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Improved Reactive Flow Modeling of the LX-17 Double Shock Experiments

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Abstract. Over driven double shock experiments provide a measurement of the properties of the reaction product states of the insensitive high explosive LX-17 (92.5% TATB and 7.5% Kel-F by weight). These experiments used two flyer materials mounted on the end of a projectile to send an initial shock through the LX-17, followed by a second shock of a higher magnitude into the detonation products. In the experiments, the explosive was initially driven by the flyer plate to pressures above the Chapman-Jouguet state. The particle velocity history was recorded by Photonic Doppler Velocimetry (PDV) probes pointing at an aluminum foil coated LiF window. The PDV data shows a sharp initial shock and decay, followed by a rounded second shock.

Here, the experimental results are compared to 2D Cheetah reactive flow modeling. Our default Cheetah reactive flow model fails to accurately reproduce the decay of the first shock or the curvature or strength of the second shock. A new model is proposed in which the carbon condensate produced in the reaction zone is controlled by a kinetic rate. This allows the carbon condensate to be initially out of chemical equilibrium with the product gas. This new model reproduces the initial detonation peak and decay, and matches the curvature of the second shock, however, it still over-predicts the strength of the second shock.

INTRODUCTION

Determining the relevant kinetic rates and the equation of state of high explosive (HE) products is crucial for predicting the performance of HE, including its momentum and energy delivered, over a wide range of pressures. Chemical equilibrium modeling of detonation (see, for example Refs. [1, 2, 3, 4, 5]) depends heavily on having an accurate equation of state of each of the product species being modeled.

It has long been recognized that carbon kinetics effects play an important role in the detonation of CHNO explosives[6, 7, 8, 9, 10] (such as TATB based explosives), particularly those with a high oxygen deficiency, which tend to produce more carbon condensate. These kinetic effects could include production, coagulation, and phase transition of the various forms of carbon in the detonation products. A detailed understanding of these effects is necessary to the proper chemical equilibrium modeling of HE, however almost no details of the carbon evolution are known.

To that end, a series of eight overdriven double shock experiments[11, 12] have been performed on the insensitive high explosive LX-17 (92.5% TATB and 7.5% Kel-F by weight) to probe the effects of reaction kinetics and the equation of state of the explosive products. These experiments build on previous research on reflected or overdriven LX-17[13, 14, 15, 16, 17, 18, 19], as well as a prior set of experiments using reflected shocks with manganin piezoresistive gauges to measure the pressure states inside a detonated explosive[20].

DESCRIPTION OF EXPERIMENTS

Here, we summarize the experimental procedure and show the PDV data. Full descriptions of these experiments are given in Refs. [11, 12]. The double shock experiments used the 2-stage gun at Lawrence Livermore National Laboratory (LLNL) to launch a series of two or more shocks through LX-17. These experiments used two flyer materials (stainless steel backed by tantalum) mounted on the end of the projectile to send an initial shock through the LX-17, followed by a second shock of a higher magnitude into the detonation products. The particle velocity history was recorded by Photonic Doppler Velocimetry (PDV) probes pointing at an aluminum foil coated LiF window. The PDV data shows a sharp initial shock and decay, followed by a rounded second shock. This decay and rounding

TABLE 1. Experimental details for the double shock experiments

Expt.	Date	Velocity (km/s)	Ta Flyer (mm)	SST Flyer (mm)	SST Plate (mm)	LX-17 (mm)
4109	4/18/12	3.511	3.022	2.019	1.508	8.013
4110	4/20/12	3.518	3.018	2.020	1.509	5.995
4111	4/24/12	3.916	3.016	2.018	1.495	5.015
4112	4/26/12	3.737	3.020	2.023	1.496	3.014
4178	8/7/13	2.49	3.019	2.022	1.507	5.013
4179	8/9/13	4.90	3.019	2.023	1.516	5.017
4180	8/13/13	3.35	3.020	2.020	1.517	6.021
4181	8/15/13	4.67	3.020	2.022	1.520	6.002

probably indicates the presence of chemical reactions occurring in the high explosive products. In this paper, we focus on the data taken by the PDV probe aligned with the center of the target.

Table 1 summarizes the experimental details for the double shock experiments. The experimental data from the central PDV probe for four of the experiments, spanning the range of impact velocities explored, are shown in Fig. 1. The data has been shifted in time so that the first shock arrives at $t = 0\,\mu$s. The experiments are arranged from lowest to highest impact velocity.

REACTIVE FLOW MODELING

In this section, we describe past reactive flow modeling of LX-17 using the Cheetah thermochemical code, and propose a new model that controls the production of a carbon condensate by a kinetic rate. This new model improves the modeling of the double shock experiments, although more work is needed in order to fully match the experimental data.

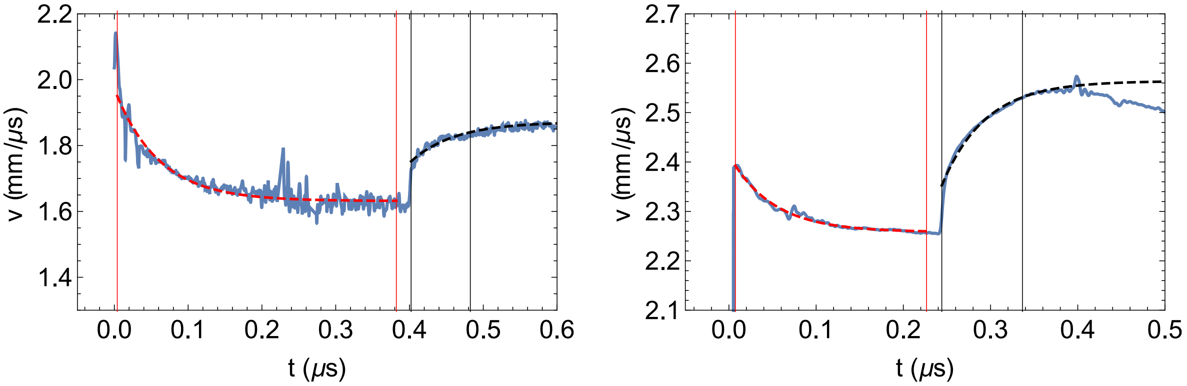
We model the double shock experiments using the Cheetah thermochemical code coupled to the multi-dimensional Arbitrary Lagrangian Eulerian (ALE) code, Ares. Cheetah solves thermodynamic equations to find chemical equilibrium between product species. In addition, Cheetah has the capability to control the rate of reaction of individual species. The current Cheetah models of LX-17 detonation includes a chemical kinetic rate model for the decomposition of the explosive as the detonation waves passes through, and keeps all product species, including the carbon condensate as well as the gas species, in chemical equilibrium. The carbon condensate is taken to be a solid mixture of carbon, nitrogen, and oxygen, roughly based on the nitrogen-rich heterocycle clusters of Manaa *et al.* [21]. All reactions in the product species "freeze-out" below a pressure of around 10 GPa.

We performed 2D and 3D simulations of the experiments using this equilibrium model. We compare the particle velocity at the PDV probes predicted by the simulations to the experimental data in Fig. 2. The results of the simulations using the equilibrium model are shown in red. The experiments are arranged from lowest to highest impact velocity.

Except in the lowest impact velocity experiment (Expt. 4178), the equilibrium model fails to predict the spike and decay as the detonation wave burns the explosive. According to ZND theory (for a summary, see Ref. [22]), this spike would be the von Neumann spike, which is on the un-reacted LX-17 shock hugoniot. The decay in pressure occurs as the explosive decays into its products, and the shock approaches its C-J state (or a higher pressure state for overdriven detonations, as is the case in these experiments). A von Neumann spike is therefore related to the detonation wave progressing down in pressure and density from the un-reacted to the product hugoniot. When the hugoniots cross at higher compression, ZND theory no longer applies and the detonation goes directly to the reacted hugoniot and remains at constant pressure in the reaction zone.

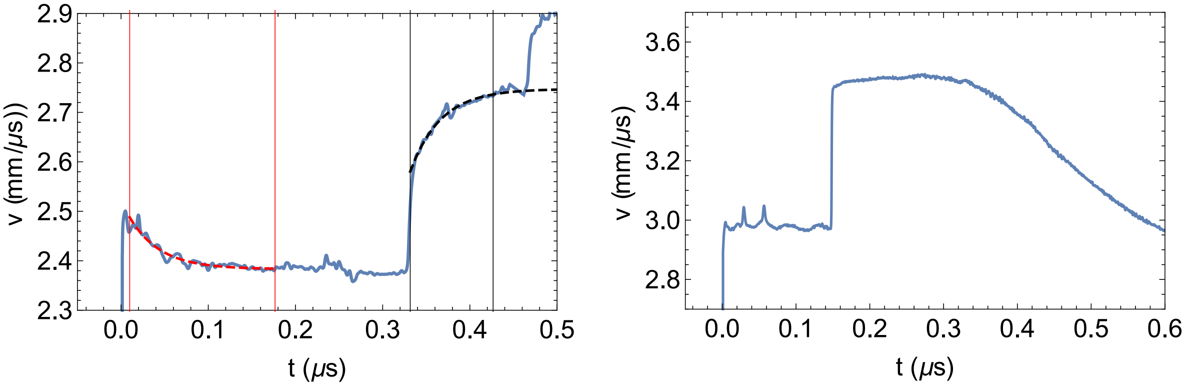
The equilibrium model fails to predict a von Neumann spike for the higher pressure double shock experiments, because the equation of states for the product species used in the Cheetah calculations predict a crossing of the LX-17 and HE products hugoniots at around 40 GPa, in between the pressures reached in Expt. 4178 and the rest of the experiments. Previous simulations of the same experiments in Vandersall *et al.* [11] used an older equation of state library, which does not predict this hugoniot crossing at 40 GPa, and therefore predicts a von Neumann spike for the higher pressure experiments.

In addition to failing to predict a spike and decay after the detonation wave burns the explosive, the equilibrium model fails to predict the rounding of the second shock seen in all experiments except Expts. 4181 and 4179. It also



(a) Expt. 4178, v_{im} = 2.49 km/s

(b) Expt. 4110, v_{im} = 3.518 km/s



(c) Expt. 4112, v_{im} = 3.737 km/s

(d) Expt. 4181, v_{im} = 4.67 km/s

FIGURE 1. The central PDV probe data from four of the double shock experiments performed. The data has been shifted in time so that the first shock arrives at $t = 0\,\mu$s. The experiments are arranged from lowest to highest impact velocity.

has trouble predicting the arrival time and strength of the second shock.

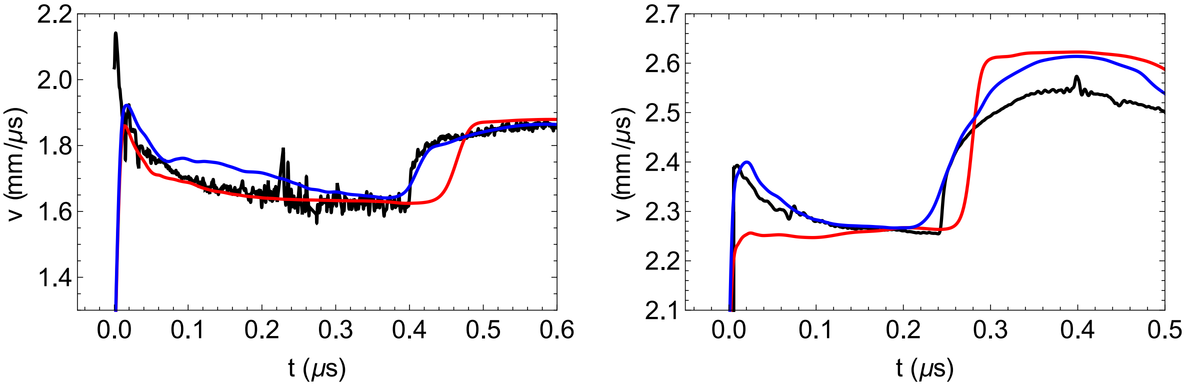
Rate Controlled Carbon Condensate Model

In this section, we propose a new reactive flow model to address some of the shortcomings of the equilibrium model. In this new model, we control the rate of production of carbon condensate. In the past, the coagulation of condensed carbon has been closely studied[7, 23, 24, 25, 26, 27]. In this new model, the size of carbon clusters is assumed to remain constant, but the condensed carbon is taken out of equilibrium with the other HE product species. The reaction rate is a constant for each experiment, i.e.

$$\frac{\mathrm{d}F}{\mathrm{d}t} = \lambda\left(v_{im}\right), \tag{1}$$

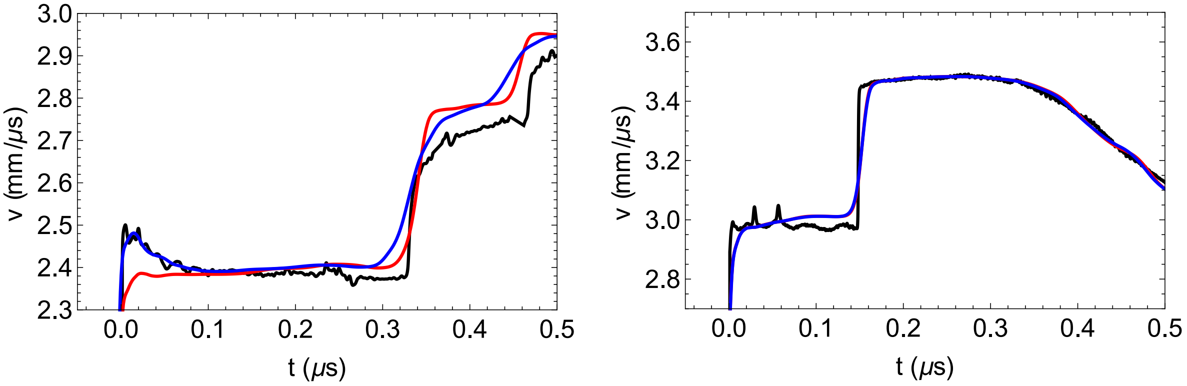
where F is the mass fraction of the reactant, and $\lambda\left(v_{im}\right)$ is a constant, which depends on the impact velocity of the projectile in each experiment. $\lambda\left(v_{im}\right)$ is defined as $\lambda\left(v_{im}\right) \equiv 1/\tau\left(v_{im}\right)$, where $\tau\left(v_{im}\right)$ is defined by fitting a decaying exponential to the first peak of the experimental data from each experiment. The fit is shown in the dashed red lines in Fig. 1. This fit defines the decay time τ for each experiment. Fig. 3 shows τ as a function of flyer velocity. We then fit a line through the data in Fig. 3, excluding Expt. 4178, and use the multiplicative inverse of this line as the rate constant in Eq. 1. The decay time found by fitting the first shock and decay also fits the second, rounded shock as well (see the dashed black lines in Fig. 1). This suggests that the same chemical process is the main cause of both the decay of the initial shock and the rounding of the second shock. We did not perform the fitting procedure on Expts. 4179 and 4180 because in these experiments, the products appear to reach chemical equilibrium faster than can be resolved by the PDV probes.

The results of 2D Cheetah simulations using the new rate controlled carbon condensate model are shown in blue in Fig. 2, along with the experimental data (black lines) and the results of 2D simulations using the equilibrium model



(a) Expt. 4178

(b) Expt. 4110



(c) Expt. 4112

(d) Expt. 4181

FIGURE 2. A comparison of experimental data (black lines) to the results of 2D Cheetah simulations using the equilibrium soot model (red lines) and the new rate-controlled soot model (blue lines). The red and blue lines overlap in Expts. 4181. The experiments are arranged from lowest to highest impact velocity.

(red lines). Before discussing the lower pressure experiments, we first note that the carbon condensation reaction rates are so high for Expt. 4181, that the new model closely matches with the equilibrium model.

The new model predicts an initial spike as the detonation wave passes through the LX-17, as the carbon condensate is created and relaxes to chemical equilibrium with the other product species. This spike from controlling the production of the carbon condensate fits the experimental data better than the equilibrium model in all the experiments, except Expt. 4178, which has pressures below the hugoniot crossing, and where the von Neumann spike from the LX-17 decomposition reaction will interact with the spike from the creation of the carbon condensate. It appears that the interplay between the LX-17 decomposition reaction and the creation of the carbon condensate needs to be accounted for in a better way.

The second shock predicted by the new model more closely matches the experimental data than does the old model. A curvature is introduced to the second shock, and the prediction of the arrival time is improved. The strength of the second shock still does not match well with the data. This is not a surprise, as in the new model, the carbon condensate tends to relax to chemical equilibrium over the characteristic timescale, eventually meeting up with the results of the equilibrium model. This over-prediction of the strength of the second shock is likely not related to kinetic modeling, but rather to the high pressure equation of state of the HE products, and in particular, that of the carbon condensate. Future work should be performed to address this discrepancy.

SUMMARY AND CONCLUSIONS

In this paper we have made a close examination of previous reactive flow modeling of the double shock experiments. The default Cheetah reactive flow model, which keeps the carbon condensate in chemical equilibrium with the other

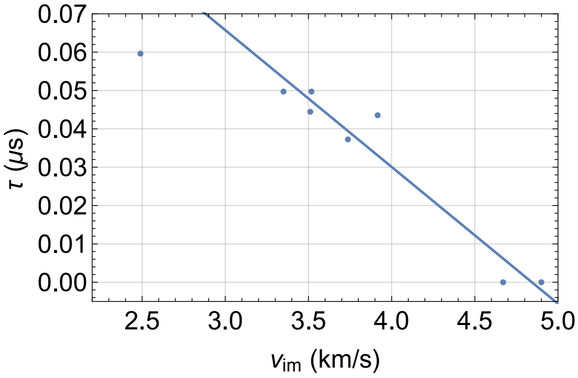


FIGURE 3. The characteristic time scales as a function of flyer impact velocity.

HE products, fails to accurately reproduce the decay of the first shock or the curvature or strength of the second shock. We have proposed a new model in which the carbon condensate produced in the reaction zone is controlled by a kinetic rate. This allows the carbon condensate to be initially out of chemical equilibrium with the product gas. This new model reproduces the initial detonation peak and decay better than the equilibrium model, particularly for the higher pressure experiments, and matches the curvature and arrival time of the second shock well. However, it still over-predicts the strength of the second shock.

There is still work that should be done to improve the agreement of reactive flow modeling with the experimental data. The high pressure equation of state of the HE product species, and in particular, that of the carbon condensate, should be examined to try to better match the measured strength of the second shock in these experiments. In addition, the carbon condensate used for this model could be replaced with other forms of carbon, such as graphite or diamond. It could also be useful to explore the effects of a kinetically controlled transition from graphite to diamond on these experiments. Finally, the model we have proposed in this paper calls for a reaction rate determined by the flyer impact velocity. A rate that is a function of the pressure would be more useful, and likely more consistent with the actual physical processes.

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