curve separates the stable and unstable zones as shown in Fig. 1. From Fig. 1 (curves B, C, and D) it is evident that an increase in the magnitude of surface heat-release rate tends to shrink the stable combustion zone. Similar results have been reported by De Luca and Verri.⁴⁷ It is seen that all neutral stability curves (curve A, B, C, D, and E) tend to merge at the high E_g limit because of gas-phase response time tending to zero, a regime where quasi-steady gas-phase assumptions are valid. This issue is discussed in detail by Anil Kumar and Lakshmisha.⁴⁶

Table 4 Stability of the solution for zero surface heat-release rate and A_{DB} of 10.5

Sl. No.	E_g , kJ/g-mole	$1/\alpha_{DB}$	A_{DB}	Stability
1	2.1	1.090	10.5	Stable
2	4.2	1.137	10.5	Stable
3	8.4	1.230	10.5	Not stable
4	6.3	1.183	10.5	Not stable
5	5.2	1.160	10.5	Not stable

Table 5 Sensitivity study

Parameter	Burn-rate pressure index	Temperature sensitivity of burn rate
$E_g/R = 3800$ K	0.8	0.0026–0.00235 K ⁻¹
$E_g/R = 3300$ K	0.77	0.00247–0.00227 K⁻¹
$E_g/R = 2800$ K	0.73	0.00236–0.00221 K ⁻¹
$E_s/R = 6542$ K	0.74	0.0025–0.00235 K ⁻¹
$E_s/R = 6039$ K	0.77	0.00247–0.00227 K⁻¹
$E_s/R = 5536$ K	0.79	0.0025–0.00223 K ⁻¹
$f = 0.7$ (–560 kJ/kg) ^a	0.693	0.0028–0.0024 K ⁻¹
$f = 0.6$ (–205 kJ/kg)^a	0.77	0.00247–0.00227 K⁻¹
$f = 0.5$ (+147 kJ/kg) ^a	0.81	0.0023–0.00218 K ⁻¹

^aValues indicated within parentheses refer to the surface heat release at 2.07 MPa and $T_i = 26^\circ\text{C}$.

Choice of f , E_s , and E_g

The values for f , E_s , and E_g are chosen in keeping with the philosophy of this paper that the results obtained must agree with the most certain experimental observations stated in the introduction to the paper. Towards this end a parametric study was undertaken; a small portion of the same is presented as the sensitivity analysis in Table 5. The nominal values chosen are indicated in bold, and variations around these parameters are considered.

As already observed, an increase in f or surface heat release shrinks the stable zone (refer Fig. 1). From Table 5, it is noticed that σ_p is strongly affected by the choice of f . Besides, if the heat release is lowered further, the temperature sensitivity approaches the experimental value but in the process makes the surface pyrolysis endothermic. Hence, the choice of $f = 0.6$ is made (σ_p of 0.24%/K, being close to the experimental value) with the expectation that the surface pyrolysis is exothermic in nature as revealed in a significant part of the literature. Once a choice of f is made, the values of E_s and E_g must be so chosen that the results obtained are stable in comparison to curve C (corresponding to $f = 0.6$) of Fig. 1 even at the highest pressure (14 MPa) and highest initial temperature (150°C). This forces a choice of lower E_s , which is in keeping with the observations made in the "Introduction" section. In addition, the particular value of E_s given in Table 3 has been made based on a parametric study as stated earlier.

The gas-phase reaction parameters have been obtained by calibrating them against known experimental data over a wide range of conditions. The detailed chemistry calculations of Tanaka and Beckstead⁴ reveal that the reaction of perchloric acid with radicals such as OH, Cl, and HNO are chain-carrying steps and hence quite important. These reactions have low activation energies, and these could be rate-controlling processes. The use of low values of E_g has led to better prediction of σ_p as evident from Table 5. Analogously, WSB model^{48–50} has been successful in obtaining a better prediction of σ_p for double-base propellants through the use of $E_g/RT \ll 1$ limit. From the preceding discussions, it can be deduced that a choice of lower E_g is more appropriate to the combustion of AP than a higher one. Moreover, stability (comparison to curve C of Fig. 1) as explained earlier restricts the choice of E_g to a low value. The results obtained with the parameters (especially f , E_s , E_g) given in Tables 2 and 3 are stable under all conditions of pressure and initial temperature as seen from Fig. 1 (curves F and G).

The results of some of the earlier models^{1–6,8,51} of AP combustion that have used a steady-state condensed-phase approximation (Denison and Baum²⁹ parameters were calculated based on the pa-

rameters used by them for a burn rate of 3.3 mm/s) are also presented in Fig. 1. The steady-state condensed-phase models^{1–6,8,51} have preferred higher values of E_s and E_g , and these values when used in an unsteady model (like the current one) result in an unstable solution.

In recent times Beckstead and Erickson⁵² have taken experimental data of n , σ_p , and surface temperature of many propellants to determine their response characteristics using flame modeling approaches to combustion instability. The region occupied by Beckstead and Erickson⁵² on the stability plot is in the stable zone with reference to the Denison and Baum²⁹ neutral stability curve (curve A of Fig. 1). But the Denison and Baum²⁹ analysis does not include the surface heat release, which (refer to curves B, C, and D of Fig. 1) is a key parameter that decides the location of the neutral stability curve on the stability plot. Hence, a more cautious approach towards claiming the results as stable would be to verify the time invariance of the nondimensional burn rate.

From the preceding discussions the following broad inferences can be drawn:

1) Comparison of the results of monopropellant combustion with the stability criterion of Denison and Baum²⁹ and other investigators reviewed in Refs. 31–34 does give a reasonable estimate of stability, but it would be appropriate to check the stability of a propellant composition based on the time invariance of the nondimensional burning rate.

2) Low values of E_s and E_g are essential to achieve stability of AP combustion.

3) Previous modeling efforts^{1–6,8,51} have used relatively high E_s reported by experimental investigators^{18–24} and have preferred high-activation-energy asymptotics in the gas phase, which when used in an unsteady model results in an unstable solution.

4) These have not been noticed by them^{1–6,8,51} because of the convention of use of one-dimensional quasi-steady solution in the condensed phase.

Burn-Rate Sensitivity to Pressure and Initial Temperature

With the input parameters given in Tables 2 and 3, the predicted dependence of burn rate on pressure along with experimental values obtained by Boggs and Zurn⁹ and Boggs¹¹ at an initial temperature of 26°C is as shown in Fig. 2. The solid lines correspond to predictions while the experimental values are the dashed lines. The circles indicate experimental results of all other investigators (refer Guirao and Williams²). It is seen that the agreement of predictions with the experiment is good. The predictions show a burn pressure index of 0.77 up to 6.9 MPa, which decreases with further increase in the pressure. These are in agreement with the experimental results of Boggs.¹¹

The predicted variation of melt layer thickness with pressure is also shown in Fig. 2. The melt layer thickness is seen to increase rapidly at low pressure end and slowly decreases towards the high pressure end. Qualitatively similar trends have been reported by

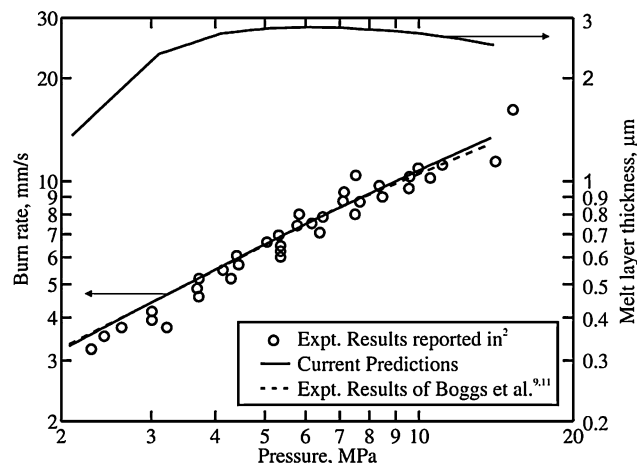


Fig. 2 Predicted values of burn rate and melt layer thickness (top curve) of AP at various pressures along with experimental results.

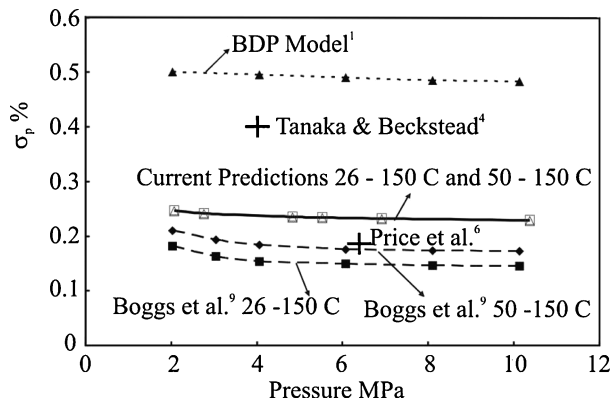


Fig. 3 Predicted initial temperature sensitivity of burn rate as a function of pressure along with experimental results.

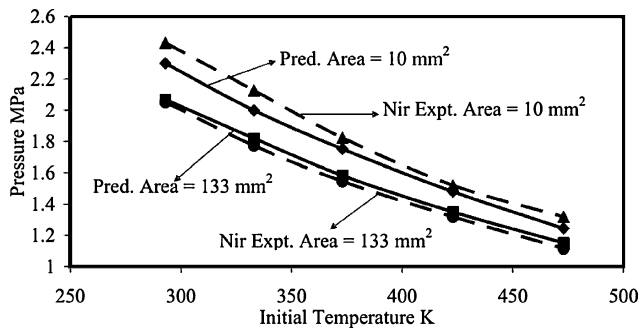


Fig. 4 Predicted values of LPDL of AP as a function of the initial temperature for the different strand sizes along with the experimental results.

Guirao and Williams,² although they observe a sharper increase in the melt layer thickness at the low pressure end.

The predicted initial temperature sensitivity of burn rate along with the experimentally obtained values of Boggs and Zurn⁹ and predicted values of other investigators are shown in Fig. 3. The predicted values of $\sigma_p = 0.0024\text{--}0.0022\text{ K}^{-1}$ are in reasonable agreement with experimental results of $\sigma_p = 0.0021\text{--}0.0015\text{ K}^{-1}$. The results obtained here are better than 0.005 K^{-1} of BDP¹ model, 0.004 K^{-1} of Tanaka and Beckstead,⁴ and 0.003 K^{-1} of our previous work⁵ as evident from Fig. 3. The experimental results of Watt and Petersen¹⁶ and Friedman et al.⁴⁵ have not been used here for comparison, as the AP used by them has been later proved⁵³ to have small traces of impurity, which led to higher-pressure index of burn rate and a higher σ_p . Price et al.⁶ have predicted the values of σ_p that are closer to the experimental values than those obtained here, but the solution obtained by them is in the unstable zone as discussed earlier.

Low-Pressure Deflagration Limit

To understand the role of heat loss in causing LPDL of AP, the heat-loss model outlined in section "Heat Loss Model" was used. The factor F [in Eq. (5)] accounts for multidimensional effects in a one-dimensional framework. This was obtained so as to realize a LPDL of 2.05 MPa for a strand size of 13 mm (identical to those obtained by Nir,¹³ refer to Fig. 4). This value was around 20. This appears high, although it is the best one can get from a one-dimensional model. This needs further careful consideration (multidimensional effects) and has not been pursued here as it has little bearing on the other aspects being examined in this paper.

With the heat-loss model as already outlined, the variation of LPDL of AP with the initial temperature for two different strand sizes along with the experimental results of Nir¹³ are plotted in Fig. 4. The predicted results capture the broad trends exhibited by the experimental results. The predicted and experimental curves have a negative slope indicating a reduction in LPDL with increase in the initial temperature. The two curves for the different strand sizes get

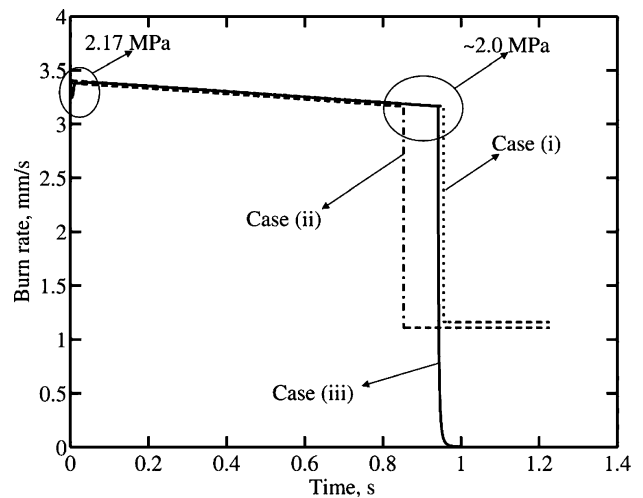


Fig. 5 Variation of burn rate with time for the three different cases when the pressure is gradually reduced from 2.17 to 2.0 MPa for an initial temperature of 293 K.

closer as one moves to a higher initial temperatures pointing to a reduction in the heat-loss effect associated with strand size at higher initial temperatures.

At a pressure of 2.0 MPa and an initial temperature of 299 K for a strand diameter of 25 mm, experimental observation¹³ reveals that AP ceases to burn. This corresponds to the limit wherein the strand size has little effect on LPDL. To assess LPDL correctly, the calculations were started from a steady solution obtained for a pressure higher than the LPDL at 2.17 MPa. The development of the solution in time was then followed by slowly reducing the pressure to a value lower than LPDL (2.0 MPa) at a rate of 0.2 MPa/s. This rate is very much lower than the rates required to quench the propellant by rapid depressurization⁵⁴ (400 MPa/s). The results of these calculations with a melt layer model as described in the section "Kinetic Details" are shown for three cases: 1) when the solution is obtained with quasi-steady condensed-phase approximation without heat loss from the sides (as adopted by BDP model¹), 2) when the solution is obtained with quasi-steady condensed-phase approximation with heat loss from the sides (as adopted by Guirao and Williams²), and 3) when solution is obtained with transient conduction in the condensed phase without the heat-loss model being incorporated (current study).

With quasi-steady condensed phase with and without heat loss (refer Fig. 5), it is possible to obtain steady burning (around 1 mm/s) at pressures below LPDL, despite the loss of liquid layer (as described in section "Kinetic Details") and consequent loss of activity on the surface. When transient conduction in the condensed phase accompanied by the loss of the liquid layer (case 3) is introduced without the heat loss, the results show quenching (zero burn rate). The burn rate obtained with case (3) should have stabilized at some point below 3 mm/s, but because of the nature of the liquid layer model the surface activity and consequently the surface temperature (burn rate) begin to decrease. This causes the surface pyrolysis process to become endothermic (2.035 MJ/kg) from an initial exothermic process (-205 kJ/kg). This in turn increases the temperature profile thickness in the condensed phase (refer to Fig. 6) leading to an increased demand in gas-phase heat flux at the surface with low rates of surface pyrolysis. As this cannot be sustained, the surface temperature and consequently the burn rate fall further taking it away from the equilibrium point. These processes continue until the propellant ceases to burn. Thus, LPDL is obtained with case 3, wherein heat loss was not considered. This conclusively shows that the LPDL of AP is not caused by the heat loss alone as argued by Johnson and Nachbar.³ The LPDL of AP was not attained with conditions defined in cases 1 and 2, indicating that the LPDL of AP is not caused by the loss of liquid layer alone with or without heat loss either as argued by Guirao and Williams² and BDP model,¹ respectively. In conclusion, it can be stated that the transient heat conduction in the

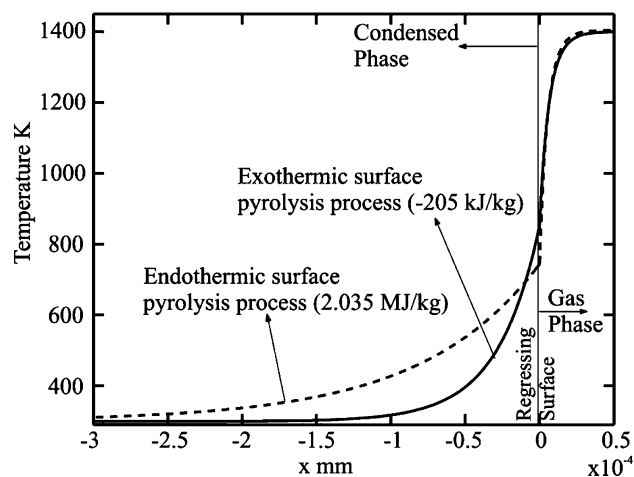


Fig. 6 Temperature profiles in the gas and the condensed phase when the surface pyrolysis process changes from an exothermic process to an endothermic process.

condensed phase accompanied by the loss of liquid layer causes the LPDL of AP.

Conclusions

Computations have been carried out with a simple single-step gas-phase reaction and surface heat-release model with a view of simulating the burn-rate dependence on pressure and initial temperature and LPDL of AP combustion. The pressure index of AP combustion is 0.77 for pressures ranging from 2.07 to 6.91 MPa and decreases from 6.91 to 13.82 MPa, which is in good agreement with the experimental observations. The initial temperature sensitivity of burn rate σ_p is around $0.0024\text{--}0.0023\text{ K}^{-1}$, which is slightly higher than the experimental value of $0.0016\text{--}0.0021\text{ K}^{-1}$. The low-pressure deflagration limit is correctly predicted and is shown to be caused not by heat loss or by loss of melt layer alone but by the loss of melt layer accompanied by transient conduction into the condensed phase. Perhaps, the most important contribution of this work is in identifying stability as an important element in AP combustion.

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