PROPAGATION OF COMBUSTION WAVES IN VAN DER WAALS GASES

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Abstract. This work analyses the propagation of combustion waves in Van der Waals gases. The Chapman-Jouguet approach is adopted and analytical expressions are obtained for the jump conditions across the combustion waves, the Mach number and detonation velocity in the CJ point. The influence of covolumes and intermolecular force parameters on the Rayleigh line and Hugoniot curve are considered. Examples of the application of the theoretical results are presented and a comparison is made with experimental data.

Keywords: Detonation, Deflagration, Chapman-Jouguet, Van der Waals, Hugoniot curve

1. INTRODUCTION

The equation of state (EOS) for a perfect gas can be used with good accuracy at low pressures. At high pressures should be considered the effects of the volume of molecules and the forces of molecular attraction. The equation of Van der Waals (VDW) was the first semi-empirical approach used to represent the behavior of real gases and is an approximation to the Virial EOS with two expansion terms.

The VDW-EOS was derived from the perfect gas equation considering the excluded volume (covolume) of molecules, *b*, which is approximately equal to four times the volume of a molecule, and the effect of molecular attraction forces, which is proportional to the square of the concentration, $a v^2$, where *a* is a constant and *v* is the gas specific volume.

Therefore, the VDW-EOS can be written in the form $\left(P + a/v^2 \right)\left(v - b \right) = RT$ where *P* is the pressure, *T* is temperature and *R* is the gas constant. The constants *a* and *b* can be calculated from critical properties or, alternatively, can be adjusted experimentally for given ranges of pressure and temperature.

Combustion waves propagate in solid, liquid, gas or multiphase medium with different speeds, depending of the initial or boundary conditions. Combustion waves that propagate with subsonic velocities are called deflagrations and usually have speeds less than 1 m/s under ambient conditions and combustion waves that propagate with supersonic velocities are called detonations and usually have speeds of about 1500-2000 m/s under ambient conditions. It has been verified that the deflagration waves are approximately isobaric whereas detonation waves have high compression ratio, varying from 1.5 to 2.5 MPa.

Shepherd (2007) stated that a detonation is not just a shock wave initiated by combustion. It is a combustion wave propagating at supersonic velocities and characterized by a unique coupling between a shock wave and a zone of chemical energy release.

The classical Chapman-Jouguet (CJ) theory considers detonation waves as a discontinuity with an infinite reaction rate. Based on the CJ-theory it is possible to calculate the detonation velocity and jump conditions across the wave, for a specified gas mixture. During the Second World War, the CJ model was improved by Zeldovich, Döring and Von Neumann who considered a finite reaction rate. Their model (ZND) describes the detonation wave as a shock wave immediately followed by a reaction zone, with the thickness of this zone given by the reaction rate. The ZND theory provides the same detonation velocities and pressures than the CJ theory, the only difference between the models is the wave thickness (Zeldovich and Kompaneets, 1960; Williams, 1985; Glassman, 1996; Wingerden et al, 1999).

In real detonations the wave structure is tridimensional due to hydrodynamic instabilities. However the CJ and ZND solutions give good results for the average properties (Fickett and Davis, 2000, Kuo, 2005).

Since the pressures and temperatures attained in combustion waves can be extremely high, it is pertinent to analyse the effects of molecular volume and of the intermolecular attraction on the propagation characteristics of combustion waves.

Next, an analysis of the propagation of combustion waves in VDW gases is presented based on the (CJ) approach (Williams, 1985; Glasmann, 1996). The results are compared to the perfect gas solution. This work extends the theoretical results related to Noble-Abel gases presented previously (Gonzales et al., 2009; Gonzales, 2010).

2. THE CONSERVATION EQUATIONS

Assuming a one-dimensional steady-state flow of a VDW gas, and adopting a fixed coordinate system on the combustion wave, the conservation equations of continuity, momentum and energy are given, respectively, by:

$$\rho_1 u_1 = \rho_2 u_2 = m \tag{1}$$

$$P_1 + \rho_1 u_1^2 = P_2 + \rho_2 u_2^2 \tag{2}$$

$$h_1 + \frac{u_1^2}{2} = h_2 + \frac{u_2^2}{2} \tag{3}$$

The subscripts 1 and 2 denotes properties of reactants and products, respectively, m is the mass flow rate per unit area, P is the pressure, ρ, the density, u, the flow velocity and h is the enthalpy. The VDW-EOS for reactants and products are written, respectively, as:

$$\left(P_1 + a_1/v_1^2\right)\left(v_1 - b_1\right) = R_1 T_1 \quad \text{and} \quad \left(P_2 + a_2/v_2^2\right)\left(v_2 - b_2\right) = R_2 T_2 \tag{4}$$

where the specific volumes are obtained from:

$$v_1 = 1/\rho_1 \quad \text{and} \quad v_2 = 1/\rho_2 . \tag{5}$$

The enthalpy of a real gas (Van Wylen et al., 1994) can be obtained from:

$$dh = c_p dT + \left[v - T\left(\frac{\partial v}{\partial T}\right)_P\right] dP \tag{6}$$

and integrating for a VDW gas with constant properties, it yields:

$$h_1 = h_{f,1} + c_{P,1}^0 T_1 + b_1 P_1 \left(1 + \varepsilon_1\right) - 2 P_1 v_1 \varepsilon_1 \quad \text{and} \quad h_2 = h_{f,2} + c_{P,2}^0 T_2 + b_2 P_2 \left(1 + \varepsilon_2\right) - 2 P_2 v_2 \varepsilon_2 \tag{7}$$

where $c_{P,1}^0$ and $c_{P,2}^0$ are the specific heats at low pressure (perfect gas equivalent), $\varepsilon_1 = a_1/P_1 v_1^2 \ll 1$ and $\varepsilon_2 = a_2/P_2 v_2^2 \ll 1$.

2.1. Rayleigh line

Combining Eqs. (1) and (5) it yields $u_1 = m/\rho_1 = m v_1$ and $u_2 = m/\rho_2 = m v_2$. Substituting into Eq. (2), it gives:

$$\frac{P_2 - P_1}{v_2 - v_1} = -m^2 \tag{8}$$

This equation is known as the Rayleigh line and relates the pressure variation to the specific volume variation along a combustion wave. Eq. (8) is independent of the equation of state. For a VDW gas, Eq. (8) can be rewritten as:

$$\frac{p - 1}{v - 1} = -\mu \tag{9}$$

where $p = P_2/P_1$ and $v = v_2/v_1$ are non-dimensional pressure and specific volume, and $\mu = m^2 v_1/P_1 = u_1^2/P_1 v_1$ is the dimensionless mass flow rate, that can be expressed in terms of the Mach number of the reactants, $M_1 = u_1/c_1$, where c_1 is the speed of sound in the reactants. The speed of sound c can be obtained (Oates, 1984) from:

$$c^2 = \left(\frac{\partial P}{\partial \rho}\right)_s = \gamma v^2 \left(\frac{\partial P}{\partial T}\right)_v \left(\frac{\partial T}{\partial v}\right)_P \tag{10}$$

Substituting the VDW-EOS for the reactants in Eq. (10), gives:

$$c_1^2 = \gamma_1 P_1 v_1 \left(\frac{1+\varepsilon_1}{1-b_1^*} + 2\varepsilon_1 \right) \tag{11}$$

where $b_1^* = b_1/v_1$ is the non-dimensional covolume of reactants. Considering Eq. (11) and rearranging, the expression for μ is:

$$\mu = \gamma_1 \left(\frac{1+\varepsilon_1}{1-b_1^*} + 2\varepsilon_1 \right) M_1^2 \tag{12}$$

where $M_1^2 = \dfrac{1}{\gamma_1} \left(\dfrac{1+\varepsilon_1}{1-b_1^*} + 2\varepsilon_1 \right)^{-1} \left(\dfrac{p-1}{1-v} \right)$ and the specific heat ratio of reactants is $\gamma_1 = \dfrac{\gamma_1^0 (1+\varepsilon_1) - 2\varepsilon_1 (1-b_1^*)}{1-\varepsilon_1 + 2\varepsilon_1 b_1^*}$.

2.2. Rankine-Hugoniot relations

Substituting Eq. (7) into Eq. (3), the equation of energy becomes:

$$c_{P1}^0 T_1 + (1+\varepsilon_1) b_1 P_1 - 2\varepsilon_1 P_1 v_1 + \frac{u_1^2}{2} + q = c_{P2}^0 T_2 + (1+\varepsilon_2) b_2 P_2 - 2\varepsilon_2 P_2 v_2 + \frac{u_2^2}{2} \tag{13}$$

where $q = h_{f,2} - h_{f,1}$ is the heat of reaction. Substituting in Eq. (13) $u_1^2 = m^2 v_1^2$ and $u_2^2 = m^2 v_2^2$ from Eq. (1), $T_1 = P_1 (1+\varepsilon_1)(v_1 - b_1)/R_1$ and $T_2 = P_2 (1+\varepsilon_2)(v_2 - b_2)/R_2$ from Eq. (4), the Eq. (8) for m^2 and dividing by $P_1 v_1$, using the dimensionless variables p and v, the following is obtained:

$$\left[\frac{c_{P,1}^0}{R_1} + \left(\frac{c_{P,1}^0}{R_1} - 2 \right) \varepsilon_1 \right] - \left(\frac{c_{P,1}^0}{R_1} - 1 \right)(1+\varepsilon_1) \frac{b_1}{v_1} + \frac{q}{P_1 v_1} = -\frac{P_2 - P_1}{v_2 - v_1} \frac{(v_2 + v_1)(v_2 - v_1)}{2 P_1 v_1} +$$
$$+ \left[\frac{c_{P,2}^0}{R_2} + \left(\frac{c_{P,2}^0}{R_2} - 2 \right) \varepsilon_2 \right] pv - \left(\frac{c_{P,2}^0}{R_2} - 1 \right)(1+\varepsilon_2) \frac{b_2}{v_1} p \tag{14}$$

Defining the non-dimensional heat release, $\alpha = q/(P_1 v_1)$, the non-dimensional covolumes, $b_i^* = b_i/v_1$, and using $c_{P,1}^0/R_1 = \gamma_1^0/(\gamma_1^0 - 1)$ and $c_{P,2}^0/R_1 = \gamma_2^0/(\gamma_2^0 - 1)$, where γ_i^0 are specific heat ratios at low pressure (perfect gas equivalent), and making several algebraic manipulations, Eq. (14) reduces to:

$$pv - \left[\frac{\gamma_2^0 + 2b_2^* - 1 + 2\varepsilon_2 b_2^*}{\gamma_2^0 + 1 + 2(2-\gamma_2^0)\varepsilon_2} \right] p + \left[\frac{\gamma_2^0 - 1}{\gamma_2^0 + 1 + 2(2-\gamma_2^0)\varepsilon_2} \right] v =$$
$$= \left[\frac{\gamma_2^0 - 1}{\gamma_2^0 + 1 + 2(2-\gamma_2^0)\varepsilon_2} \right] \left\{ 2\alpha + \left[\frac{\gamma_1^0 - 2b_1^* - 1 + 2(2-\gamma_1^0 - b_1^*)\varepsilon_1}{\gamma_1^0 - 1} \right] \right\} \tag{15}$$

The canonical form of a hyperbola is $(p - p_0)(v - v_0) = K^2$, where $p = p_0$ and $v = v_0$ are horizontal and vertical asymptotes, respectively. The canonical form of the hyperbola can be rewritten as:

$$pv - v_0 p - p_0 v + p_0 v_0 = K^2 \tag{16}$$

Comparing this equation with Eq. (15), yields:

$$v_0 = \frac{\gamma_2^0 + 2b_2^* - 1 + 2\varepsilon_2 b_2^*}{\gamma_2^0 + 1 + 2(2-\gamma_2^0)\varepsilon_2} \quad \text{and} \quad p_0 = -\frac{\gamma_2^0 - 1}{\gamma_2^0 + 1 + 2(2-\gamma_2^0)\varepsilon_2}. \tag{17}$$

By algebraic manipulation of Eq. (15), the equation of the Hugoniot curve for VDW gas is obtained:

$$
\left[p + \frac{\gamma_2^0 - 1}{\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2} \right]\left[v - \frac{\gamma_2^0 + 2b_2^* - 1 + 2\varepsilon_2 b_2^*}{\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2} \right] = \left[\frac{\gamma_2^0 - 1}{\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2} \right] \cdot
$$

$$
\cdot \left[2\alpha + \frac{\gamma_1^0 - 2b_1^* + 1 + 2\left(2 - \gamma_1^0 - b_1^*\right)\varepsilon_1}{\gamma_1^0 - 1} - \frac{\left(\gamma_2^0 + 2b_2^* - 1 + 2\varepsilon_2 b_2^*\right)}{\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2} \right]
$$

(18)

If the properties of reactants and products are constant, then Eq. (18) describes a hyperbola. If γ_2 and b_2 are function of the temperature and pressure of the products, then the hyperbola is degenerate. When $\varepsilon_i = 0$ in Eq. (18), the equation of the Hugoniot curve for a Noble-Abel gas (Gonzales et al., 2009) is obtained:

$$
\left(p + \frac{\gamma_2^0 - 1}{\gamma_2^0 + 1} \right)\left(v - \frac{\gamma_2^0 + 2b_2^* - 1}{\gamma_2^0 + 1} \right) = \left(\frac{\gamma_2^0 - 1}{\gamma_2^0 + 1} \right)\left(2\alpha + \frac{\gamma_1^0 - 2b_1^* + 1}{\gamma_1^0 - 1} - \frac{\gamma_2^0 + 2b_2^* - 1}{\gamma_2^0 + 1} \right)
$$

(19)

Initial values for ε_2, b_2 e γ_2^0 can be estimated from the perfect gas solution and, then, more accurate values can be obtained by iteration of the VDW solution.

Figure 1 shows the effects of parameters b_i, γ_i, ε_i and α on the Hugoniot curves for VDW gases.

a) $\gamma_1^0 = \gamma_2^0 = \gamma^0 = 1.3$ b) $\gamma_1^0 = \gamma_2^0 = \gamma^0 = 1.4$
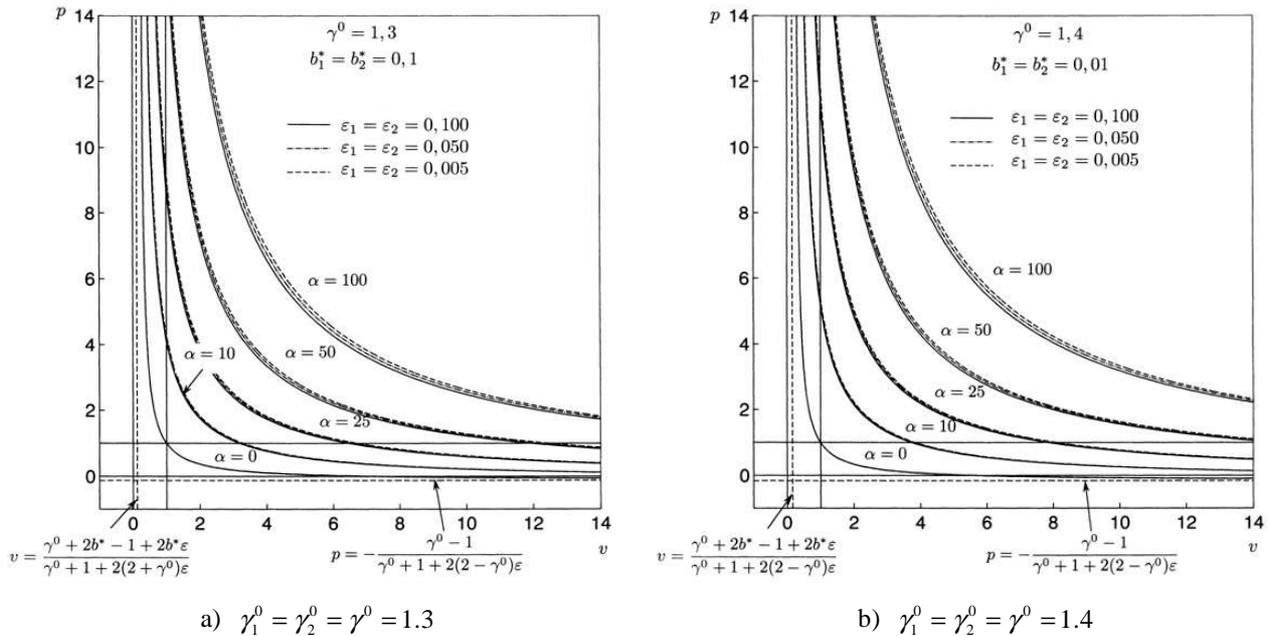
Figure 1. Hugoniot curves for VDW gases.

When the Rayleigh line is tangential to the Hugoniot curve, the upper and lower CJ points are determined. It can be proved that $M_2 = 1$ in the CJ points. The Rayleigh line, Eq. (9), for a VDW gas, can be written as:

$$
\frac{dp}{dv} = -\mu = \frac{p - 1}{v - 1}
$$

(20)

Differentiating Eq. (18) yields:

$$
\frac{dp}{dv} = -\frac{p\left[\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2 \right] + \left(\gamma_2^0 - 1 \right)}{v\left[\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2 \right] - \left[\gamma_2^0 + 2b_2^* - 1 + 2\varepsilon_2 b_2^* \right]}
$$

(21)

Equating the derivatives (20) and (21), an expression for p in terms of v can be obtained and replaced into Eq. (18), then providing the following expressions for p and v:

$$p_\pm = \frac{B}{2A}\left[1 \pm \left(1 - \frac{4AC}{B^2}\right)^{1/2}\right] \tag{22}$$

$$v_\mp = \frac{B'}{2A'}\left[1 \pm \left(1 - \frac{4A'C'}{B'^2}\right)^{1/2}\right] \tag{23}$$

where
$$A = \left(\gamma_2^0 + 1\right)\left[\left(1 - \gamma_2^0 \varepsilon_2\right) - \left(1 - \varepsilon_2\right)b_2^*\right] + 6\varepsilon_2\left(1 - b_2^*\right)$$
$$B = -\left(\gamma_2^0 - 1\right)\left[\gamma_2^0 + 1 + 2\left(2 - \gamma_2^0\right)\varepsilon_2\right]\left(2\alpha + X_1 - 1\right)$$
$$C = \left(\gamma_2^0 - 1\right)\left\{\left[1 + \left(2 - \gamma_2^0\right)\varepsilon_2\right]\left(2\alpha + X_1\right) - \left(1 + \varepsilon_2\right)b_2^*\right\}$$
$$A' = \left(\gamma_2^0 + 1\right)\gamma_2^0\left(1 - 3\varepsilon_2\right) + 2\left(4\gamma_2^0 + 1\right)\varepsilon_2$$
$$B' = -2b_2^*\left(\gamma_2^0 + 1\right)\left(1 - \varepsilon_2\right) - 12b_2^*\varepsilon_2 + \left[\left(\gamma_2^0 + 1\right)\left(1 - 2\varepsilon_2\right) + 6\varepsilon_2\right]\left(\gamma_2^0 - 1\right)\left(2\alpha + X_1 + 1\right),$$
$$C' = \left(\gamma_2^0 - 1\right)\left(2\alpha + X_1 + 1\right)\left[\left(\gamma_2^0 + b_2^*\right)\left(1 - \varepsilon_2\right) + 2\left(b_2^* + 1\right)\varepsilon_2\right] - \left[\gamma_2^0\left(\gamma_2^0 - 1\right)\left(1 - \varepsilon_2\right) + 2\left(\gamma_2^0 - 1\right)\varepsilon_2\right]$$
$$X_1 = \left[\gamma_1^0 - 2b_1^* + 1 + 2\left(2 - \gamma_1^0 - b_1^*\right)\varepsilon_1\right]/\left(\gamma_1^0 - 1\right).$$

In Eqs. (22) and (23), the plus signs correspond to a CJ detonation and the minus signs correspond to a CJ deflagration. Dividing the VDW-EOS for reactants and products gives the temperature ratio:

$$T_\pm = \frac{R_1}{R_2}\frac{\left(1 + \varepsilon_2\right)\left(1 - b_2^*\right)}{\left(1 + \varepsilon_1\right)\left(1 - b_1^*\right)}p_\pm v_\pm \tag{24}$$

The expression for M_1^2 as a function of p and v can be rewritten:

$$M_{1\pm}^2 = \frac{1 - \varepsilon_1 + 2\varepsilon_1 b_1^*}{\gamma_1^0\left(1 + \varepsilon_1\right) - 2\varepsilon_1\left(1 - b_1^*\right)}\left(\frac{1 - b_1^*}{1 + 3\varepsilon_1 - 2\varepsilon_1 b_1^*}\right)\left(\frac{p_\pm - 1}{1 - v_\pm}\right) \tag{25}$$

Figures 2 and 3 show the dependence of the properties of the combustion waves upon the dimensionless heat-release parameter α in VDW gases.

2.4. Stagnation properties

Stagnation properties are obtained when the flow velocity is reduced to zero by an isentropic process. The energy equation can be written as

$$c_v T_0 + P_0 v_0\left(1 - \varepsilon_0\right) = c_v T + Pv\left(1 - \varepsilon\right) + \frac{u^2}{2} \tag{30}$$

where $\varepsilon_0 = a/\left(P_0 v_0^2\right)$, c_v is the specific heat at constant volume and the subscript 0 indicates stagnation condition. Consequently, stagnation temperatures can be obtained from:

$$\frac{T_0}{T} = \frac{1 + \dfrac{\left(\gamma_0 - 1\right)}{\left(1 + \varepsilon\right)\left(1 - b^*\right)}\left[1 - \varepsilon + \dfrac{\gamma^0\left(1 + \varepsilon\right) - 2\varepsilon\left(1 - b^*\right)}{2\left(1 - \varepsilon + 2\varepsilon b^*\right)}\left(\dfrac{1 + \varepsilon}{1 - b^*} + 2\varepsilon\right)M^2\right]}{1 + \left(\gamma_0 - 1\right)\dfrac{\left(1 - \varepsilon_0\right)}{\left(1 + \varepsilon_0\right)\left(1 - b_0^*\right)}} \tag{31}$$

An expression for the stagnation pressure P_0 is obtained from the VDW-EOS. Indices for reactants and products were omitted in Eqs. (30) and (31). Figure 4 shows effects of M, b and ε upon stagnation conditions of VDW gases.

It can be verified in Fig. 4 that the influence of the intermolecular force coefficient is not significant in the range considered, since decreasing it one order of magnitude the variation obtained on stagnation properties by the influence of ε is not perceptible. When the Mach number increases the kinetic energy of molecules and the stagnation temperatures increase and other energy modes in the molecules (rotational and vibrational) are excited, causing a smaller influence of intermolecular forces. For low values of M (from 0 to approximately 3) the influence of intermolecular forces is small. Fig. 4 does not show the influence of γ on stagnation properties but it can be significant.

Once known the stagnation temperatures, their ratio can be calculated. Given the parameters (a_i, b_i, γ_i, R_i and α) of reactants and products, the pressure, specific volume and temperature ratios, Mach numbers and stagnation conditions in the CJ combustion waves (deflagrations and detonations) in VDW gases can be calculated.
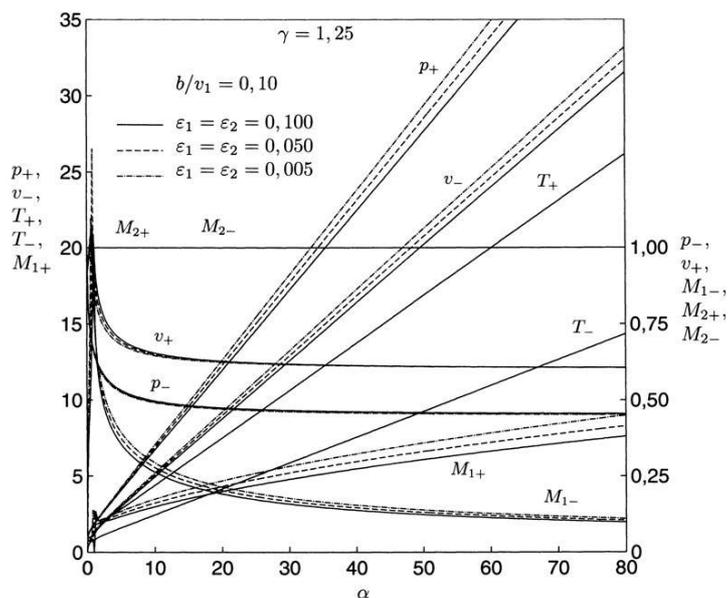
Figure 2. Properties of combustion waves in VDW gases for b^*=0.1=$const$, $\varepsilon_i = const$ and γ^0= 1.25.
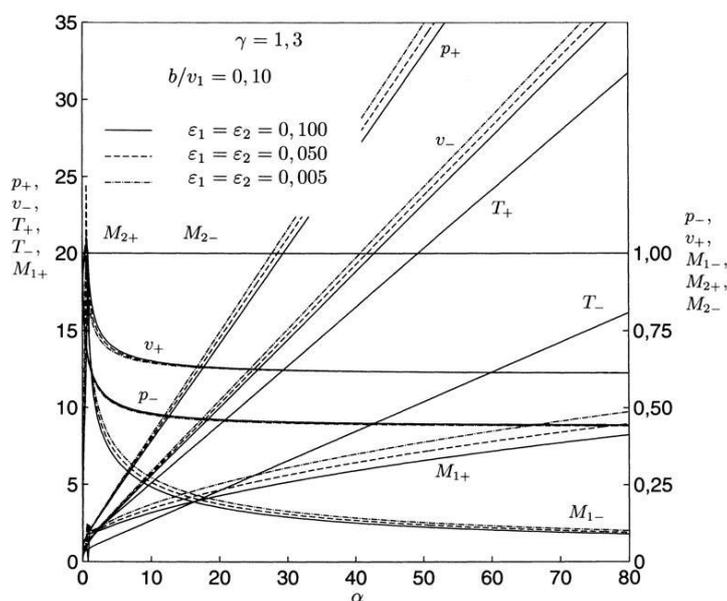
Figure 3. Properties of combustion waves in VDW gases for b^*=0.1=$const$, $\varepsilon_i = const$ and γ^0 = 1.3.
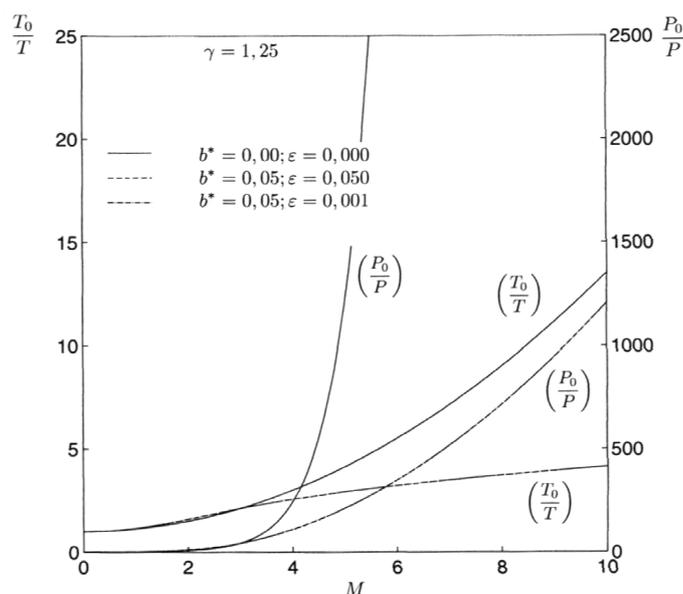
Figure 4. Stagnation properties for VDW gases.

3. EXAMPLES

This chapter shows examples of application of the previous equations for calculations of propagation characteristics of combustion waves in VDW gases. Details of these calculations are described by Gonzales (2009).

3.1. Burning of stoichiometric mixtures of n-octane and air

The burning characteristics of combustion waves propagating in a stoichiometric mixture of n-octane and air are calculated with the derived equations. The parameters used in the calculations were obtained considering three cases: *i)* γ, c_P, R, *a e b* of reactants; *ii)* γ, c_P, R, *a e b* of products; and *iii)* γ, c_P, R, *a e b* average of reactants and products.

Results are depicted in Table 1 and are compared to the results of simulations with the NASA CEA 2004 code. This code considers burning of perfect gases with chemical equilibrium, dissociation of products and different parameters for reactants and products.

Table 1. Properties of combustion waves in stoichiometric mixtures of n-octane and air, for VDW and perfect gases.

Property	VAN DER WAALS GASES			CEA NASA
	reactants	products	reac.& prod.	
P_2 (det.) (MPa)	2.5756	1.8837	1.84598	1.89001
v_2 (det.) (m³/kg)	0.5192	0.53197	0.52773	0.55184
T_2 (det.) (K)	4600.75	3388.6	2977.6	2830.72
P_2 (def.) (MPa)	0.04441	0.04697	0.04696	
v_2 (def.) (m³/kg)	15.445	11.7254	11.4746	
T_2 (def.) (K)	2360	1894.87	1660	

3.2. Effects of initial pressure on detonations of mixtures of propane and diluted air

A study of the effects of initial pressure on the detonation velocity of a mixture of propane and diluted air was made, using the derived equations. The following reaction was considered:

$$C_3H_8 + 4.3O_2 + 8.7N_2 \rightarrow products$$
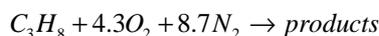
Figure 5 compares theoretical results to experimental results. Theoretical results were obtained for VDW and NA gases using parameters of the products.
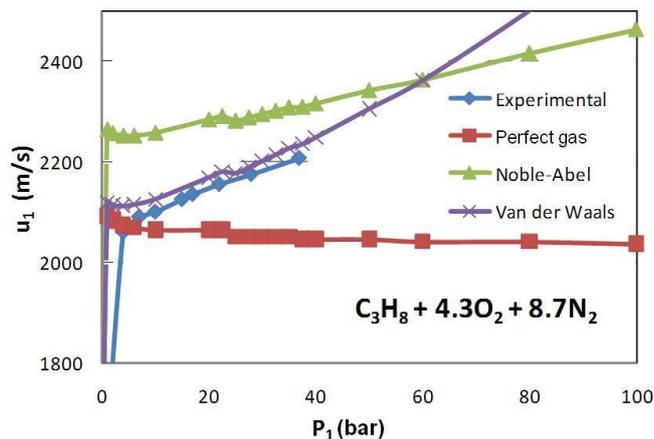
Figure 5. Influence of initial pressure on detonation velocities of a mixture of propane and diluted air.
Source of experimental data: Schmitt and Butler, (2008)

The detonation velocities calculated using the VDW-EOS follow closely the experimental detonation velocities for initial pressures up to 40 bar. Calculated values for detonation velocities using the perfect gas equation are lower than experimental data, whereas results for Noble-Abel gases are larger than experimental data, for initial pressures up to 40 bar. Therefore the effects of intermolecular forces and covolumes should be taken into account for initial pressures above about 3 bar.

4. CONCLUSIONS

This paper presented a simplified description of the propagation of combustion waves in VDW gases, considering the CJ approach. Analytical expressions for the properties along combustion waves were obtained in terms of nondimensional covolumes and intermolecular force parameters. The theoretical results matched closely the experimental results for mixtures propane and diluted air. Effects of initial pressure are significant on detonation velocities.

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