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ScienceDirect
Proceedings of the Combustion Institute 32 (2009) 2115–2122
Proceedings of the Combustion Institute
www.elsevier.com/locate/proci

Regression fronts in random sphere packs: Application to composite solid propellant burning rate

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Abstract

The burning rate of a composite solid propellant may be estimated by global modeling, such as the widely used BDP model. The backbone of such models is the "mixture law" that links the propellant burning rate r_p with the burning rate of its own components, i.e., oxidizer r_{ox} and binder r_b. However, different laws are available in literature which all read: $1/r_p = q(\xi)/r_{ox} + (1 - q(\xi))/r_b$, with $q(\xi)$ a function of oxidizer volume fraction ξ. This work attempts in analyzing numerically the validity of those empirical formulations by surface regression computation. Composite propellants are modeled by a random packing of monomodal spheres and the evolution of the regression front is computed via the resolution of Hamilton–Jacobi equations. It is shown that the popular choice $q(\xi) = \xi$ is fairly valid but only provided that burn rate ratio $Z = r_{ox}/r_b$ is about 1. When $Z > 1$, combustion surface is no longer plane and global burning rate deviates from postulated laws. A special regime is also noticed for high rate ratio Z (typically $Z > 5$) because combustion then preferentially takes place through adjacent oxidizer particles. Computed results occur to be correctly modeled by percolation theory. This hints that percolation is a common feature of propellant combustion and a critical percolation threshold on volume fraction is numerically found to be about $\xi_c \sim 0.2$. First validations show encouraging correlations with experimental data.

Keywords: Solid propellant; Burning rate; Percolation

1. Introduction

A reliable prediction of solid propellant combustion is of great importance for industrial applications because solid propellants for propulsion applications must meet serious requirements as far as ballistic properties are concerned (e.g., burning rate, pressure exponent, temperature sensitivity, ...). Therefore, some *a priori* estimation of those properties is a valuable aid for propellant formulation. In spite of the deem complexity of solid propellant combustion, some theoretical and numerical approaches have emerged, from 0D global modeling [1–3] to recent tentative 3D mesoscopic modeling [4]. However, the underlying physics is so intricate that all models need some calibration (e.g., fitting unknown chemical rates for some reactions), so that they might be considered as semi-empirical models. Consequently, they all give a correct estimation of burning rate most of the time. Therefore, global 0D models remain appealing, like the widely used BDP model from Beckstead [1]. They demand virtually no computational resources, are able to deal with composite propellants (albeit in a lumped fashion) and are known to provide correct burning rate predictions for AP (ammonium perchlorate)/HTPB (hydroxyl-terminated polybutadiene) propellants [1,3]

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doi:10.1016/j.proci.2008.06.017

but also propellants based on HMX (cyclotetramethylenetetranitramine) (HMX/HTPB [2] or HMX/active binder [3]), and more recently for advanced CL20 (hexanitrohexaazaisowurtzitane)/GAP (glycidyl azide polymer) propellants [5].

The important backbone of such global modeling lies in the relation linking the global propellant burning rate r_p with the burning rate of its own components, namely oxidizer and binder. The problem may be recast in the following form: consider a composite propellant with oxidizer loaded at volume fraction ξ and binder (at fraction $1-\xi$). Let r_{ox} (resp. r_b) be the burning rate of oxidizer (resp. binder), then the question is what is the burning rate r_p of the composite propellant, assuming that burning rates r_{ox} and r_b are known (experimentally or given by a monopropellant model).

Various approaches have been developed to tackle this problem. The first attempts [1,6] were based on a space-averaging procedure where global rate is calculated by a surface-weighted arithmetic average of binder and oxidizer burning rates but this led to inconsistencies and gave unrealistic results when oxidizer and binder rates differ significantly. Instead, a further improvement was to consider a time-averaging procedure as postulated later on [2,3] and which is now considered from now on. In a randomly packed arrangement of spherical oxidizer particles, let L_{ox} (resp. L_b) be the total length of intersection between a straight line, perpendicular to surface, and oxidizer spheres (resp. binder). We have [3]: $r_p = \sum \text{distances} / \sum \text{times} = (L_{ox}+L_b)/(L_{ox}/r_{ox}+L_b/r_b)$. Some statistical results show that total intersection length L_{ox} is proportional to volume fraction ξ so that the final relation reads [3]:

$$\frac{1}{r_p} = \frac{\xi}{r_{ox}} + \frac{1-\xi}{r_b} \tag{1}$$

This relation is used in many models [3,7,8] and is the one with the strongest theoretical background. It is also found in several other domains such as electrical conductivity or mechanics (“slab layer model”).

However, some works are reported to make use of alternative forms of Eq. (1), such as:

$$\frac{1}{r_p} = \frac{\xi^{1/3}}{r_{ox}} + \frac{1-\xi^{1/3}}{r_b} \tag{2}$$

which is used in the Beckstead model [2] but also in Russia under the “geometrical model” denomination [9,10]. Some variants also consider [10,11]:

$$\frac{1}{r_p} = \frac{(6\xi/\pi)^{1/3}}{r_{ox}} + \frac{1-(6\xi/\pi)^{1/3}}{r_b} \tag{3}$$

Hence, the motivation of present work is to assess whether Eq. (1) is valid or not and why alternative empirical formulations, such as Eq. (2) or Eq. (3), are also reported. In other words, what is the adequate formulation to adopt in a global prediction model?

The proposed approach consists in modeling a composite propellant as a random arrangement of spherical particles (packing) which will be “numerically burnt” by some surface evolution computation. This supposes that both burning rates for oxidizer and binder are prescribed: it is then possible to compute the overall burning rate and hence confirm Eq. (1) or variants. Imposing burning rates for oxidizer and binder suggests they burn on their own, which might be of limited practical interest in some cases (for instance, when diffusion flames are present). However, this is a deliberate choice since the present study does not intend to develop a complex predictive model but rather aims at validating only the mixture laws. This work also intends to draw a bridge between simple global models and advanced 3D models by attempting to incorporate, though in a simple way, geometrical information on propellant microstructure.

2. Packing and front regression

2.1. Packing simulation

As most of solid propellants used in industry are composite, it is necessary to build a numerical model for their microstructure. This model typically consists in a random arrangement of hard spheres in a representative volume element (RVE). This packing allows to model multimodal propellants and particles with any diameter distribution. However, this work will essentially focus on monomodal packs with particles of constant diameter D.

General and refined algorithms for packing may be found in literature [12–14] because it is a problem of importance for a wide variety of physical systems. The algorithm used in present study is close to the Jodrey–Tory algorithm [13]. Basically, sphere centers C_i are randomized and diameters are progressively increased (growing phase). If two spheres i and j overlap, they are both spread apart symmetrically along vector $\mathbf{C}_i\mathbf{C}_j$ to a prescribed distance. The procedure is repeated until no overlap remains. Diameters are then increased again and the whole procedure is resumed until the desired packing fraction and particle diameter are reached.

In the forthcoming computations, the RVE is a cube of fixed size L with wall boundary conditions (spheres are not allowed to extend beyond the boundaries). For all the study, it is chosen the following parameters: $L = 1$ mm and constant particle diameters $D = 100$ μm, which basically leads to roughly 1000 particles at $\xi = 0.5$. This choice results from a trade-off between computational