

Plastic Dissipation Sensitivity to Mechanical Properties in Polycrystalline β -HMX Subjected to Impact Loading

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Abstract

This manuscript investigates the sensitivity of plastic dissipation expressed in the form of temperature rise to anisotropic elasticity constants and crystal plasticity properties of polycrystalline β -HMX (1,3,5,7-tetranitro-1,3,5,7-tetrazocane) under impact loading conditions. Parametric sensitivity analyses are performed using a global sensitivity analysis framework to quantify the relative roles of the elasticity constants of the monoclinic β -HMX crystal, as well as to delineate thermal activation and phonon drag induced slip mechanisms that contribute to the nonlinear response. The plastic behavior of β -HMX is modeled using a Crystal Plasticity Finite Element model incorporating the slip mechanisms of thermal activation and phonon drag driven by the evolution of dislocation generation and annihilation. The results of the sensitivity analyses show that the anisotropic elasticity coefficients of the monoclinic crystal have a nominal effect on the energy dissipation and temperature rise, dominated by sensitivities of a few coefficients. Among the two primary slip mechanisms, phonon drag appears dominant within the load rate and amplitude regimes considered in this study.

Keywords: Energetic materials, Crystal plasticity, Global Sensitivity Analysis, Dynamic behavior, Polycrystalline materials

1 Introduction

Dynamic behavior of energetic materials (e.g., HMX, TATB, PETN) subjected to high amplitude transient loads is complicated due to high crystal anisotropy, interacting thermo-plastic

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processes within the polycrystalline microstructure, and the interactions between different phases acting at multiple length scales [29]. A significant body of work exists on numerical modeling and experimental characterization of various mechanisms within energetic particles (e.g., pore collapse, inter-granular friction, particle fracture, dislocation pile-ups [10, 17, 47, 66, 15]) in order to understand and predict the overall performance of the energetic particles under dynamic loadings. Particularly in mesoscale simulations that involve complex anisotropic polycrystalline particles deforming at non-uniform high rate load regimes, characterization of the relative roles of the competing and interacting deformation and failure mechanisms within the energetic particles is important [6, 14, 78]. This manuscript investigates and quantifies the sensitivities of the elasticity constants and plasticity mechanisms of β -HMX polycrystals subjected to dynamic loading.

HMX (1,3,5,7-tetranitro-1,3,5,7-tetrazocane) is a polymorphic molecular crystal. β phase is of significance since it is the stable phase at room temperature and pressure. The dynamic deformation behavior in β -HMX crystals is crystal orientation dependent [16, 40, 74], and the low symmetry monoclinic structure of β -HMX plays an important role in its thermo-mechanical response. A number of previous efforts focused on measuring the anisotropic elastic constants of β -HMX. Zaug [76] reported, through experiments, five of the thirteen monoclinic elastic constants. Stevens and Eckhardt [62] reported a complete tensor of elastic moduli for crystalline β -HMX by Brillouin scattering. Sun et al. [63] reported elastic constants obtained by the impulsive stimulated thermal scattering (ISTS) method, which are significantly different than those obtained previously using Brillouin scattering. Sewell et al. [57] performed molecular dynamics simulations to compute the thirteen elasticity constants. Bedrov et al. [8] also performed molecular dynamics simulations using a quantum chemistry-based atomistic force field to obtain unit cell parameters (a , b , c , β), coefficients of thermal expansion and heats of sublimation for the β -, α -, and δ -HMX. Pressure-temperature dependency of the elastic constants also influences deformation within crystalline HMX particles particularly when subjected to shock, which induces large variations in pressure and temperature. Piermarini et al. [45] employed a Fourier transform infrared (FTIR) microspectroscopy method to measure the effects of pressure and temperature on the thermal decomposition rate of β -HMX in a diamond anvil high-pressure cell. Sewell et al. [58] calculated the complete elastic tensor at room temperature and pressure via analysis of microscopic strain fluctuations for β -, α -, and δ -HMX. Cui et al. [13] performed isothermal-isobaric molecular dynamics simulations to investigate the variation of the mechanical properties of β -HMX in the temperature range of (5-555) K at 0 GPa and the pressure range of (0-40) GPa at 298 K.

Dynamic behavior of HMX is marked by the presence of complex plastic deformation mechanisms. Menikoff and Kober [38] reported that, for relatively weak waves, plastic deformation is the dominant dissipative mechanism and leads to dispersed waves that spread out in time. Winter and Field [69] noted that the deformation is concentrated in narrow adiabatic shear bands, and investigated the role of localized plastic flow in the impact initiation of explo-

sives. Numerical simulations complement the study of visco-plastic behavior of HMX crystals since experimental measurements are very difficult under high-rate loading given the time and length scales involved. Austin et al. [4] developed a numerical model of HMX, and found that the shear bands are an important mode of localization for HMX in void collapse region as documented in earlier experimental studies [12, 59]. Menikoff and Sewell [39] employed a numerical model of HMX incorporating crystallographic, thermal, mechanical and transport properties to understand the physics associated with the formation and growth of hot spots. Wang et al. [67] investigated the effects of crystal anisotropy and microstructural heterogeneity on mesoscale thermo-mechanical response of PBXs based on a crystal plasticity model. Typically, the plastic deformation stage results in substantial heat generation due to viscous flow [7], adiabatic shear [69], fracture [44] and interfacial friction effects [18, 29].

In view of the complexity of the deformation and failure mechanisms associated with the mesostructure of energetic particles such as HMX, we seek to systematically understand how each mechanism incorporated into a numerical model affects and contributes to the overall behavior of the material. Various sensitivity analysis methods such as the One-at-a-time (OAT) [52], screening method [41], differential analysis/local method [48], scatter plot [64], regression analysis [11], global sensitivity analysis [54], and others [19, 53] have been proposed to understand parametric sensitivities in the general context of particulate composites [51, 56, 72, 73]. For energetic materials, the OAT method has been primarily employed. OAT probes the vicinity of a calibrated parameter set by varying one parameter at a time while other parameters are kept constant [6, 14]. The main drawback of the OAT method is that it cannot account for the interactions between parameters since only one parameter is probed each time [52]. In contrast, global sensitivity analysis (GSA) methods offer the capability to quantify the sensitivity of all parameters along with the interactions between them [54]. Recently, Zhang and Oskay [78] proposed a global sensitivity analysis framework for particulate composites, and investigated the material and morphology parameter sensitivities for polymer bonded explosive, with focus on the particle and binder interface properties.

In this manuscript, we focus on investigating the roles of anisotropic elasticity and plastic deformation mechanisms on the overall behavior of β -HMX single crystal and polycrystalline materials. The dynamic behavior of β -HMX is captured by a polycrystal plasticity model validated at intermediate impact loading conditions, where the material deformation is driven by thermo-visco-plastic effects. Through the sensitivity analysis studies, the monoclinic elastic constants have been analyzed to quantify their contributions to the material response over a large parameter space. Parameters that control the plastic deformation mechanisms are also analyzed through the sensitivity analysis framework. In particular, two slip mechanisms, thermal activation and phonon drag are considered as the primary contributing sources to the overall shear strain rate. The material hardening or softening described by the evolution of slip strength are represented through dislocation density evolution equations that include dislocation generation and annihilation. A novel contribution of this manuscript is the identification

of the most consequential elasticity coefficients, and slip and dislocation evolution mechanisms within the given dynamic loading regime for single crystal and polycrystalline configurations.

The remainder of this manuscript is structured as follows: Section 2 provides the crystal plasticity constitutive relations employed in the forward simulation of the dynamic response of β -HMX at the mesoscale, as well as the detailed flow rule and evolution equations. Section 3 describes the basic formulation of GSA, including the surrogate model employed to accelerate sensitivity computation. Section 4 presents the parameter calibration and model verification for β -HMX single crystal. Numerical examples that utilize the GSA framework to quantify the sensitivity of elastic properties and plastic mechanisms for both single crystal and polycrystalline configurations are discussed in Section 5. Section 6 provides the summary and conclusions.

2 Constitutive Model of β -HMX

The elasto-plastic response of HMX crystals is modeled using the Crystal Plasticity Finite Element (CPFE) approach, considering that crystallographic slip is an important deformation mechanism in the plastic stage under the moderate impact load regime. The constitutive model employed in this study describes the isothermal and large plastic deformation of the HMX polycrystal. The large deformation kinematic formulation is based on the framework of Marin [33]. A brief overview of this framework is provided below.

2.1 CPFE formulation

Consider the body of the polycrystalline solid in its current configuration denoted as $\mathbb{B}$. The initial configuration of the body is denoted as $\mathbb{B}_0$. The motion from $\mathbb{B}_0$ to $\mathbb{B}$ follows the map $\mathbf{x} = \phi(\mathbf{X}, t)$, where $\mathbf{x}$ and $\mathbf{X}$ represent positions in the current and initial configurations, and t is time. The deformation gradient, $\mathbf{F} = \partial \mathbf{x} / \partial \mathbf{X}$, is decomposed using the classical multiplicative split:

$$\mathbf{F} = \mathbf{F}^e \cdot \mathbf{F}^p = \mathbf{V}^e \cdot \mathbf{R}^e \cdot \mathbf{F}^p \quad (1)$$

where $\mathbf{F}^e$ is the elastic part of the deformation gradient, $\mathbf{F}^p$ represents the plastic deformation which is modeled by dislocation evolution and shear stress induced slip in the crystallographic slip systems. Polar decomposition of $\mathbf{F}^e$ generates $\mathbf{R}^e$ and $\mathbf{V}^e$, where $\mathbf{R}^e$ is the orthogonal tensor defining the rotation and reorientation of the grains, $\mathbf{V}^e$ is the left stretch tensor representing the pure elastic stretch of the lattice.

Two intermediate configurations, $\tilde{\mathbb{B}}$ and $\bar{\mathbb{B}}$, are introduced. In what follows, all quantities with tilde and over bar respectively indicate representation in the $\tilde{\mathbb{B}}$ and $\bar{\mathbb{B}}$ configurations. Plastic update is performed in the intermediate and stress free configuration $\tilde{\mathbb{B}}$, which is obtained through unloading the elastic stretch, $(\mathbf{V}^e)^{-1}$, from current configuration $\mathbb{B}$. The use of two configurations provides a better setting for elastic-viscoplastic behavior, in contrast with a single intermediate configuration approach typically employed for rigid-viscoplastic

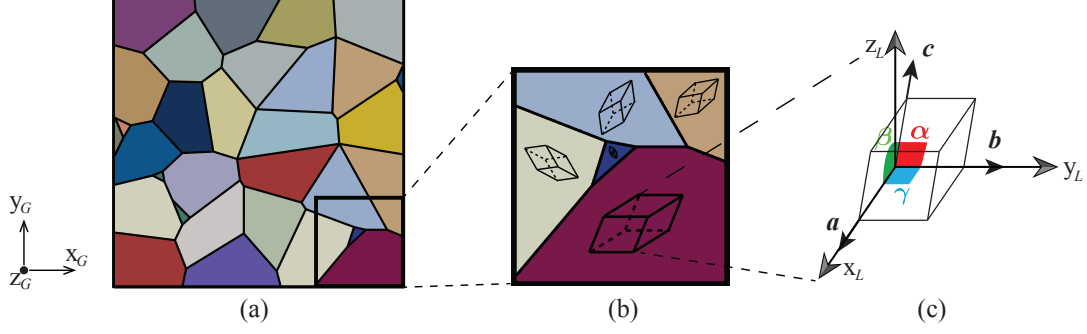


Figure 1: Schematic example of polycrystal β -HMX. x_G , y_G and z_G represent the global coordinates, while x_L , y_L and z_L represent the local coordinates. y_L axis is chosen as the unique axis (parallel to $\mathbf{b}$) in the monoclinic lattice structure with $\alpha = \gamma = 90^\circ$, $\beta \neq 90^\circ$. The transformation between local coordinates and global coordinates is through Euler angles (Kocks convention).

behavior. The above finite deformation crystal plasticity model is specialized to the case of small elastic strains:

$$\mathbf{V}^e = \mathbf{1} + \boldsymbol{\epsilon}^e, \quad \dot{\mathbf{V}}^e \approx \dot{\boldsymbol{\epsilon}}^e, \quad (\mathbf{V}^e)^{-1} \approx \mathbf{1} - \boldsymbol{\epsilon}^e \quad (2)$$

where $\boldsymbol{\epsilon}^e$ is the elastic strain tensor.

The stress-strain relationship is expressed as $\boldsymbol{\tau} = \tilde{\mathbb{C}}^e : \boldsymbol{\epsilon}^e$, where $\boldsymbol{\tau} = (\det \mathbf{F}) \boldsymbol{\sigma}$ is the Kirchhoff stress, $\boldsymbol{\sigma}$ is the Cauchy stress, and $\tilde{\mathbb{C}}^e$ is the elasticity tensor. For the plasticity behavior, the resolved shear stress (RSS) on the α^{th} slip system, τ^α , is defined by the Schmid's law:

$$\tau^\alpha = \tilde{\mathbf{S}} : \text{sym}(\tilde{\mathbf{C}}^e \cdot \tilde{\mathbf{Z}}^\alpha) \approx \boldsymbol{\tau} : \text{sym}(\tilde{\mathbf{Z}}^\alpha) = \boldsymbol{\tau} : \tilde{\mathbf{Z}}^\alpha \quad (3)$$

where $\tilde{\mathbf{S}}$ is the 2^{nd} Piola-Kirchhoff stress, and $\tilde{\mathbf{C}}^e = \mathbf{V}^{eT} \mathbf{V}^e$ is the right Cauchy-Green tensor. $\tilde{\mathbf{Z}}^\alpha = \tilde{\mathbf{s}}^\alpha \otimes \tilde{\mathbf{m}}^\alpha$ is the Schmid tensor in the α^{th} slip system. $\tilde{\mathbf{s}}^\alpha$ and $\tilde{\mathbf{m}}^\alpha$ are the unit vectors along the slip direction and normal to the slip plane, respectively.

2.2 Monoclinic lattice and elasticity

The HMX single crystals are aggregated to form a polycrystalline mesostructure with random orientations. The mesoscale geometry of polycrystal β -HMX is schematically illustrated in Fig. 1a, where each polygonal grain represents a single crystal with a prescribed crystal orientation as shown in Fig. 1b.

β -HMX crystals exhibit low symmetry with a monoclinic structure (Fig. 1c). In this manuscript, y_L axis is chosen as the unique axis (parallel to $\mathbf{b}$ axis), and x_L axis is parallel to $\mathbf{a}$ axis, while z_L axis deviates from the $\mathbf{c}$ axis [46]. For β -HMX crystal, the lattice can be represented using either of two unit cells, $P2_1/n$ and $P2_1/c$, with two molecules per unit cell. In the current manuscript, all coefficients are given in $P2_1/c$ space group. The transformation relationship between the two space groups are provided in Ref. [35].

For crystals of low symmetry, such as the monoclinic lattice, deviatoric and hydrostatic components of the response are coupled. Furthermore, at high rate loading conditions, temperature and pressure dependence of the elastic moduli tensor may be prominent [70]. Under moderate impact loading conditions considered in this study, volume change is smaller compared to the plastic deformation, and pressure/temperature variation is relatively small. Thirteen independent coefficients are necessary to fully describe the anisotropic elastic properties of the monoclinic lattice. The elasticity tensor $\tilde{\mathbb{C}}^e$ for the lattice is expressed in the matrix form as:

$$[\tilde{\mathbb{C}}^e] = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & C_{15} & 0 \\ C_{12} & C_{22} & C_{23} & 0 & C_{25} & 0 \\ C_{13} & C_{23} & C_{33} & 0 & C_{35} & 0 \\ 0 & 0 & 0 & C_{44} & 0 & C_{46} \\ C_{15} & C_{25} & C_{35} & 0 & C_{55} & 0 \\ 0 & 0 & 0 & C_{46} & 0 & C_{66} \end{bmatrix} \quad (4)$$

The elements of the tensor are expressed in the Voigt notation.

For computational convenience, the above elasticity tensor is decomposed to deviatoric and volumetric parts. The deviatoric and volumetric parts of the constitutive law becomes [33]:

$$\begin{aligned} \text{dev}\boldsymbol{\tau} &= \tilde{\mathbb{C}}_d^e : \text{dev}\boldsymbol{\epsilon}^e + \widetilde{\mathbf{H}}^e \text{tr}(\boldsymbol{\epsilon}^e) \\ p_\tau &= (\widetilde{\mathbf{H}}^e)^T : \text{dev}\boldsymbol{\epsilon}^e + \widetilde{M}^e \text{tr}(\boldsymbol{\epsilon}^e) \end{aligned} \quad (5)$$

where $\boldsymbol{\tau} = \text{dev}\boldsymbol{\tau} + p_\tau \tilde{I}$, $\tilde{I}$ is the second order identity, $\tilde{\mathbb{C}}_d^e$ is the deviatoric part of the elasticity tensor, $\widetilde{\mathbf{H}}^e$ is the second order deviatoric-isochoric elastic coupling tensor, and $\widetilde{M}^e$ is the elastic volumetric coefficient. Following Ref. [33], vector representations of the deviatoric stress and strain are expressed using five independent components. The corresponding matrix forms of $\tilde{\mathbb{C}}_d^e$, $\widetilde{\mathbf{H}}^e$ and $\widetilde{M}^e$ derived with respect to the crystal axes of the monoclinic lattice are shown in the appendix.

2.3 Flow rule and evolution laws

The form of the kinematic equation includes both the thermal activation and phonon drag mechanisms of dislocation slip. This is due to the fact that the velocity of a dislocation that moves through an array of obstacles such as the internal crystal boundaries [2] is determined by the time required to bypass the obstacles and the time required to move from one obstacle to another [20, 26, 30]. When the dislocation motion is relatively slow (1e-6 m/s - 1 m/s), the dislocation motion is thermally activated, and local obstacles of various types and the Peierls lattice barrier control glide resistance. At faster speeds (>1 m/s), the dislocation motion is drag-dependent, and only weakly depends on obstacles [26]. Hence, the slip rate in slip system α is expressed as:

$$\dot{\gamma}^\alpha = \left(\frac{1}{\dot{\gamma}_w^\alpha} + \frac{1}{\dot{\gamma}_r^\alpha} \right)^{-1} \quad (6)$$

where $\dot{\gamma}_w^\alpha$ and $\dot{\gamma}_r^\alpha$ respectively represent the contributions from thermal activation and phonon drag.

The thermally activated slip evolution is expressed as:

$$\dot{\gamma}_w^\alpha = \begin{cases} \left(\frac{\dot{\gamma}_{wo}^\alpha}{\sqrt{\rho_n}} \right) \left[\exp \left(- \frac{\Delta G^\alpha(\tau^\alpha)}{\kappa \theta} \right) - \exp \left(- \frac{\Delta G^\alpha(-\tau^\alpha)}{\kappa \theta} \right) \right] + h(\tau^\alpha), & \text{if } |\tau^\alpha| \leq g^\alpha \\ \left(\frac{\dot{\gamma}_{wo}^\alpha}{\sqrt{\rho_n}} \right) \text{sign}(\tau^\alpha) \left[1 - \exp \left(- \frac{2c_G \mu^\alpha}{\kappa \theta} \right) \right] + h(\tau^\alpha), & \text{if } |\tau^\alpha| > g^\alpha \end{cases} \quad (7)$$

where $\dot{\gamma}_{wo}^\alpha$ is the reference shear strain rate, κ is a constant and θ is temperature. ρ_n is a dimensionless dislocation density measure normalized by the reference dislocation density ρ_{ref} ($\rho_n = \rho/\rho_{\text{ref}}$). $\Delta G^\alpha(\tau^\alpha)$ is given by:

$$\Delta G^\alpha(\tau^\alpha) = c_G \mu^\alpha \left[1 - \left(\frac{\tau^\alpha}{g^\alpha} \right)^p \right]^q \quad (8)$$

where c_G, p, q are constants, μ^α is the shear modulus resolved in the α^{th} slip system [22]:

$$\mu^\alpha = \mathbf{Z}_0^\alpha : \mathbb{C}^e : \mathbf{Z}_0^\alpha; \quad \mathbf{Z}_0^\alpha = \mathbf{s}_0^\alpha \otimes \mathbf{m}_0^\alpha \quad (9)$$

g^α is the slip strength of the slip system α , and takes the form:

$$g^\alpha = r^\alpha (g_o + s\sqrt{\rho}) \quad (10)$$

where, s and g_o are model parameters, and r^α is the ratio of the slip system strength g^α and the reference slip system strength $g^{(010)[100]}$ ($r^{(010)[100]} = 1$).

The transition from thermal activation to phonon drag is controlled by the penalty function:

$$h(\tau^\alpha) = \text{sign}(\tau^\alpha) \left(\frac{\tau^\alpha}{g^\alpha} \right)^\zeta \quad (11)$$

where ζ is a parameter set to be a large number. The penalty function $h(\tau^\alpha)$ ensures that the phonon drag component of slip is predominant in the presence of overstress (i.e. $\tau^\alpha > g^\alpha$), and it smoothly transitions between the two mechanisms of slip. Figure 2 illustrates the role of h on the shifting between the dominant slip mechanism as a function of RSS. At low amplitude of RSS compared with the slip system strength, thermal activation is the dominant slip mechanism, whereas at higher amplitudes of RSS, phonon drag is dominant. In Fig. 2, the dislocation density and temperature are taken to be constant ($\rho=0.0307$ micro-m⁻² and $\theta=297$ K). The other parameters are set to values indicated in the verification studies below. The smoothness of the transition from thermal activation-dominated to phonon drag-dominated slip is governed by the power constant ζ . For relatively large values of ζ , the behavior is very similar to the unregularized behavior, whereas an appropriate choice for ζ provides a smoother transition, allowing better stabilization in the numerical implementation of the model.

The drag operates as the dislocation bows between obstacles before it cuts or bypasses

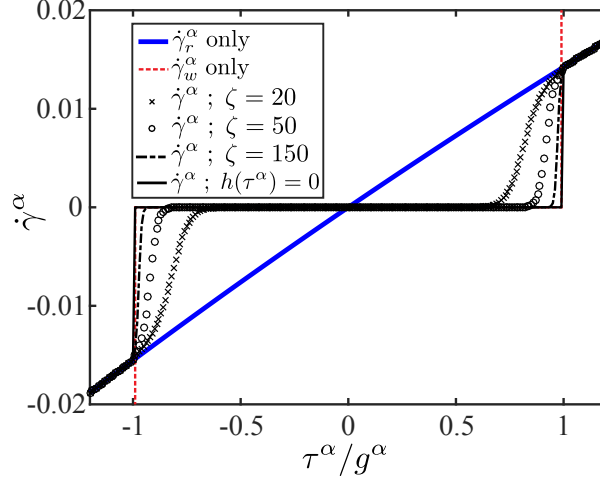


Figure 2: Transition between the dominant slip mechanisms using the power-law function $h(\tau^\alpha)$. Dislocation density is of 0.0307 micro-m⁻² in the illustration. The lines with “x x x”, “o o o”, and “---” represent the total shear strain rate with ζ equal to 20, 50 and 150, respectively.

them. For the glide of dislocations between sets of obstacles, the slip rate is:

$$\dot{\gamma}_r^\alpha = \dot{\gamma}_{ro} \rho_n \left[1 - \exp \left(- \frac{\tau^\alpha}{D_r} \right) \right] \quad \text{where} \quad D_r = D_{ro} \frac{\theta}{\theta_0} \quad (12)$$

where $\dot{\gamma}_{ro}$ is the reference shear strain rate, D_{ro} is the reference drag stress, and θ_0 is the reference temperature.

The evolution of slip strength is controlled by the dislocation density evolution which, under dynamic loading, evolves through generation and annihilation mechanisms:

$$\frac{d\rho}{d\gamma} = \frac{d\rho^+}{d\gamma} + \frac{d\rho^-}{d\gamma} \quad (13)$$

The dislocation generation term [36, 42] is expressed as $d\rho^+/d\gamma = n_1 \sqrt{\rho}$. The dynamic recovery term [36] is described by the evolution model $d\rho^-/d\gamma = -n_2 \dot{\gamma}^{-\frac{1}{n_3}} \rho$, where n_1 , n_2 and n_3 are material parameters, and the net slip system shearing rate is $\dot{\gamma} = \sum_{\alpha=1}^N |\dot{\gamma}^\alpha|$.

Under the dynamic loading conditions considered in this study, the dissipation induced by the viscoplastic slip results in localized heating through the adiabatic assumption. The adiabatic temperature evolution is expressed as:

$$c_V \dot{\theta} = \eta \sum_{\alpha=1}^N \tau^\alpha \dot{\gamma}^\alpha \quad (14)$$

where c_V is the specific heat, and η is the Taylor-Quinney parameter.

It is important to note that earlier works have attributed mechanical deformation in HMX primarily to twinning [2, 31] facilitated by the relatively large size of the molecular crystal

(compared to smaller crystals, where twinning is not as prevalent [28]). More recently, through microhardness indentation experiments, Gallagher et al. [21] observed that slip also plays an important role in the deformation of HMX particles. From the standpoint of crystal plasticity modeling, relatively scarce information is available to fully describe mechanical deformation of HMX particles using twinning alone. Of note is the work by [75], who modeled twinning on the (101)[10 $\bar{1}$] as an additional slip system idealized using the same flow and hardening evolution laws as for slip. To the best of the authors' knowledge, the other crystal plasticity modeling studies (e.g., [3, 4, 24, 77, 5, 25, 67, 68, 70, 71]) focused on describing high rate deformation in HMX using slip alone. In view of the potential importance of twinning in describing the mechanical response of HMX, additional development of experimentally-validated CPFEE models that include deformation twinning is needed and will be subject of future studies.

3 Global Sensitivity Analysis

Global Sensitivity Analysis (GSA) measures the sensitivity of parameters induced in the description of the behavior over a specific parameter subspace. The primary idea of GSA is to apportion the uncertainty or variance in the simulation output into different sources of uncertainty or variance of model inputs (i.e., model parameters). The contributions from different input parameters are quantified by sensitivity indices (Sobol's indices are used in this study) in the sense of self-contribution and interactions with the other input parameters.

3.1 Variance decomposition

Consider a target function $y = f(\mathbf{u})$, which in the current case an output of the simulations, is decomposed as [61]:

$$f(\mathbf{u}) = f(u^1, \dots, u^d) = f_0 + \left[\sum_{i=1}^d \phi_i(u^i) + \sum_{i_1=1}^{d-1} \sum_{i_2=i_1+1}^d \phi_{i_1, i_2}(u^{i_1}, u^{i_2}) + \dots + \phi_{1, 2, \dots, d}(u^1, \dots, u^d) \right] \quad (15)$$

where $\mathbf{u}$ is the vector of selected input parameters:

$$\left\{ \begin{aligned} f_0 &= \int f(\mathbf{u}) \prod_{i=1}^d [p_i(u^i) du^i] = E(y) \end{aligned} \right. \quad (16)$$

$$\left\{ \begin{aligned} \phi_i(u^i) &= \int f(\mathbf{u}) \prod_{j \neq i} [p_j(u^j) du^j] - f_0 = E_{u^{-i}}(y|u^i) - f_0 \end{aligned} \right. \quad (17)$$

$$\left\{ \begin{aligned} \phi_{i_1, i_2}(u^{i_1}, u^{i_2}) &= \int f(\mathbf{u}) \prod_{j \neq i_1, i_2} [p_j(u^j) du^j] - \phi_{i_1}(u^{i_1}) - \phi_{i_2}(u^{i_2}) - f_0 \end{aligned} \right. \quad (18)$$

with $p_i(u^i)$ is the Probability Distribution Function (PDF) of the i^{th} input parameter u^i . The constant f_0 is the expectation of the target function. The notation u^{-i} represents all possible u^j with $j \neq i$. $\int f(\mathbf{u}) \prod_{j \neq i} [p_j(u^j) du^j]$ is the expectation of y given u^i , i.e., $E_{u^{-i}}(y|u^i)$ by

definition. $\phi_i(u^i)$ represents the contribution of u_i to y only from itself, while $\phi_{i_1, i_2}(u^{i_1}, u^{i_2})$ represents the contribution from the interaction between u^{i_1} and u^{i_2} . The variance of y is obtained:

$$\text{Var}(y) = \int [f(u^1, \dots, u^d) - f_0]^2 \prod_{i=1}^d [p_i(u^i) du^i] \quad (19)$$

The parameters are taken to be independent random variables, which ensures the uniqueness of the decompositions stated above [61]. While it is possible to account for correlations (see, for instance, Li et al. [32], who introduced a new unified global sensitivity analysis framework and defined three sensitivity indices to fully describe the parameter sensitivity for systems with correlated inputs), the parameter correlation is not considered in this manuscript.

3.2 Sensitivity indices

Sensitivity index is a quantitative measure of parameter sensitivity/importance to the model output variance. The First Order Index (FOI) quantifies the contribution to output variance from the parameter itself, while Total Effect Index (TEI) also includes the interactions between the parameter and the other parameters. Substituting Eq. 19 into Eq. 15:

$$1 = \sum_{i=1}^d S_i + \sum_{i_1=1}^{d-1} \sum_{i_2=i_1+1}^d S_{i_1 i_2} + \dots + S_{1,2,\dots,d} \quad (20)$$

where the first order sensitivity index S_i is defined as:

$$S_i = \frac{\text{Var}(\phi_i(u^i))}{\text{Var}(y)} = \frac{\text{Var}_{u^{-i}}(E_{u^{-i}}(y|u^i))}{\text{Var}(y)} \quad (21)$$

$\text{Var}_{u^{-i}}(E_{u^{-i}}(y|u^i))$ is the variance of the expectation of y for given u^i .

The total effect index, S_i^T , is expressed as:

$$S_i^T = 1 - \frac{\text{Var}_{u^{-i}}(E_{u^{-i}}(y|u^{-i}))}{\text{Var}(y)} \quad (22)$$

where $E_{u^{-i}}(y|u^{-i})$ is the expectation of y for given $(u^1, \dots, u^{i-1}, u^{i+1}, \dots, u^d)$. The total effect index S_i^T is the sum of the first order index and the corresponding higher order terms [27]:

$$S_i^T = S_i + \sum_j S_{ij} + \sum_{j_1=1}^{d-1} \sum_{j_2=j_1+1}^d S_{ij_1 j_2} + \dots + S_{1,2,\dots,d} \quad (23)$$

where S_{ij} represents the interaction between u^i and u^j , $S_{ij_1 j_2}$ represents the interaction between u^i , u^{j_1} and u^{j_2} , and $S_{1,2,\dots,d}$ is the interactions between all input parameters. The interaction terms in the TEI is non-zero despite the fact that the parameter distributions are taken to be uncorrelated [32].

In the view of multiple potential failure mechanisms, GSA provides the ability to under-

stand the dominant mechanisms behind the complex input-output relationships over the input space, and eliminates the trivial influence from insensitive parameters.

3.3 Gaussian process model

The evaluation of FOI and TEI requires a substantial number of forward simulations. Unfortunately, mesoscale simulations of energetic materials using CPFEM are computationally expensive. In order to achieve computational efficiency needed to compute the sensitivity indices, we employed Gaussian Process (GP) modeling [50] as surrogate to mesoscale simulation of β -HMX crystals under dynamic loading.

A Gaussian Process model is a variant of a radial basis function that is built on Gaussian kernels. The GP model is trained to approximate the response surface of interest using an inexpensive function approximation, which must otherwise be generated by a large set of mesoscale simulations. The approximation function is expressed as a Gaussian distribution conditioned upon a set of training points that are generated using a relatively small set of mesoscale simulations. In the current study, GP models with squared exponential correlation function are employed, and the trend of the models is taken to be up to second order polynomial. It is possible to achieve higher order polynomial trends by increasing the number of training samples. The process variance and character parameter are determined through the maximum likelihood estimation.

The stratified sampling method is employed to sample the parameter subspaces to ensure that (1) information from the full domain is captured and (2) the total number of forward simulations, as the training data set for GP model, is as small as possible. The primary idea of stratified sampling method is to partition the parameter domain to multiple non-overlapping subspaces and to draw at least one sample point in each subspace [43].

The numerical computation of sensitivity indices is performed with the GP model as surrogate simulator through the Monte-Carlo based procedure [54]. The general steps of the computation procedure of S_i (S_i^T) is summarized as:

1. Generate one sample u^i (u^{-i}) from given distribution
2. Generate m_1 samples of u^{-i} (u^i)
3. Use GP model to calculate m_1 samples of y
4. Go back to *Step 1* for m_2 times
5. Calculate corresponding variance and expectation
6. Calculate sensitivity index S_i (S_i^T)

4 Model Verification

To ensure that the forward model for the energetic material captures the dynamic response with sufficient accuracy, we calibrated the model parameters to values available in the literature and compared the model predictions with those provided in Ref. [5, 36, 37], as well as the experimental results by Dick et al. [16]. The current constitutive model of β -HMX has been

verified to assess the effects of (1) specializing the formulation to small elastic strains, and (2) the use of the additional power-law function $h(\tau^\alpha)$. The verified model predictions serve as the starting point for the sensitivity analyses.

4.1 Parameter calibration

The thirteen constants of the anisotropic tensor of elastic moduli of the monoclinic lattice are set to those calculated by Sewell et al. [58] through MD simulations. The density and specific heat of β -HMX crystals are set to 1.9 g/cm³ and 1 kJ/(kg·K), respectively. The lattice structure of β -HMX is described by four cell parameters: a , b , c and β . These parameters are typically pressure dependent [13]. In view of moderate load amplitudes, the lattice parameter are taken as constants ($a=6.54$ Å, $b=11.05$ Å, $c=8.70$ Å, and $\beta=124.3^\circ$).

The CPFE model incorporates ten slip systems [4] for the monoclinic lattice structure of β -HMX in the $P2_1/c$ space group as summarized in Table 1. Slip systems and the corresponding strength ratios (r^α) are summarized in Table 1.

Table 1: Slip strength ratios.

Slip system	(010)[100]	(011)[100]	(01 $\bar{1}$)[$\bar{1}$ 00]	($\bar{1}$ 02)[010]	(001)[100]
Ratio r^α	1	0.963	0.963	0.933	1.68
Slip system	($\bar{1}$ 02)[201]	(011)[0 $\bar{1}$ 1]	(0 $\bar{1}$ 1)[0 $\bar{1}$ 1]	($\bar{1}$ $\bar{1}$ 1)[$\bar{1}$ 0 $\bar{1}$]	(1 $\bar{1}$ $\bar{1}$)[101]
Ratio r^α	0.376	0.931	0.931	0.701	0.701

Table 2: Flow rule and hardening rule parameters.

Parameter	$\dot{\gamma}_{wo}$	c_G	g_0	s	p	q	$\dot{\gamma}_{ro}$	D_{ro}
Value	1	11.47	103	0.1666	1	1	2.5	1160
Unit	micro-s ⁻¹	K/MPa	MPa	MPa·micro-m			micro-s ⁻¹	MPa
Parameter	n_1	n_2	n_3	c_V	η	θ_0	ρ_0	ρ_{ref}
Value	37.99	62	4.98	1	0.9	293	0.0307	0.452
Unit	micro-m ⁻¹			kJ/(kg·K)		K	micro-m ⁻²	micro-m ⁻²

Table 2 presents the values of the parameters associated with slip and CRSS evolution in the crystal plasticity model. The values of the model parameters are set to those proposed in Ref. [5]. The parameter n_2 for dynamic recovery is not provided in the aforementioned reference. Experimental data for HMX necessary to calibrate n_2 is not available. We estimated the value of n_2 by comparing the hardening rate of PBX 9501, whose main component (95% wt) is β -HMX. For PBX 9501 that was compressed at the room temperature (25°C) at a constant strain rate of 2000 s⁻¹ [23], the hardening rate ($\partial\sigma/\partial\gamma$) as a function of stress at a constant slip rate 2000 s⁻¹ is compared with the model predictions using $n_2=62$ in Fig. 3.

Figure 4 shows temperature dependence of the constitutive behavior of the calibrated CPFE model compared with PBX 9501 and a BCC crystal, which exhibit qualitatively similar temperature dependence of yielding. As observed in Fig. 4b, the stress-strain curves for Vanadium

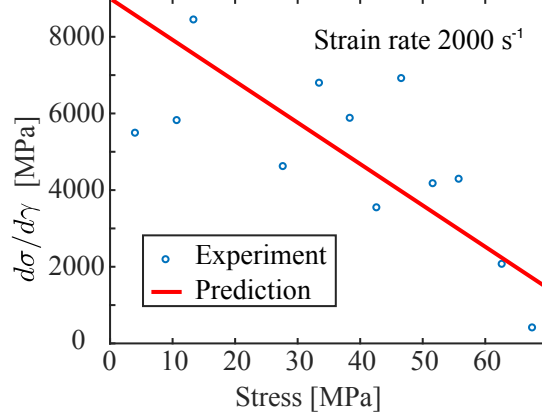


Figure 3: Hardening rate-stress curve at constant strain rate 2000 s^{-1} . The hardening rate is estimated by the finite difference method, where the stress and strain data are extracted from experimental measurements by Gray et al. [23] at room temperature.

at different temperatures reported in Ref. [65] show that higher temperatures reduce the yield strength of the material. A similar trend, in Fig. 4c, has been observed for PBX 9501 as measured by Gray et al. [23]. The constitutive law employed in the current study also exhibits a reduction in the yield strength with temperature as shown in Fig. 4a.

4.2 Verification study

The capabilities of the crystal plasticity model as well as its implementation have been verified by comparing its predictions to a similar model and experimental observations described in Ref. [16]. The assessment of the current model was performed in the context of the dynamic behavior of single crystal β -HMX specimens subjected to gas gun shots.

The setup of this verification study is shown in Fig. 5. An initial velocity is applied on the impactor (2024 Al). The dynamic wave passes to HMX specimen through the anvil (X-cut quartz anvil for impact velocity $\sim 300 \text{ m/s}$, and Kel-F¹ anvil for impact velocity $> 500 \text{ m/s}$). PMMA was chosen as the window material due to the fact that its shock impedance is close to but slightly lower than that of HMX. The slight impedance mismatch results in a rarefaction wave back to the HMX specimen after the incident wave hits the material interface. The specimen and load configurations of the six experiments are listed in Table 3 where sample type represents the loading direction in $P2_1/n$ space group. The thicknesses of the HMX specimens varies from 1.23 mm to 4.66 mm. Initial velocities of the impactor are approximately 300 m/s except for “Shot 1182” which was approximately 520 m/s.

Menikoff et al. [40] used linear Hugoniot with Mie-Gruneisen Equation Of State (EOS) for the impactor and anvils, and the shock state in the anvil is used to set the initial conditions that drive the simulations. In the current study, explicit dynamic simulations have been performed without detailed shock analysis. The loading is applied as prescribed constant velocity on

¹(Kel-F is the 3M Company brand name for polytrifluorochlorethylene)

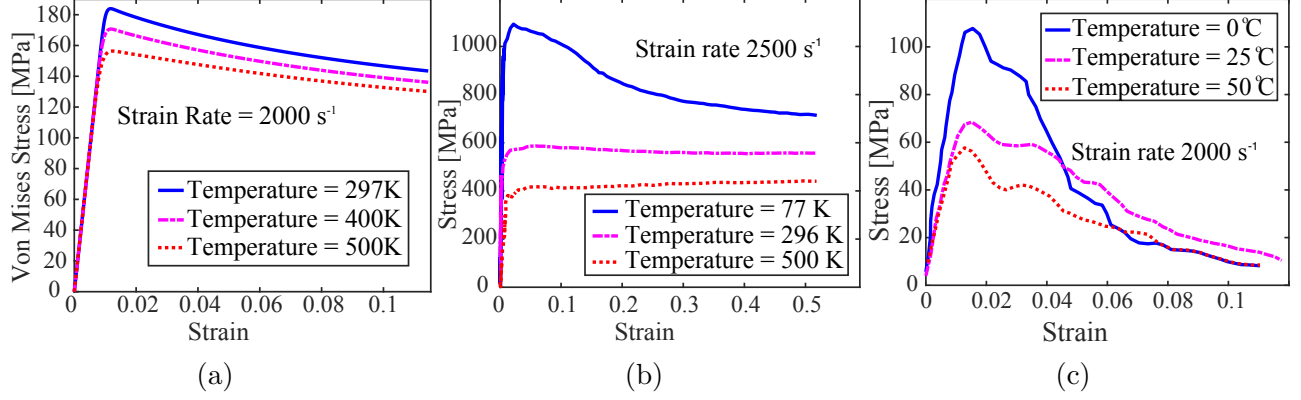


Figure 4: Stress-strain curves at different temperatures for (a) HMX in (011) direction at the constant strain rate of 2000 s⁻¹ predicted by the calibrated CPFE model, (b) experimentally observed behavior of Vanadium (experimental data from [65]) at the strain rate of 2500 s⁻¹ and (c) experimentally observed behavior of PBX 9501 (experimental data from [23]) at the strain rate of 2000 s⁻¹.

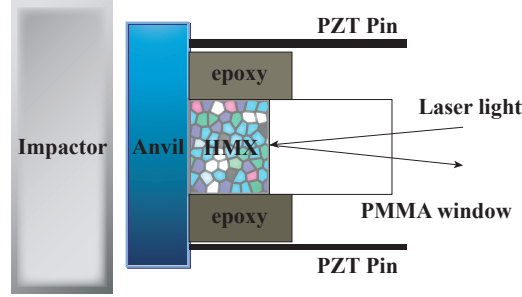


Figure 5: Schematic representation of experimental setting.

the left edge of the specimen. The amplitude of the applied velocity is determined using the measured impactor velocity through the impedance match technique, in which linear Hugoniot relationships are employed for the impactor, anvils and HMX [40, 9]. The PMMA window near the right edge of the specimen is modeled using the nonlinear viscoelastic constitutive law developed by Schuler and Nunziato [55].

In the current verification study, a quasi-one-dimensional model is employed. Dislocation density field is taken to be uniform prior to the onset of the dynamic load. The FE model contains a line of 3D hexahedral element with a total length 8.6 mm. This length ensures that there is no wave reflection during the simulation duration (1.6 micro-s). The viscoelastic PMMA window is bonded to the HMX specimen. Periodic boundary conditions are applied at the top/bottom and front/back boundaries. Barton et al. [5] observed a discrepancy between the times of arrival in the predictions and experiments due to the dependence of elastic parameters to pressure and temperature and reported the prediction results with a time shift. In the current simulations a time shift of 0.08 micro-s is used.

Verification data are collected from three shots along $(110)_{P21/n}$ direction and three shots

Table 3: Specimen and load configurations.

Shot no.	Sample type	Thickness (mm)	Impactor Velocity (m/s)
1180	110	1.23	318.5
1166	110	3.18	306.8
1182	110	3.57	520.9
1181	011	1.39	316.0
1068	011	3.00	314.0
1168	011	4.66	313.2

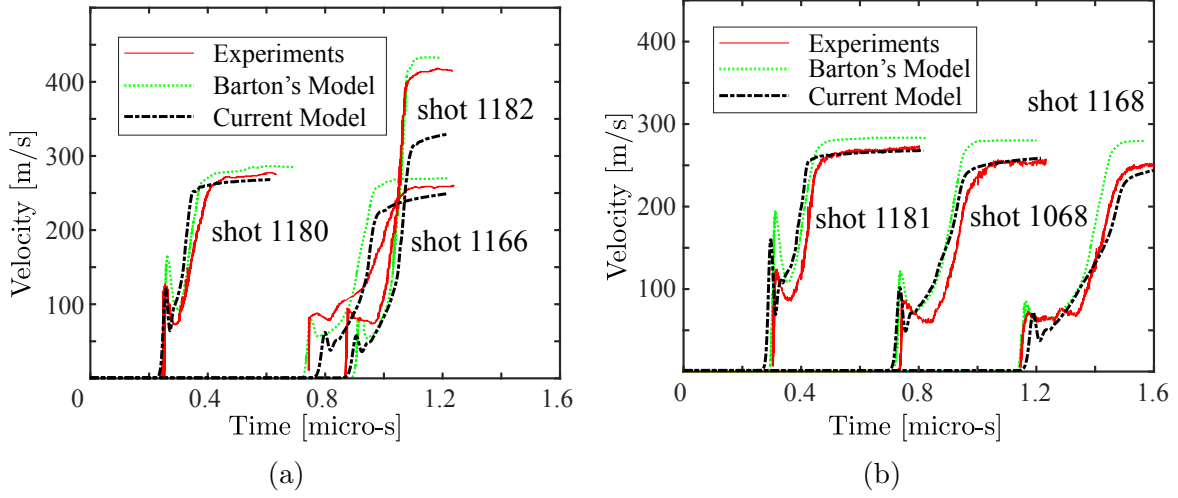


Figure 6: Model verification with respect to loading in (a) the $(110)_{P21/n}$ direction; and (b) the $(011)_{P21/n}$ direction.

along $(011)_{P21/n}$ direction. The predictions of the current model, predictions in Ref. [5] and experimental measurements are compared in Fig. 6. Overall, a reasonable agreement between the predictions and experimental data particularly for the impactor velocity of 300 m/s is observed. This indicates that the proposed model adequately captures viscoplastic response mechanisms. The experimentally observed peaks of the plastic waves (particle velocity at the HMX-PMMA interface) are captured by the current model in all shots ($< 5\%$ error) with the impactor velocity around 300 m/s. A larger discrepancy has been observed in shot 1182. A possible reason for this discrepancy is that the higher initial velocity of the impactor (520.9 m/s) in this shot results in the formation of a stronger shock. Capturing the propagation of the stronger shock requires incorporation of the Equations-of-State (EOS) for the materials into their constitutive models.

5 Sensitivity Analyses

Employing the verification study and the associated model parameters as baseline, we performed sensitivity analyses to understand the relative roles of the material parameters and

viscoplastic mechanisms on the response of β -HMX subjected to impact loading. The analyses are performed in the context of impact response of a single crystal, as well as polycrystalline mesostructures.

5.1 Parameters subspace

The sensitivity analyses have been performed to understand the behavior of a range of parameters that define the elastic and plastic behavior of the energetic crystals. In the subsequent analyses, we restrict the parameter subspaces to a range centered around the set of parameters calibrated for the numerical verification study. The parameter ranges are selected such that the parameter subspace reasonably spans the experimental observations and computationally feasible, i.e., surrogate models can be constructed with a reasonable number of training simulations.

5.1.1 Elastic coefficients ranges

Ranges of the elasticity coefficients are estimated through the available experimental measurements and molecular simulations. Zaug [76] determined thirteen elastic constants via Impulsive Stimulated Light Scattering (ISLS) after the ultrasonic sound speed measurements in single crystal β -HMX at various temperatures. Stevens and Eckhardt [62] measured the complete elasticity tensor for crystalline β -HMX by scattering from a variety of acoustic phonons. Sun et al. [63] used the Impulsive Stimulated Thermal Scattering (ISTS) method to resolve the thirteen elastic constants from acoustic velocity measurements using samples cut parallel to three different crystal planes. Sewell et al. [57] employed two primary simulation cells with 48 and 96 molecular respectively to calculate the thirteen elasticity constants. Sewell et al. [58] also used atomistic simulations to calculate the isothermal elastic properties of HMX crystal. Cui et al. [13] investigated the mechanical properties (elastic coefficients, Young’s modulus, shear modulus and others) of β -HMX at different temperature and pressure by performing isothermal-isobaric molecular dynamics simulations.

In Zaug’s experiments, five (C_{11} , C_{33} , C_{55} , C_{15} , C_{35}) of the thirteen coefficients were well determined while others were not. C_{33} , C_{55} and C_{15} have significant discrepancies with the molecular dynamics predictions [58]. Among the remaining coefficients, C_{22} and C_{46} are within a reasonably tight range. Hence, four coefficients (C_{11} , C_{35} , C_{22} , C_{46}) are taken as constants and excluded from sensitivity studies. C_{33} , C_{44} , C_{55} , C_{66} , C_{12} , C_{13} , C_{23} , C_{15} and C_{25} are regarded as variables whose ranges are selected to span the values available in the literature and summarized in Table 4.

5.1.2 Plastic deformation properties

Compared with the elastic constants, identification of appropriate parameter subspaces for plastic behavior is more challenging. This is due to the fact that different models available in the literature employ different internal state variables and evolution forms to describe the slip and strength evolution, and that relatively small number of studies focused on accurate

Table 4: Parameter ranges of elasticity coefficients (unit: GPa).

Parameter	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{23}	C_{15}	C_{25}
Zaug [76]	17.6	2.9	6.6	3.8	4.0	13.0	6.6	0.6	-1.5
Sewell [57]	17.8	9.1	9.2	9.8	5.9	8.4	8.2	-1.1	3.2
Sewell [58]	23.4	9.2	11.1	10.1	9.6	13.2	13.0	-0.1	4.7
Stevens [62]	12.44	4.77	4.77	4.46	6.37	10.5	6.42	-1.1	0.83
Sun [63]	18.24	9.92	7.69	10.67	9.65	9.75	12.93	-0.61	4.89
Cui [13]	13.2	6.41	4.04	4.68	3.75	4.66	5.07	-0.15	-2.71
Upper boundary	24.0	10.0	12.0	11.0	10.0	18.0	13.0	1.0	5.0
Lower boundary	11.0	2.0	4.0	3.0	3.0	4.0	5.0	-5.0	-3.0

characterization of these parameters. The current crystal plasticity model consists of 16 parameters to describe the slip evolution by thermal activation ($\dot{\gamma}_{wo}^\alpha$, c_G , p , q , ζ), slip evolution by phonon drag ($\dot{\gamma}_{ro}^\alpha$, D_{ro} , θ_o), hardening evolution (g_o , s), adiabatic temperature evolution (c_V , η) and dislocation evolution (n_1 , n_2 , n_3 , ρ_0). In what follows, we focus on the investigation of the phonon drag, thermal activation and dislocation evolution mechanisms. ζ controls the smoothness of the transition of dominant slip mechanism. It is employed for convergence control in this study and set to $\zeta = 150$. The reference temperature θ_o is set as constant (i.e., the room temperature). p and q , which influence low-rate hardening behavior, are also taken as constants (set to unity). The remaining 8 parameters ($\dot{\gamma}_{wo}^\alpha$, c_G , $\dot{\gamma}_{ro}^\alpha$, D_{ro} , ρ_0 , n_1 , n_2 , n_3) are considered as variables in the sensitivity analysis studies below. In view of limited information available, we set the range for each parameter to be $[50\%\mu, 150\%\mu]$, where μ is the calibrated values shown in Table 2, with the exception of c_G and ρ_0 . The upper limit of c_G is slightly reduced since high values require very significant time step size reduction for convergence in CPFE simulations. The upper and lower bounds for the parameters are summarized in Table 5.

Table 5: Ranges for the plastic parameters.

Parameter	$\dot{\gamma}_{wo}$	c_G	$\dot{\gamma}_{ro}$	D_{ro}	ρ_0	n_1	n_2	n_3
Unit	micro-s ⁻¹	K/MPa	micro-s ⁻¹	MPa	micro-m ⁻²	micro-m ⁻¹		
Upper bound	1.5	12.5	3.75	1740	0.046	56.99	93	7.47
Lower bound	0.5	5.735	1.25	580	0.0153	18.99	31	2.49

5.2 Single crystal sensitivity analysis

The response of single crystal β -HMX subjected to impact loading described in Section 4 is employed to investigate the parametric sensitivities. In the first case, the anisotropic elastic properties in β -HMX single crystal are investigated to understand the role that crystal elasticity plays in the material dynamic behavior. In the second case, the relative roles of the viscoplastic deformation mechanisms are investigated by considering the experimental setup for shot 1166 shown in Table 3. In both cases, sensitivities are assessed based on the maximum rise in temperature within the specimen.

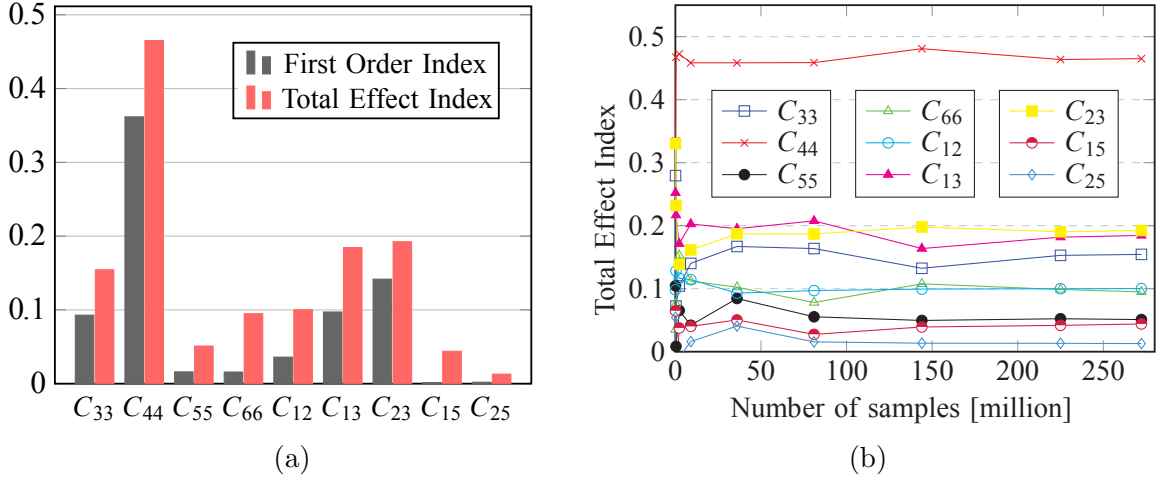


Figure 7: Summary of (a) sensitivity indices and (b) convergence for elastic coefficients in single crystal specimen.

5.2.1 Elasticity coefficients

Approximately 3,000 forward simulations were performed with parameters randomly sampled from parameter ranges shown in Table 4 assuming uniform distribution for each parameter. Approximately 272 million predictions were then generated using the surrogate model to collect information of the response surface over the entire parameter space. In this study, only the elastic parameters are considered as variables, whereas all plastic parameters are set to values in Table 2.

First order and total effect sensitivity indices computed for elastic coefficients are shown in Fig. 7a. The figure indicates C_{44} (or C_{2323} in expended notation) is the most sensitive parameter in terms of the maximum temperature rise. The temperature rise is due to the plastic processes (Eq. 14), and thermoelastic heating is neglected in the analyses. C_{55} , C_{15} and C_{25} have negligible influence on the plasticity and peak temperature under the impact load in $(110)_{P21/n}$ direction. The convergence for each index as a function of number of prediction points is shown in Fig. 7b.

Figure 8 demonstrates the sensitivities of the nine elastic constants (displayed as probability distributions) computed using the One-at-A-Time (OAT) method with the same surrogate model in GSA. In the OAT method, the sensitivity of a single parameter is computed by sampling that parameter from within its range, while keeping all other parameters fixed at the mean. OAT therefore provides “local” variability of the response as a function of that parameter. In contrast with GSA which identifies C_{13} as the third sensitive parameter based on both FOI and TEI, varying C_{13} alone generates the largest temperature variation (≈ 14 K). This discrepancy points to the fact that characterization of the variation in the response surfaces locally (such as captured by the OAT method) provides an incomplete description and assessment of sensitivity. Separately varying the parameters C_{25} , and C_{55} does not produce

significant temperature variation (<3 K), which is consistent with the results shown in Fig. 7a.

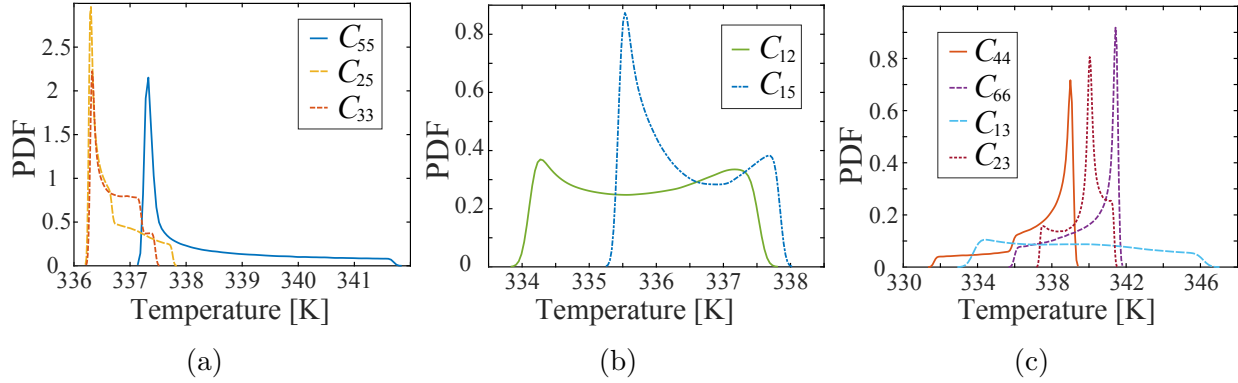


Figure 8: Probability distributions for peak temperature resulting from the OAT analysis of individual elastic constants for single crystal cases.

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Interaction between C_{44} and C_{23} , the two most sensitive parameters identified by GSA, are further investigated by plotting the variation of peak temperature over the C_{44} - C_{23} plane, as shown in Fig. 9a. Within the given parameter range, the temperature distribution with respect to C_{23} is convex for a fixed C_{44} , whereas the variation with C_{44} is linear for a fixed C_{23} value. The fairly uniform variation of the temperature field indicates only a mild interaction between the two most sensitive elastic constants. The peak of this surface occurs near $(C_{44}, C_{23}) = (2.0, 10.0)$ with maximum temperature around 343 K.

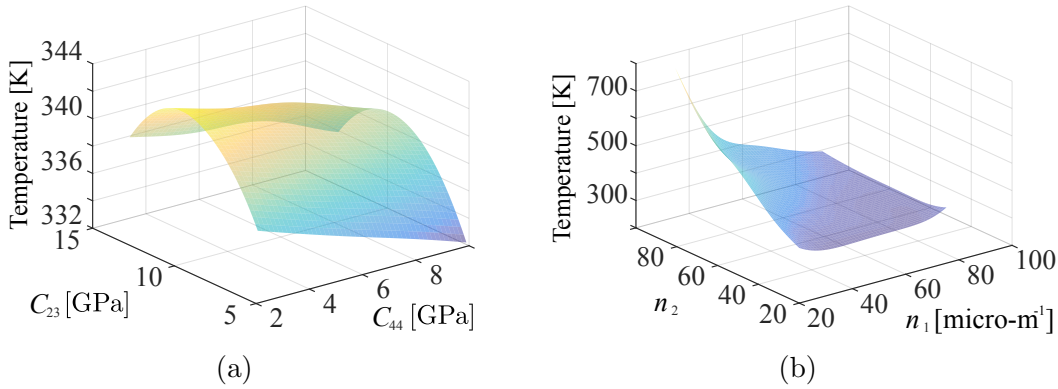


Figure 9: Variation of peak temperature over (a) C_{44} - C_{23} plane and (b) n_1 - n_2 plane for single crystal specimen.

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5.2.2 Plastic deformation mechanisms

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Approximately 3,500 mesoscale simulations were performed as the training set of the GP model that has the target function of maximum temperature rise under the applied impact load. 242 million predictions were then generated by the surrogate to study the variation of the target function and compute the sensitivity indices. In this study, only the plastic parameters are

considered as variables, whereas elastic constants are set to values calculated by Sewell et al. [58].

Figure 10a shows the sensitivity indices for all eight plastic parameters. The convergence of the sensitivity values as a function of the number of prediction points is shown in Fig. 10b.

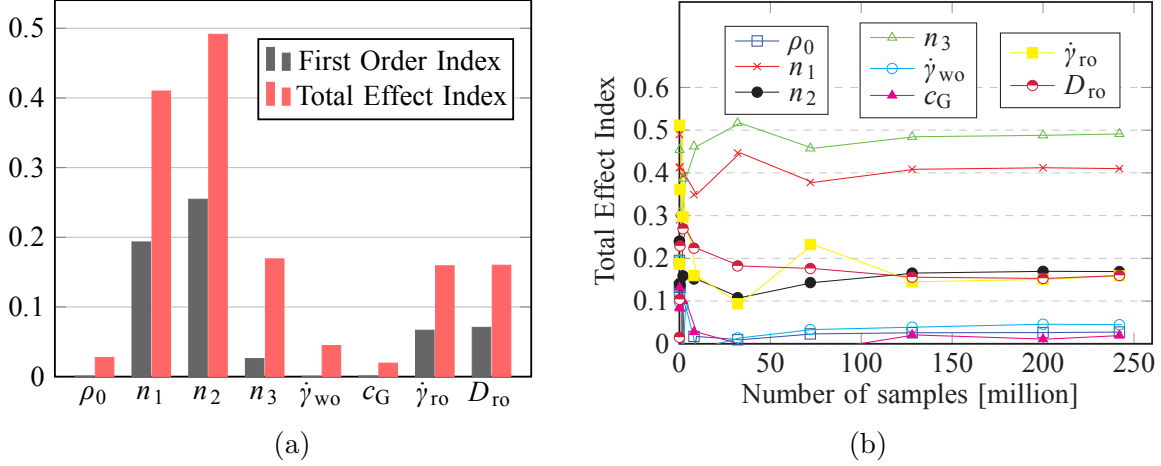


Figure 10: Summary of (a) sensitivity indices and (b) convergence for plastic parameters in single crystal specimen.

The dislocation annihilation coefficient n_2 and the dislocation generation coefficient n_1 are identified as the most influential parameters. The significance of dislocation evolution parameters indicates that the material impurities or defects govern the plastic deformation in terms of temperature rise regardless of which of the slip mechanism is predominant. The parameters, c_G and $\dot{\gamma}_{wo}$ that control the thermal activation mechanism, have little effect, which indicates that under given impact velocity (≈ 300 m/s), thermal activation does not contribute significantly to the plastic deformation compared with the phonon drag mechanism, and this is consistent with Ref. [26]. Furthermore, the initial density of dislocation prior to the onset of the impact loading appears to have insignificant effect on the plastic behavior of the crystal. It is important to note that the void collapse mechanism under shock loading was observed to be sensitive to initial dislocation density in Ref. [5]. Void collapse behavior therefore appears to be substantially different than the shear dominated viscoplastic process probed in this study.

The peak temperature distribution for each parameter obtained by the OAT method is plotted in Fig. 11. Near the calibrated parameter set, varying D_{ro} generates the largest temperature variation (of approximately 200 K). However, D_{ro} is identified as the fourth sensitive parameter using the TEI and the third sensitive parameter using the FOI. The two most sensitive parameters identified by TEI, n_1 and n_2 , generates small temperature variations (≈ 15 K and 70 K) compared with D_{ro} in the OAT analysis. These discrepancies show that the plastic processes are sensitive to the change and interaction of material parameters, which makes the GSA meaningful as it accounts for the parameter variance and relation. Varying

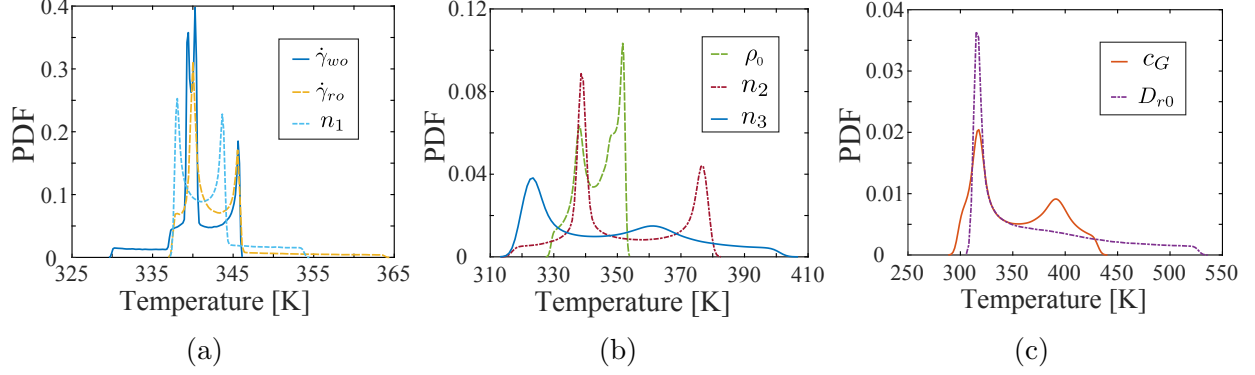


Figure 11: Probability distribution for peak temperature resulting from the OAT analysis of individual plastic parameters for single crystal specimen.

the parameter $\dot{\gamma}_{w0}$ does not produce significant temperature rise (≈ 15 K) consistent with GSA results. As c_G directly amplifies or reduces the activation energy, it also offsets the ratio τ^α/g^α at which the transition from thermal activation mechanism to phonon drag mechanism occurs.

The variation of peak temperature over the n_1 - n_2 plane is plotted in Figure 9b. The maximum temperature occurs near the $(n_1, n_2) = (20, 100)$ corner, which indicates that lower n_1 (i.e., less dislocation generation) and higher n_2 (i.e., more dislocation annihilation) generate more severe plastic deformation whose work is represented by temperature rise up to 800 K subjected to the given load. Compared with the mild interaction observed in Figure 9a, the shape of temperature variation as a function of the two parameters also indicate that there is a strong interaction effect in this case. This is also apparent from the relatively large discrepancies between the first order and total effect indices shown in Fig. 10a.

5.3 Polycrystalline sensitivity analysis

Parameter sensitivity analysis within a polycrystalline mesostructure is also of significant interest, as such an analysis explains the behavior at the scale of a particle. In this section, we investigate the sensitivity of elasticity and plasticity within polycrystalline HMX subjected to impact loading.

Figure 12 shows the morphology, loading and boundary conditions of the mesostructure considered in this study. The mesh of the mesostructure consists of 33,530 wedge elements to capture the localized stress evolution and heat generation. The size of the numerical specimen is 3 mm $\times$ 1 mm that is made of 91 single crystals with random orientations sampled from uniform random distribution. The impact loading is applied from the left edge of the specimen, where the edge velocity is linearly increased from rest to 250 m/s in 10 ns and kept constant thereafter. The total duration of each mesoscale simulation is 1 micro-s, with time step size of 1e-6 micro-s. The geometry is modeled as a quasi 2D domain with three dimensional discretization constrained in the third (out of plane) direction with a single set of elements along the thickness direction.

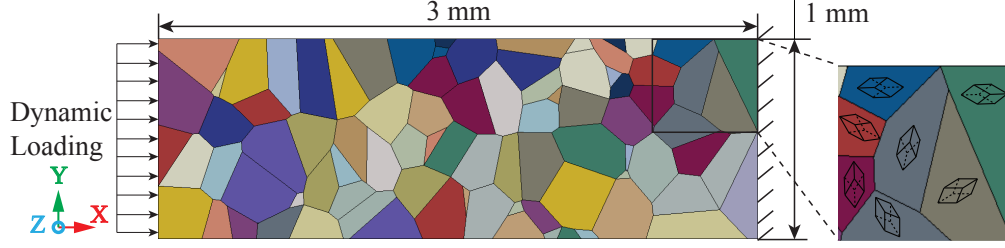


Figure 12: Polycrystal specimen geometry. For the first normal distribution of particle size, $N(\mu_1, \sigma_1)$, the weight $w=0.2$, $\mu_1=61.8$ micro-m, $\sigma_1=13.9$ micro-m. For the second normal distribution $N(\mu_2, \sigma_2)$, $\mu_2=225.7$ micro-m, $\sigma_2=44.9$ micro-m.

The size of the grains are sampled from a bimodal distribution, which is constructed according to the following available experimental data and numerical investigations [25, 49, 60, 75]. Previous studies in energetic crystals point to a strong crystal size effect on the hot spot formation and initiation sensitivity, where the temperature rise is correlated with the square root of the crystal diameter [1, 2]. The current study does not consider a size-dependent constitutive behavior. The Euler angles (Kocks convention) defining the orientation of a crystal is assumed to be independent of each other, and subjected to a uniform random distribution within the given range ($0 < \psi_1 < 2\pi$, $0 < \phi < \pi$ and $0 < \psi_2 < 2\pi$). The normalized histogram of the Euler angles of all crystals of the polycrystal specimen is shown in Fig. 13.

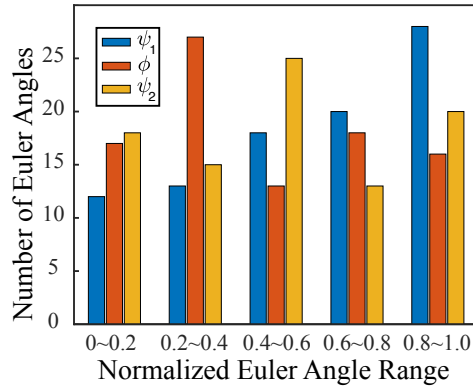


Figure 13: Crystal orientation histogram for the specimen in Fig. 12. The x axis is the normalized angle range ($0 < \psi_1/(2\pi) < 1, 0 < \phi/\pi < 1, 0 < \psi_2/(2\pi) < 1$).

Figure 14 illustrates the stress contours as the stress wave progresses through the polycrystal specimen generated using the model parameters stated in Section 4. Local stress concentrations are apparent particularly along grain boundaries and triple junctions.

The dislocation density generation and annihilation continuously drive local increase or decrease of the material strength, slip and plastic work in a complex fashion. Evolution of dislocation density distribution over the specimen is shown in Fig. 15. The misorientation between adjacent grains leads to the concentration of dislocations along grain boundaries and within particles.

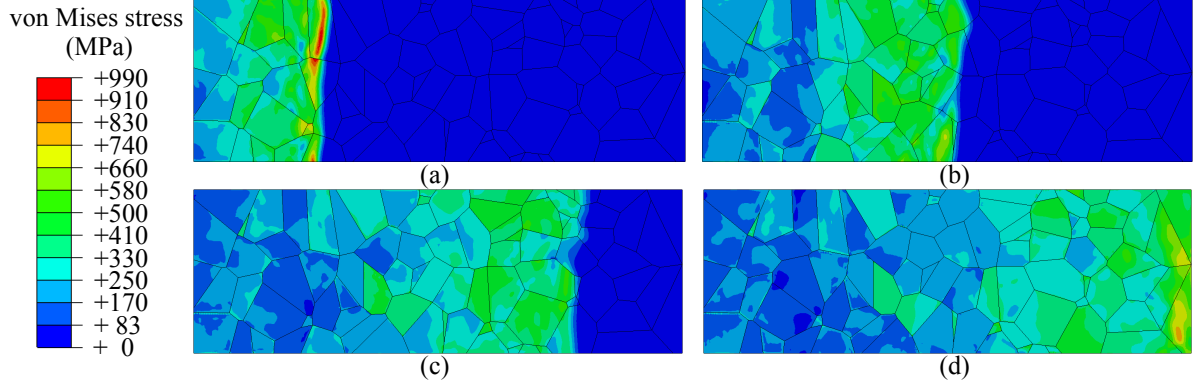


Figure 14: Von Mises stress contours at (a) $t = 0.25$ micro-s, (b) $t = 0.5$ micro-s, (c) $t = 0.75$ micro-s, and (d) $t = 1.0$ micro-s.

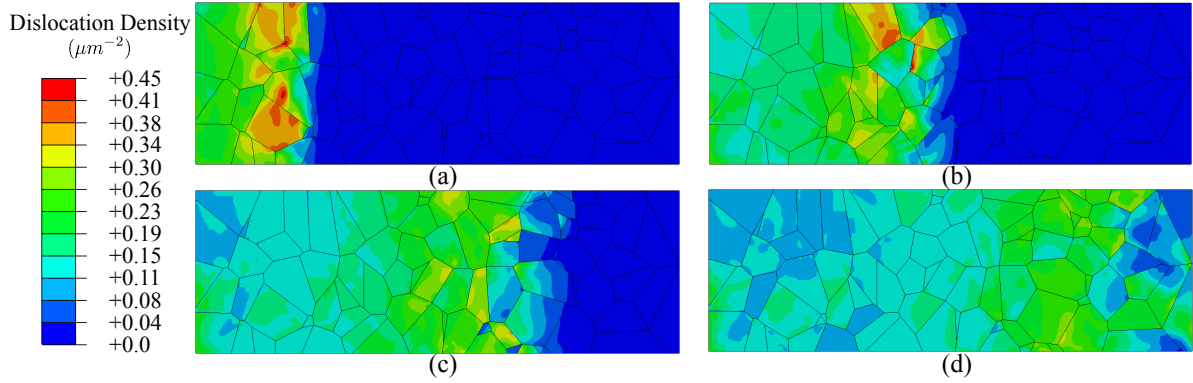


Figure 15: Dislocation density contours at (a) $t = 0.25$ micro-s, (b) $t = 0.5$ micro-s, (c) $t = 0.75$ micro-s, and (d) $t = 1.0$ micro-s.

Similar to the previous study, the primary response function of interest is the peak temperature within the domain, which represents the most critical dissipative state across the mesostructure. In what follows, the mesostructural morphology is fixed and we focus on the analysis of the effects of elastic and plastic properties of the polycrystal.

5.3.1 Elasticity coefficients

Approximately 2,000 forward mesoscale simulations are performed with randomly sampled parameters to serve as training data set for the GP model, and 140 million predictions were made using the surrogate to compute sensitivity indices.

Figure 16a shows the sensitivity indices for the elastic parameters along with sensitivity convergence plot as a function of sample size. The plot clearly indicates that C_{33} is the most sensitive parameter under given impact loading. C_{33} is the lone volumetric component among these nine coefficients, and the corresponding high sensitivity is due to the confinement effect induced by the boundary condition employed in the lateral direction.

OAT variabilities of the elastic parameters are plotted in Fig. 17. C_{13} and C_{66} separately generate the largest temperature variations (~ 11 K) even though they are far less sensitive

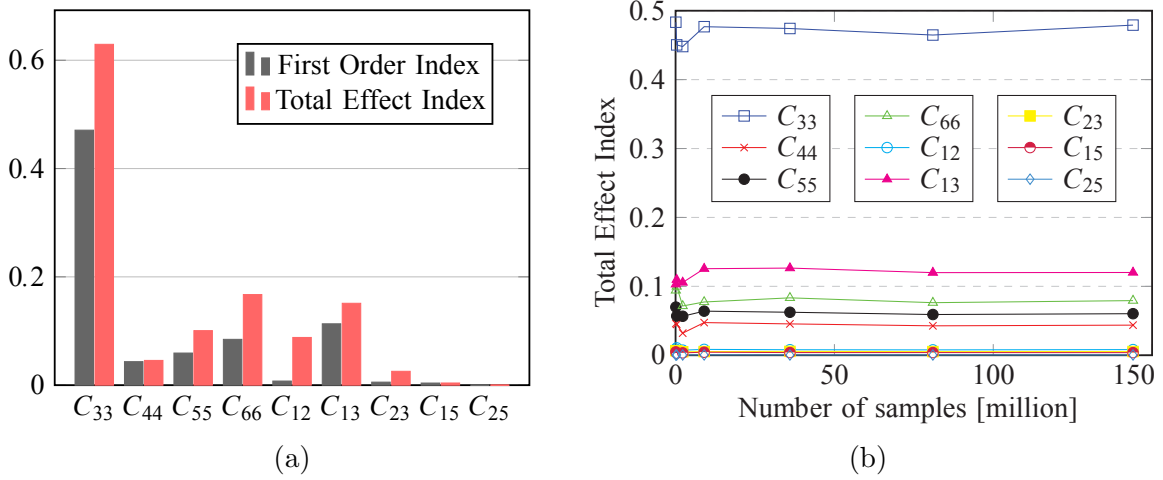


Figure 16: Summary of (a) sensitivity indices and (b) convergence for elastic parameters in polycrystal specimen.

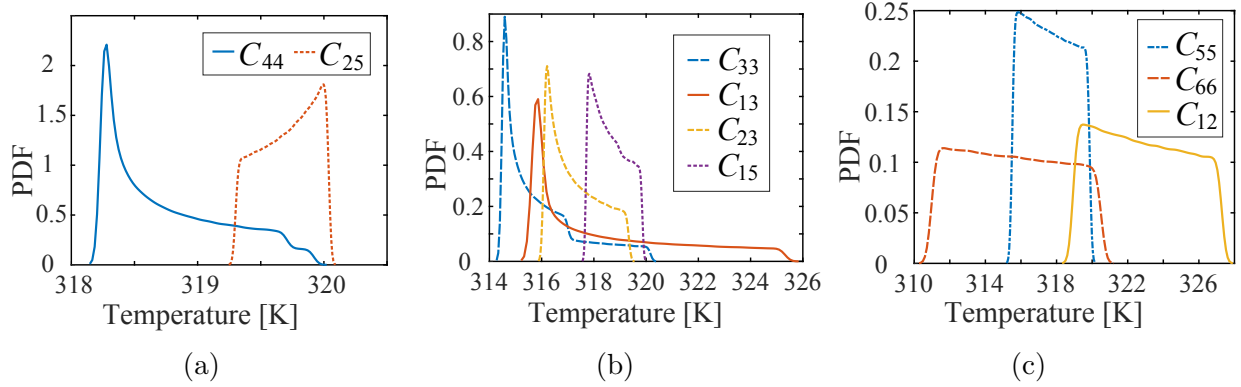


Figure 17: Probability distributions for peak temperature resulting from the OAT analysis of individual elastic constants for polycrystal specimen.

compared with C_{33} across the parameter subspace investigated in this study. Separately varying the parameter C_{15} , C_{25} and C_{44} does not produce significant temperature rise (<3 K), which is consistent with the corresponding low sensitivity indices in Fig. 16a. The variation of peak temperature over C_{33} - C_{66} plane is shown in Fig. 18a. Similar to the single crystal case, the plot indicates a convex shape with varying C_{66} for fixed C_{33} and a linear variation with C_{33} for a fixed C_{66} . In the present case, the curvature does vary with the value of C_{33} , which indicates some interactive effects between the two most influential parameters.

5.3.2 Plastic deformation mechanisms

Plastic deformation within the polycrystal mesostructure includes the crystallographic slip within each crystal, and the interaction between the crystals. The initial dislocation density range is taken to be larger ($[0.0307 \text{ micro-m}^{-2}, 3.07 \text{ micro-m}^{-2}]$) in view of the high potential variability of this parameter as a function of processing conditions.

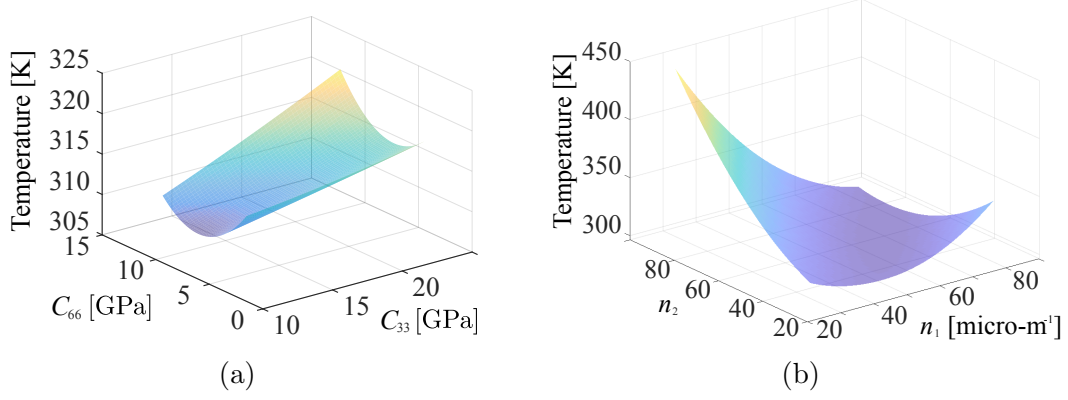


Figure 18: Variation of peak temperature over (a) C_{44} - C_{23} plane and (b) n_1 - n_3 plane for polycrystal cases.

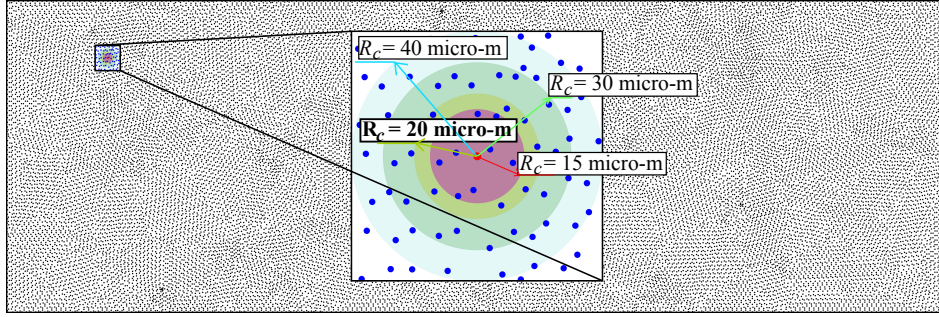


Figure 19: Computation of the nonlocal peak temperature. The dots represent integration points within the specimen discretization. The circles represent the searching areas with the critical radii. The central point (marked as red in the electronic version of the manuscript) is the position where the nonlocal average is computed.

In the current case, we have employed a nonlocal definition of the primary response function (i.e., peak temperature) to ensure that numerical singularities introduced due to irregular element shapes or artificial viscosity do not pollute the sensitivity results. The response function is described as the distance-weighted average maximum temperature over a circular domain. As shown in Fig. 19, the peak nonlocal temperature at an arbitrary position $\hat{x}$ is computed as the weighted temperature over neighbor elements within the circular domain defined by the critical radius R_c . The nonlocal weight $w(x, \hat{x})$ of temperature at x with respect to the center $\hat{x}$ is expressed using the Wendland Radial Basis function:

$$w(x, \hat{x}) = \begin{cases} \left(1 - \frac{\|x - \hat{x}\|}{R_c}\right)^4 \left(4 \frac{\|x - \hat{x}\|}{R_c} + 1\right), & \|x - \hat{x}\| \leq R_c \\ 0, & \|x - \hat{x}\| > R_c \end{cases} \quad (24)$$

The time evolution of the peak nonlocal temperatures (i.e., the maximum nonlocal temperature over the entire domain of the specimen) obtained using different critical radii are shown in the Fig. 20. The results are generated using the model parameters stated in Section 4. The

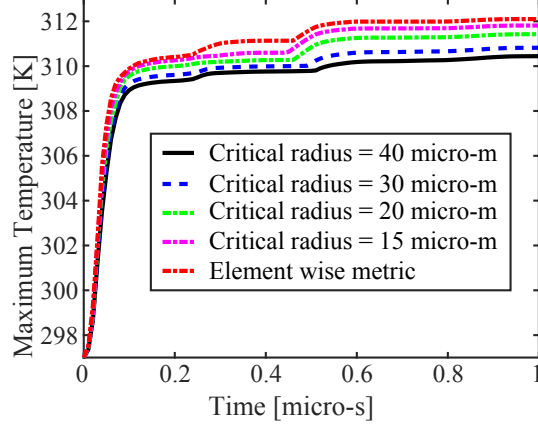


Figure 20: Maximum temperature-time curves with different critical radii.

element size used in the discretization of the domain is approximately 10 micro-m. The general trend of the time evolution of the peak nonlocal temperature does not significantly change by the nonlocal radius, indicating that the numerical singularities do not appear to significantly affect the peak temperature even when local (i.e., element-wise) peak temperature is chosen as the response function. The magnitude of the temperatures naturally reduce with increasing nonlocal radius. The sensitivity analysis below employs a nonlocal radius of 20 micro-m.

The sensitivities were assessed based on 128 million predictions using the surrogate model trained with approximately 1280 FE simulations. The sensitivity indices and index convergence are shown in Fig. 21.

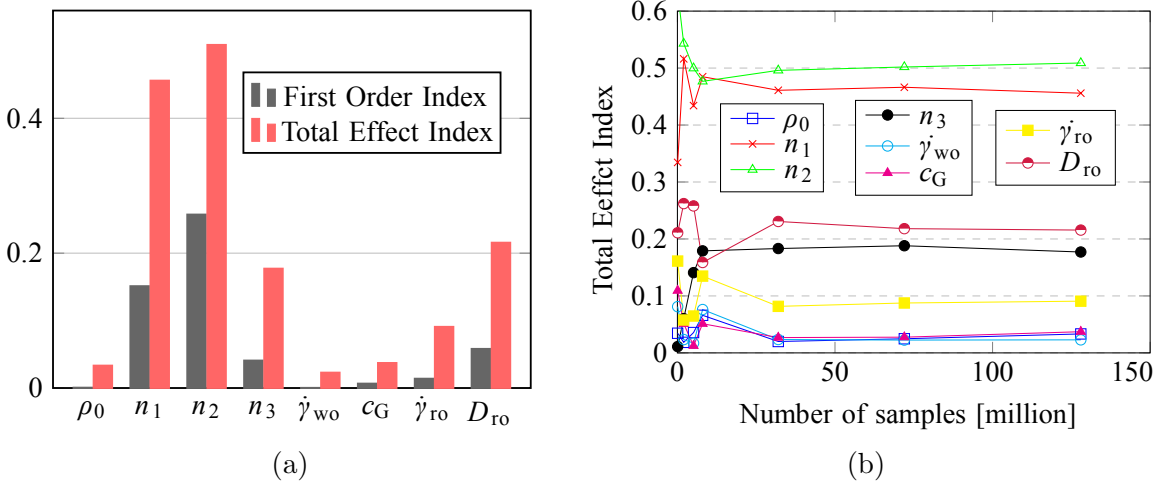


Figure 21: Summary of (a) sensitivity indices and (b) convergence for plastic parameters in polycrystal specimen.

The results of the sensitivity indices are consistent with those from the single crystal analyses that the dislocation generation (n_1) and annihilation (n_2 or n_3) along with phonon drag mechanisms ($\dot{\gamma}_{ro}$ and D_{ro}) primarily describe the plastic deformation and dissipation processes

that occur under the applied dynamic loading. The distribution functions computed based on the OAT approach (Fig. 22) also indicate that, while the overall contributions and variabilities differ from those computed based on the GSA, the effects of phonon drag and dislocation density evolution are dominant. The key dislocation density evolution parameters that describe the generation and annihilation terms interact in a nonlinear fashion to describe the overall dissipative response and consequent temperature rise in the mesostructure, as shown in Fig. 18b. An interesting observation is that less dislocation generation (small n_1) and more dislocation annihilation (larger n_2 or n_3) lead to larger plastic deformation. This is due to the fact that the transition to phonon drag mechanism is significantly influenced by the ratio τ^α/g^α while smaller slip strength leads to earlier transition.

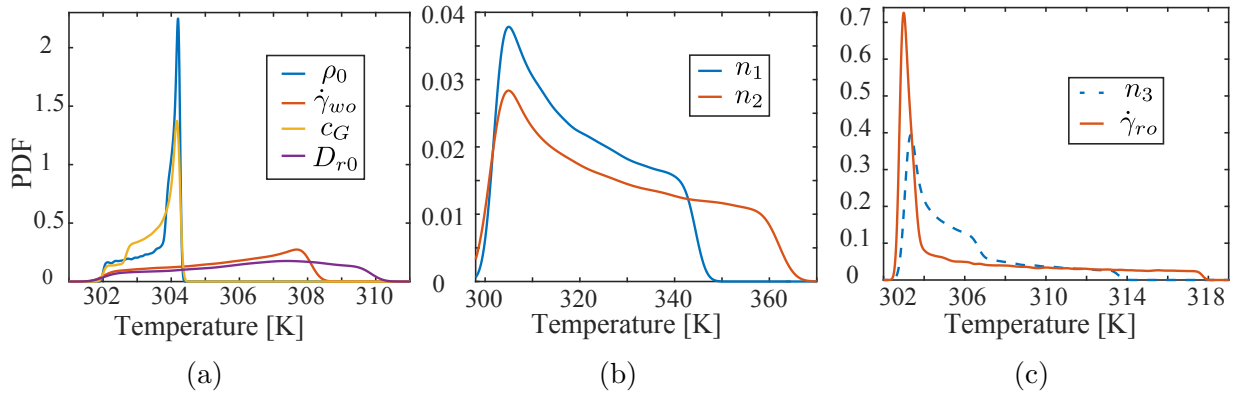


Figure 22: Probability distributions for peak temperature resulting from the OAT analysis of the individual plasticity parameters for polycrystal specimen.

Both OAT and GSA in single crystal and polycrystal specimen indicate that the thermal activation mechanism has little contribution to the temperature at the current time scale and load amplitude, while the phonon drag mechanism, instead, has much larger slip rate and dominates the shear deformation.

6 Conclusion

This manuscript presented the investigations of sensitivity of the response of crystalline β -HMX with respect to anisotropic elasticity constants and crystal plasticity properties under given impact loading through the proposed parameter sensitivity analysis framework. The results show that the anisotropic elasticity coefficients in the monoclinic crystalline have a modest effect on the energy dissipation and temperature rise dominated by sensitivities of a few coefficients. Among the two primary slip mechanisms, phonon drag appears dominant within the load rate amplitude regimes considered in this study. The dominating and non-dominating coefficients, slip mechanisms or dislocation evolution kinetics are identified through both OAT and GSA method, and the discrepancy between results are observed. The initial dislocation density appears to be not particularly influential. The quantified sensitivities of

these mechanisms point to the main deformation mechanisms contributing to the complicated physical phenomena observed at mesoscale. The outcomes of the present analyses will be employed to suggest evolution laws that can capture the localized behavior by only including the physics contributing to the behavior of interest.

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7 Appendix: Tensor of elastic moduli

The vector representations of the deviatoric stress and strain are expressed with five independent components as [34]:

$$\begin{aligned} \text{dev}\boldsymbol{\tau} \rightarrow \{\tau'\} &= \left\{ \frac{1}{\sqrt{2}}(\tau'_{11} - \tau'_{22}) \quad \sqrt{\frac{2}{3}}\tau'_{33} \quad \sqrt{2}\tau'_{12} \quad \sqrt{2}\tau'_{13} \quad \sqrt{2}\tau'_{23} \right\}^T \\ \text{dev}\boldsymbol{\epsilon}^e \rightarrow \{\epsilon^{e'}\} &= \left\{ \frac{1}{\sqrt{2}}(\epsilon^{e'}_{11} - \epsilon^{e'}_{22}) \quad \sqrt{\frac{2}{3}}\epsilon^{e'}_{33} \quad \sqrt{2}\epsilon^{e'}_{12} \quad \sqrt{2}\epsilon^{e'}_{13} \quad \sqrt{2}\epsilon^{e'}_{23} \right\}^T \end{aligned} \quad (25)$$

The deviatoric and hydrostatic components of the constitutive law are written in the matrix notation as:

$$\begin{aligned} \{\tau'\} &= [\tilde{\mathbb{C}}_d^e]\{\epsilon^{e'}\} + \{\tilde{H}^e\}\text{tr}(\boldsymbol{\epsilon}^e) \\ p_\tau &= \{\tilde{H}^e\}^T\{\epsilon^{e'}\} + \tilde{M}^e\text{tr}(\boldsymbol{\epsilon}^e) \end{aligned} \quad (26)$$

where $[\cdot]$ indicates the matrix representation of a tensor. The contracted forms of the fourth order deviatoric elasticity tensor $\tilde{\mathbb{C}}_d^e$, the deviatoric-isochoric elastic coupling tensor $\tilde{H}^e$ and the elastic volumetric coefficient $\tilde{M}^e$ derived with respect to the crystal axis (indicated by the subscript 0) of the monoclinic lattice are:

$$[\mathbb{C}_d^e]_0 = \begin{bmatrix} C_{11}^* & C_{12}^* & 0 & C_{14}^* & 0 \\ C_{12}^* & C_{22}^* & 0 & C_{24}^* & 0 \\ 0 & 0 & C_{33}^* & 0 & C_{35}^* \\ C_{14}^* & C_{24}^* & 0 & C_{44}^* & 0 \\ 0 & 0 & C_{35}^* & 0 & C_{55}^* \end{bmatrix} \quad (27)$$

where

$$\begin{aligned}
C_{11}^* &= \frac{C_{11}}{2} - C_{12} + \frac{C_{22}}{2} \\
C_{12}^* &= -\frac{\sqrt{3}}{6}(C_{11} - 2C_{13} - C_{22} + 2C_{23}) \\
C_{14}^* &= C_{15} - C_{25} \\
C_{22}^* &= \frac{1}{6}(C_{11} + 2C_{12} - 4C_{13} + C_{22} - 4C_{23} + 4C_{33}) \\
C_{24}^* &= -\frac{\sqrt{3}}{3}(C_{15} + C_{25} - 2C_{35}) \\
C_{33}^* &= 2C_{44}, \quad C_{35}^* = 2C_{46}, \quad C_{44}^* = 2C_{55}, \quad C_{55}^* = 2C_{66}
\end{aligned} \tag{28}$$

$$\{H^e\}_0 = [H_1^* \quad H_2^* \quad 0 \quad H_4^* \quad 0] \tag{29}$$

where

$$\begin{aligned}
H_1^* &= \frac{\sqrt{2}}{6}(C_{11} + C_{13} - C_{22} - C_{23}) \\
H_2^* &= -\frac{\sqrt{6}}{18}(C_{11} - 3C_{23} - 3C_{33} - 3C_{13} + 2C_{12} + C_{22} + \frac{2|C_{13}|^2}{C_{13}} + \frac{2|C_{23}|^2}{C_{23}} + \frac{|C_{33}|^2}{C_{33}}) \\
H_4^* &= \frac{\sqrt{2}}{3}(C_{15} + C_{25} + C_{35})
\end{aligned} \tag{30}$$

$$M_0^e = \frac{1}{9}(C_{11} + 2C_{12} + 2C_{13} + C_{22} + 2C_{23} + C_{33}) \tag{31}$$

The tensor of elastic moduli in the intermediate configuration is obtained as:

$$\tilde{\mathbb{C}}_d^e = (\underline{\mathbf{C}} \otimes \underline{\mathbf{C}}) : \mathbb{C}_{d0}^e : (\underline{\mathbf{C}} \otimes \underline{\mathbf{C}})^T \tag{32}$$

where $\mathbf{C} = \mathbf{R}^e \mathbf{C}_0$ represents the rotation from crystal coordinates to the global coordinates, and $(\underline{\mathbf{C}} \otimes \underline{\mathbf{C}})_{ijAB} = C_{iA} C_{jB}$. The initial orientation $\mathbf{C}_0$ is prescribed as part of the crystal initial state in terms of the Euler angles in Kocks convention.

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