ARTICLE

Improvements of the CARTE Thermochemical Code Dedicated to the Computation of Properties of Explosives

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ABSTRACT: In the field of detonation science or shock physics, predicting the thermo-dynamic properties and chemical compositions of detonation products of CHON systems (outcome of the decomposition of explosives) over a wide range of temperature and pressure with a very small computational cost is of great importance. Thermochemical codes such as the CARTE code use several methods published in the literature to compute the properties of these CHON systems. In this work, we evaluate the accuracy of the recently improved KLRR perturbation method and we propose a new mixing rule to reduce the fluid mixture to an effective pure fluid. Then, the results obtained with these two contributions are presented.
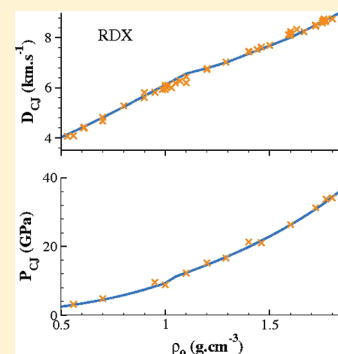
■ INTRODUCTION

The evaluation of high explosive performances is of great interest to shed light on the chemistry and physics of shock and detonation processes. In the ideal description of a detonation,[1,2] the detonation wave brings the inert explosive onto a particular point: the Chapman–Jouguet (hereafter CJ) state. This point lies on the Crussard curve of the detonation products mixture (defined by the Rankine–Hugoniot relations). The CJ point is the starting point of the subsequent isentropic expansion of the detonation products. As a consequence, the prediction of the CJ state properties and then the equation of state of detonation products are crucial to properly model the detonation interaction with the surrounding materials. These properties can be computed by means of the CARTE thermochemical code[3,4] or with other codes such as CHEETAH[5,6] or TDS.[7,8] The general scheme of the code is as follows for a given temperature and density. In a first step, the ideal properties of each species of the detonation products mixture have to be computed. This can be done very simply with the NASA thermochemical tables, for example.[9–12] Then, the effective pure fluid corresponding to a given chemical composition is computed by using a mixing rule such as the one proposed by Ree.[13] With this effective fluid, the excess properties of the mixture can be calculated by different methods such as the MCRSR perturbation method[14] or the HMSA integral theory.[5] In perturbation theories, the free energy of the system is composed of a hard sphere part and a perturbation part which takes into account the real interaction. On the other hand, integral theories are based on the resolution of integral equations (Ornstein–Zernike equation plus a closure relation) to obtain the pair distribution function. In the current version of the CARTE code, NASA tables, Ree's mixing rule, and the MCRSR perturbation method are used.[4] Additional models taking into account ionic and/or multipolar interactions are usually added.[15] Of course, an appropriate calibration of the parameters of the potentials describing the detonation products species has to be performed. An example of such a work is detailed in ref 4.

In this work, we intend to evaluate the accuracy of the MCRSR method and the improved KLRR method proposed recently by Victorov and Gubin[7] by comparison to accurate Monte Carlo (hereafter MC) simulations results from ref 5 and from this work. Then, in a second part, the mixing rule proposed by Ree[13] is compared to reference MC simulation results of binary mixtures. The inaccuracy of this rule led us to propose a new mixing rule. The last part is dedicated to results concerning CJ state properties of model explosives and CJ detonation velocity as a function of the initial density of real explosives.

■ EXCESS PROPERTIES: MCRSR VS IMPROVED KLRR

To evaluate the accuracy of perturbation or integral theories, comparisons to reference MC simulation results can be performed. This work has already been done for HMSA by Fried and Howard.[5] They computed by MC simulations the pressure and the energy of a system of a pure fluid in which particles interact through an EXP-6 potential (composed of three parameters ε, α, and r_m) along several isotherms. These simulation results are the reference data to which perturbation or integral theories have to be compared. Nonetheless, the work of Fried and Howard is limited to temperatures that are relatively high when compared to temperatures occurring at the end of the isentropic expansion or in Diamond Anvil Cell experiments ($\sim$900 K) and to values of the potential parameter α lower than 15.5 and higher than 11.5 (soft repulsion case). These restrictions led us to compute thermodynamic properties for lower temperatures and wider values of α by MC simulations in the same way as ref 5. All the Monte Carlo simulations performed in this work have been done with the GIBBS code, owned by the Institut Français

Table 1. Results of our MC Simulations for α = 11.5, 13.5, and 15.5 and T^* = 2.5[a]

α	T^*	ρ^*	Z	U^*
11.5	2.5	1.00	1.8308 (14)	−1.6554 (3)
11.5	2.5	1.25	3.2870 (15)	−1.8798 (3)
11.5	2.5	1.50	5.8357 (23)	−1.8526 (6)
11.5	2.5	1.75	9.7472 (46)	−1.4531 (14)
13.5	2.5	1.00	2.3972 (13)	−1.5421 (2)
13.5	2.5	1.25	4.4134 (23)	−1.7216 (5)
13.5	2.5	1.50	8.0524 (34)	−1.5867 (8)
13.5	2.5	1.75	13.957 (13)	−0.9351 (34)
15.5	2.5	1.00	2.8416 (15)	−1.4753 (2)
15.5	2.5	1.25	5.3515 (31)	−1.6283 (5)
15.5	2.5	1.50	10.051 (6)	−1.4109 (11)

[a] $T^* = kT/\varepsilon$, $\rho^* = \rho \cdot r_m^3$, and $U^* = U/NkT$ are the reduced temperature, density, and energy, $Z = PV/NkT$. Statistical uncertainties are reported in parentheses; 9.7472 (46) means 9.7472 ± 0.0046.

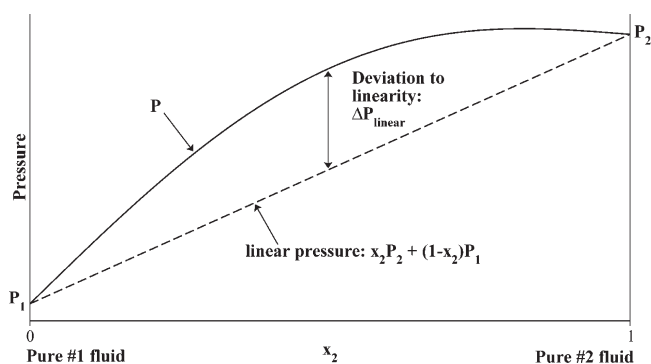**Table 2. Results of Our MC simulations for α = 10.5**

α	T^*	ρ^*	Z	U^*
10.5	2.5	1.00	1.4548 (6)	−1.7451 (2)
10.5	2.5	1.25	2.5821 (9)	−2.0033 (2)
10.5	2.5	1.50	4.5271 (14)	−2.0492 (5)
10.5	2.5	1.75	7.4354 (21)	−1.7953 (8)
10.5	2.5	2.00	11.377 (22)	−1.1729 (9)
10.5	5	0.6661	1.3479 (7)	−0.4825 (1)
10.5	5	0.8000	1.5441 (9)	−0.5564 (2)
10.5	5	0.9998	1.9664 (9)	−0.6376 (1)
10.5	5	1.1003	2.2505 (8)	−0.6608 (2)
10.5	5	1.3336	3.1315 (12)	−0.6521 (4)
10.5	5	1.4991	3.9715 (6)	−0.5796 (2)

Table 3. Results of Our MC Simulations for α = 17.5

α	T^*	ρ^*	Z	U^*
17.5	2.5	1.00	3.2160 (12)	−1.4333 (2)
17.5	2.5	1.25	6.1900 (46)	−1.5691 (7)
17.5	2.5	1.50	11.953 (12)	−1.2847 (22)
17.5	5	0.6661	1.9826 (9)	−0.3782 (1)
17.5	5	0.8000	2.4596 (13)	−0.4320 (1)
17.5	5	0.9998	3.5222 (13)	−0.4697 (1)
17.5	5	1.1003	4.2760 (18)	−0.4585 (2)
17.5	5	1.3336	6.8218 (18)	−0.3071 (3)
17.5	20	1.2728	4.3905 (12)	0.3492 (2)
17.5	20	1.4142	5.3228 (14)	0.4909 (3)
17.5	20	1.7678	8.6287 (18)	1.0488 (4)
17.5	20	2.1213	13.8401 (55)	2.0234 (13)
17.5	20	2.4749	21.7381 (111)	3.6206 (28)
17.5	100	1.7876	4.6920 (12)	0.7441 (2)
17.5	100	2.0831	6.1312 (11)	1.0644 (2)
17.5	100	2.5003	8.8697 (16)	1.6962 (3)
17.5	100	3.4380	19.1772 (59)	4.2300 (18)
17.5	100	3.8198	25.5376 (110)	5.8731 (33)

Figure 1. Linear pressure and deviation to linearity in a binary mixture.

du Pétrole, the Université Paris-Sud, and the CNRS, and developed in collaboration between those three owners and the CEA.[16] Our simulation results in reduced units are gathered in Tables 1−3. Additional curves of preliminary work are reported in ref 17.

These reference results together with the reference results given in ref 5 form a large database of reference data that can be used to evaluate any perturbation method or integral theory dedicated to the computation of excess properties of EXP-6 fluids. One can note that this database, in reduced units, is independent from the ε and r_m values of the parameters of the fluid potential . The MCRSR[14] and improved KLRR[7] perturbation theories are then compared to that database. The relative discrepancies on Z and U^* are reported in Table 3. The improved KLRR method appears to be very accurate. Relative discrepancies of improved KLRR never exceed ∼1% while relative discrepancies of MCRSR are often higher than 2%. On average, the improved KLRR theory is 3 times more accurate than MCRSR. In the work of Fried and Howard[5] (that is on the original database), the relative average error of the HMSA integral method is 0.7% on Z and 1.65% on U^*. The HMSA integral method is therefore more precise than MCRSR but less precise than improved KLRR on that original database. The improved KLRR method is then used in our CARTE thermochemical code.

IMPROVING THE MIXING RULE

Misleadings of the VdW-1f Mixing Rule. To compute the excess properties of detonation products composed of different species, the fluid mixture has to be reduced to an effective pure fluid that mimics the properties of the mixture. The well-known VdW-1f mixing rule proposed by Ree[13] for EXP-6 potentials has been almost always used in the literature (ref 5, for example). This rule reads as follows:

$$r_{m_x}^a = \sum_{ij} x_i x_j r_{m_{ij}}^a \tag{1}$$

$$\varepsilon_x^c r_{m_x}^b = \sum_{ij} x_i x_j \varepsilon_{ij}^c r_{m_{ij}}^b \tag{2}$$

$$\alpha_x^f \varepsilon_x^e r_{m_x}^d = \sum_{ij} x_i x_j \alpha_{ij}^f \varepsilon_{ij}^e r_{m_{ij}}^d \tag{3}$$

where subscript x stands for the effective fluid and ε_{ij} stands for the cross-interaction parameter ε between species i and j. The cross-interaction parameters are defined by the extended Lorentz−Berthelot rules. x_i is the molar fraction of species i.

Parameters (a, b, c, d, e, f) are equal to $(3, 3, 1, 3, 1, 1)$ in the original work of Ree.

That mixing rule has been deduced from comparisons to global thermodynamic properties of H_2–He binary mixtures computed by MC simulations. From that point of view, results obtained with the VdW-1f rule seem to be in fair agreement with MC results. Nevertheless, in our opinion, this is not relevant. In a binary mixture, one can define, for example, the linear pressure

Table 4. Relative Discrepancies ΔZ and ΔU^* in % between Perturbation Theories and Monte Carlo Reference Results for Each Value of α (Averaged over the Different Isotherms) and Global Relative Discrepancies

	MCRSR		KLRR	
α	ΔZ	ΔU^*	ΔZ	ΔU^*
10.5	3.91	1.71	0.95	0.82
11.5	2.72	2.24	0.71	1.01
13.5	1.81	2.06	0.56	0.70
15.5	1.49	3.00	0.74	1.06
17.5	1.08	2.90	0.44	0.64
all	2.20	2.38	0.68	0.85

Table 5. Fictitious Fluids Used in the MC Simulations of Binary Mixtures

fluid	α	ε (K)	r_m (Å)	M (g·mol^{-1})
A	12.8	108	4.17	28
B	11.8	419	4.22	44
C	11.5	310	3.3	18
D	10.9	265	3.9	17
E	14.2	200	4.2	44
F	11.4	300	3.4	18

P_{linear} and the deviation to linearity ΔP_{linear}, which are simply (see also Figure 1) $P_{linear} = P_1 \cdot (1 - x_2) + P_2 \cdot x_2$ and $\Delta P_{linear} = P - P_{linear}$. The deviation to linearity is generally very small when compared to the pressures P_1 and P_2 of the pure fluids.

To check the accuracy of the VdW-1f mixing rule, reference MC simulations of various binary mixtures have been performed. First, six fictitious fluids are defined (see Table 5). The values of parameters of these fluids span the usual range of values. For a given temperature and a given molar volume, pressure and energy are computed for three chemical compositions (0.25, 0.50, and 0.75) together with the same quantities for the pure fluids. Results of these MC simulations are reported in Table 6 and in Figure 2. The computed pressures span the usual high pressure regime in detonation processes. Then, Monte Carlo simulations of the corresponding VdW-1f effective pure fluids are performed and compared to the reference database. Results concerning the deviation to linearity are reported in Figure 2. Large discrepancies (up to 50%) can be viewed, even in the case of the H_2–He mixture from Ree. Similar results are observed for reduced energy. This confirms our initial intuition that the VdW-1f mixing rule proposed on the basis of global thermodynamic properties is not relevant.

Calibration of a New Mixing Rule. To calibrate a new mixing rule that captures the deviation to linearity, the use of Monte Carlo simulations of effective fluid is not possible due to the huge computational cost that would be required. Computing the 42 properties of the effective pure fluids, that is, evaluating one particular mixing rule, costs 68 h on 15 CPU. To circumvent this problem, the improved KLRR perturbation method can favorably be used. Indeed, the same work made with the improved KLRR method can be done in only a few seconds provided the results do not stray too much from the MC simulation results of effective fluids. Results obtained with the improved KLRR method and the VdW-1f mixing rule are reported in Figure 2. The agreement between MC simulations of effective fluids and KLRR calculations of the same fluids is very good.
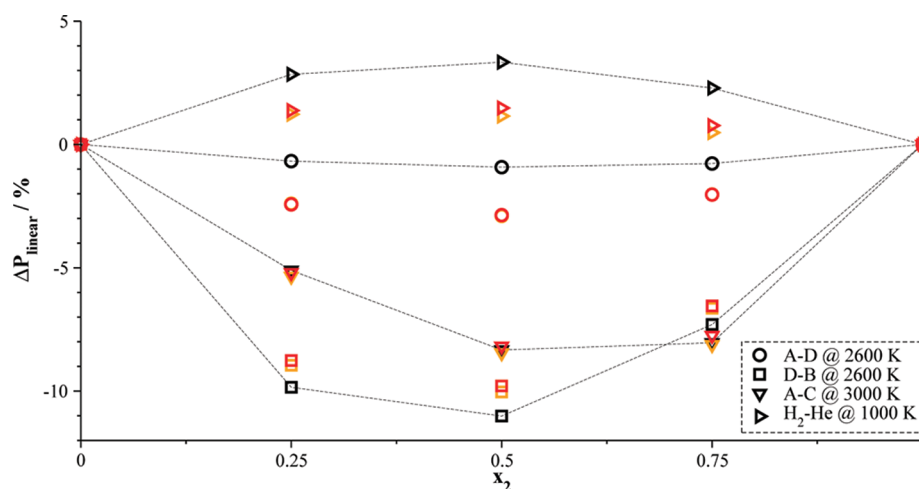
Table 6. Pressures P (Upper Number) in GPa and Reduced Energy U^* (Lower Number) for Various Binary Mixtures for Given Temperatures T in K and Given Molar Volumes v in cm^3·mol^{-1} ^{a}

fluids				x_2				
#1	#2	T	v	0.00	0.25	0.50	0.75	1.00
A	D	2600	11.2	47.02 (2)	43.01 (2)	39.22 (1)	35.59 (2)	32.15 (1)
				6.740 (6)	6.031 (5)	5.328 (4)	4.624 (4)	3.925 (3)
D	B	2600	21.642	4.266 (1)	5.319 (4)	6.704 (3)	8.498 (4)	10.801 (6)
				0.199 (1)	0.277 (1)	0.414 (1)	0.631 (2)	0.955 (2)
A	C	3000	16.26	14.707 (4)	11.679 (5)	9.082 (6)	6.904 (4)	5.108 (4)
				2.101 (1)	1.4202 (9)	0.8476 (13)	0.378 (1)	0.0028 (6)
A	B	3600	16.26	15.795 (7)	18.920 (7)	22.758 (11)	27.461 (9)	33.18 (1)
				1.921 (1)	2.186 (1)	2.515 (2)	2.933 (2)	3.471 (3)
A	D	3600	11.2	50.36 (2)	46.16 (2)	42.17 (2)	38.343 (8)	34.683 (7)
				5.316 (3)	4.790 (3)	4.269 (3)	3.747 (2)	3.225 (1)
D	B	3600	35.723	1.726 (1)	1.916 (1)	2.145 (1)	2.421 (2)	2.757 (1)
				0.0016 (4)	−0.0233 (3)	−0.0476 (3)	−0.0690 (7)	−0.0854 (5)
E	F	3000	16.26	31.079 (24)	20.859 (16)	13.744 (11)	8.938 (8)	5.765 (5)
				3.9780 (51)	2.3556 (36)	1.2570 (23)	0.5427 (16)	0.0946 (10)
H_2	He	1000	9	5.852 (1)	5.252 (2)	4.507 (2)	3.699 (1)	2.871 (1)
				1.5025 (6)	1.3070 (7)	1.0655 (7)	0.8101 (6)	0.5530 (5)

^{a} Statistical uncertainties are reported in parentheses. The results for the H_2–He mixture correspond to the simulations of Ree.[13]

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Figure 2. Deviations to linearity of pressure for four of the eight binary mixtures computed. Different symbols correspond to the four binary mixtures. Black symbols, MC simulations of the real mixture (with two interacting species); red symbols, MC simulations of the corresponding VdW-1f effective pure fluid; yellow symbols, improved KLRR calculations of the corresponding VdW-1f effective pure fluid. Dashed lines are guides to the eyes.
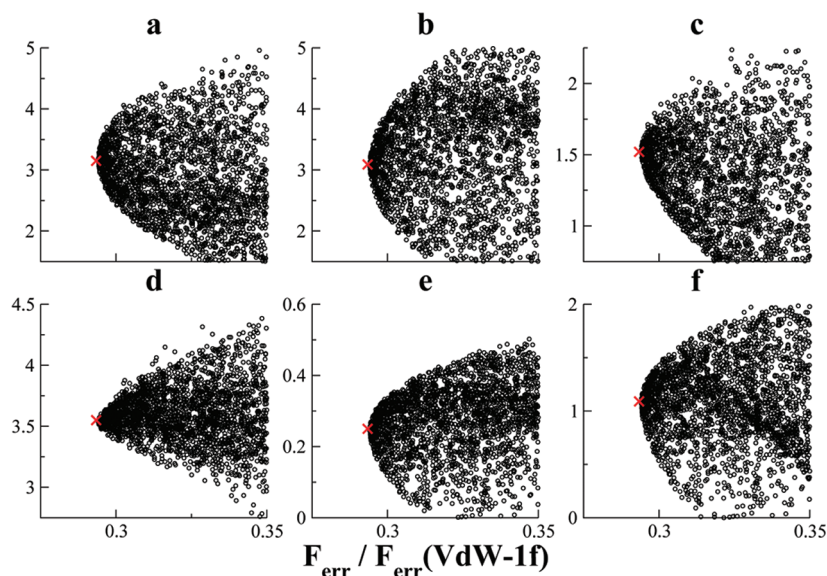

Figure 3. Calibration of the six parameters (a, b, c, d, e, f). The abscissa is the ratio of the error functions $F_{err}/F_{err}(\text{VdW-1f})$ measuring the discrepancy with MC simulation results. The minimum is marked with red crosses.

Calibration of the mixing rule by means of KLRR calculations can be performed.

The calibration of the parameters (a, b, c, d, e, f) on the reference MC simulation results of binary mixtures is realized. This consists of a minimization step using a Nelder–Mead scheme with random starting points. The error function which measures the difference between the exact MC simulation results and the calculations made with a particular mixing rule τ is

$$F_{err}(\tau) = \left(\frac{1}{N}\sum_{i=1}^{N} \left[\frac{P_i(\tau) - P_i(\mathscr{B})}{\delta P_i(\mathscr{B})} \right]^2 + \frac{1}{N}\sum_{i=1}^{N} \left[\frac{U_i(\tau) - U_i(\mathscr{B})}{\delta U_i(\mathscr{B})} \right]^2 \right)^{1/2}$$

(4)

where $N = 24$ (the 24 points of Table 6 for which $0 < x_2 < 1$), $P_i(\tau)$ is the deviation pressure of mixture i computed with the

rule τ, $P_i(\mathscr{B})$ is the reference deviation pressure of mixture i given by MC simulations, and $\delta P_i(\mathscr{B})$ is the statistical uncertainty of the MC simulations. The results are reported in Figure 3. An obvious minimum, called VdW-6p in the sequel and marked by red crosses in Figure 3, is obtained at (3.15, 3.09, 1.52, 3.55, 0.25, 1.09). In Figure 4, we reported the relative deviation to ideality for pressure obtained with the VdW-6p mixing rule. It then clearly appears that the VdW-6p mixing rule brings an important improvement in the computation of the thermodynamic properties of binary mixtures. We point out here that the VdW-6p rule is fitted on properties computed at high pressure and temperature; hence, this rule could not be well suited to the computation of properties in a very different regime (very low temperature, for example). The VdW-6p mixing rule is now used in the CARTE code.
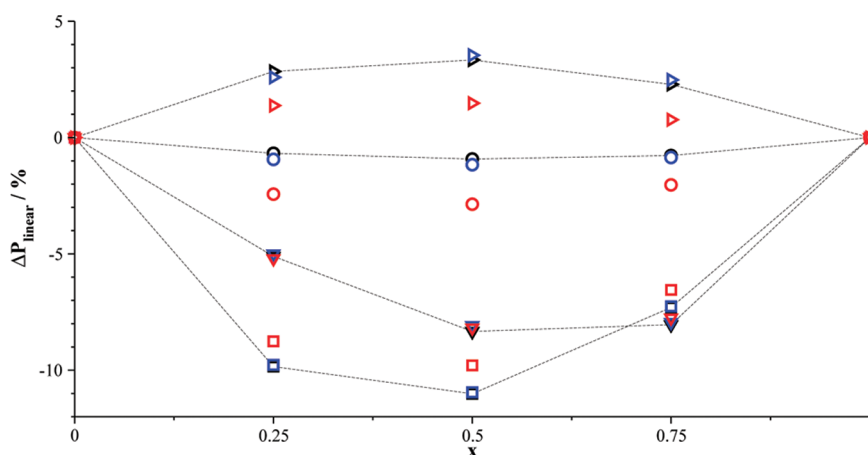
Figure 4. Relative deviation to ideality of pressure for four different binary mixtures. Same legend as that for Figure 2. Blue symbols: MC simulations of the corresponding VdW-6p effective pure fluid.

Test Cases: TNM, HNE, PETN, RDX. By Monte Carlo simulations, CJ states of explosives can be computed.[18] Such a work has been done for four explosives: TNM (CN_4O_8), HNE ($C_2N_6O_{12}$), PETN, and RDX. Detonation product species for each explosive are detailed in Table 7, and parameters of potential for these species are taken from ref 19. The goal of this part is not to reproduce the experimental CJ properties but to evaluate the accuracy of calculations of CJ states using both improved KLRR and VdW-6p when compared to reference MC simulation results.

These CJ state properties have also been computed in three different ways—MCRSR with VdW-1f, KLRR with VdW-1f, and KLRR with VdW-6p (all other things being equal)—and are reported in Table 8.

MCRSR calculations with the VdW-1f mixing rule reproduce CJ state properties computed by MC simulations with a 1.5—2% accuracy. Although not so bad, turning to improved KLRR enables the accuracy to be enhanced between 0.2 and 1.16%. Using the improved KLRR method together with the VdW-6p mixing rule enables one to reproduce reference Monte Carlo simulation results with an average error of 0.25% on the CJ state properties.

CJ State Properties for Usual Explosives: D_{CJ} as a Function of ρ_0. The improved KLRR perturbation theory and the VdW-6p mixing rule have proven to be very accurate when compared to reference Monte Carlo simulation results. For the CARTE code to provide accurate thermodynamic properties and chemical compositions of a large variety of explosives, a huge calibration procedure has been performed. This procedure together with details concerning the equation of state of the solid carbon phase are fully described in ref 4 and hence not repeated here. Then, the CJ detonation velocity can be computed as a function of the initial density of the explosive. Results for six common explosives are detailed in Figures 5—10 and compared to experimental results. The CARTE code reproduces with a high accuracy the CJ state properties of these explosives.

Table 7. Detonation Product Species Used to Compute the CJ State Properties of the Explosives

species	TNM	HNE	PETN, RDX
CO_2, N_2, NO	✓	✓	✓
O_2	✓	✓	
CO		✓	✓
H_2O, H_2, NH_3			✓

Table 8. CJ State Properties of the Four Explosives Computed by Reference Monte Carlo Simulations (MC), MCRSR + VdW-1f (Named A), KLRR + VdW-1f (B), and KLRR + VdW-6p (C) (the Relative Average Difference Is Also Reported)

		P_{CJ} (GPa)	T_{CJ} (K)	D_{CJ} (m·s^{-1})	overall diff (%)
TNM	MC	14.97	2250	6364	
	A	15.50	2228	6464	2.03
	B	15.19	2225	6421	1.16
	C	14.97	2238	6381	0.27
HNE	MC	6.14	6017	4828	
	A	6.29	6048	4907	1.53
	B	6.19	6026	4853	0.49
	C	6.16	6038	4837	0.29
PETN	MC	8.32	4677	5599	
	A	8.57	4686	5692	1.62
	B	8.34	4669	5612	0.21
	C	8.29	4671	5591	0.21
RDX	MC	9.38	4325	6028	
	A	9.72	4331	6143	1.89
	B	9.45	4313	6061	0.53
	C	9.34	4314	6027	0.23

■ CONCLUSION

We have evaluated the accuracies of the largely known MCRSR perturbation method and the recently improved KLRR one. The improved KLRR method has been shown to reproduce with a close agreement the reference Monte Carlo simulation results of a pure fluid in a large range of thermodynamic conditions. In a second step, comparisons with Monte Carlo simulation results of binary mixtures showed that the largely used VdW-1f mixing rule used to reduce a mixture to an effective pure
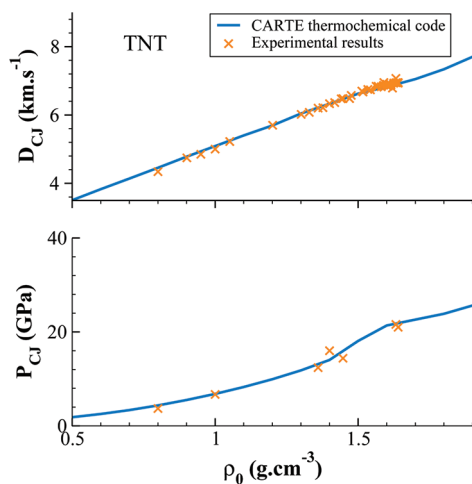
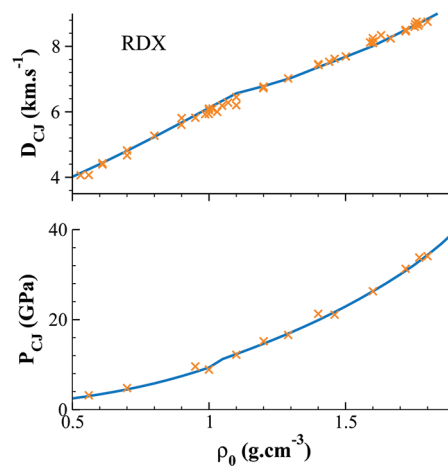**Figure 5.** D_{CJ} and P_{CJ} as functions of the initial density for TNT.

Figure 8. D_{CJ} and P_{CJ} as functions of the initial density for RDX.
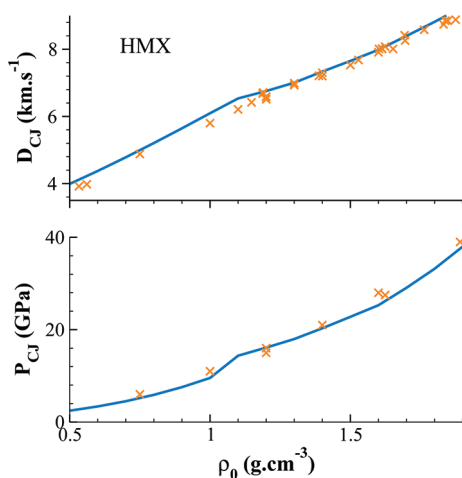

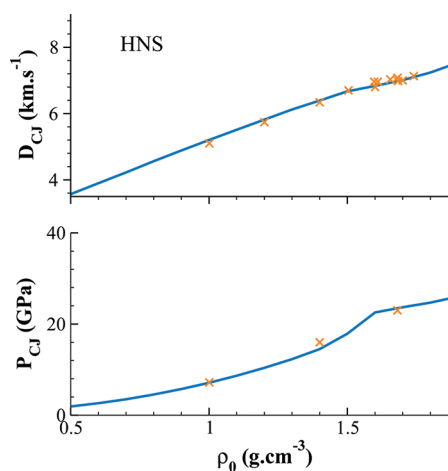**Figure 6.** D_{CJ} and P_{CJ} as functions of the initial density for HMX.

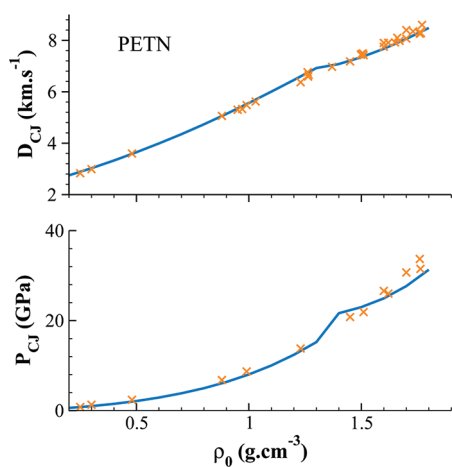**Figure 9.** D_{CJ} and P_{CJ} as functions of the initial density for HNS.

Figure 7. D_{CJ} and P_{CJ} as functions of the initial density for PETN.
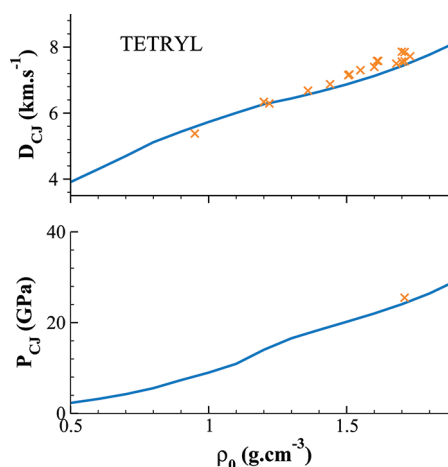

Figure 10. D_{CJ} and P_{CJ} as functions of the initial density for TETRYL.

fluid is not very accurate. A new VdW-6p mixing rule calibrated to reproduce the properties of binary mixtures is proposed. Using both the improved KLRR method and the proposed VdW-6p mixing rule to compute CJ state properties of explosives enables one to approximate the reference Monte Carlo simulation results of the same systems with an error of ~0.25% on average. This is a great improvement when compared to the former version of the CARTE code (MCRSR+VdW-1f) for which an error of ~1.8% was observed.

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Using these improved models, properties of CHON systems at high temperature and pressure can be computed by means of the CARTE code with almost the same accuracy as Monte Carlo simulations but with a much lower computational cost.

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■ ACKNOWLEDGMENT

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