On the thermodynamics of higher-order gradient plasticity for size-effects at the micron and submicron length scales

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Abstract: A physically motivated and thermodynamically consistent formulation of higher-order gradient plasticity theory is presented. This proposed model is a two non-local parameter framework that takes into consideration: (1) the presence of plastic strain gradients, which is motivated by the evolution of dislocation density tensor that results from non-vanishing net Burgers vector and, hence, incorporating additional kinematic hardening and (2) the presence of gradients in the equivalent (effective) plastic strain (history variable), which is motivated by the accumulation of geometrically necessary dislocations and, hence, incorporating additional isotropic hardening. It is demonstrated that the non-local yield condition, flow rule and non-classical microscopic boundary conditions can be derived directly from the principle of virtual power. It is also shown that the local Clausius-Duhem

inequality does not hold for gradient-dependent material and, therefore, a non-local form should be adopted. Applications of the proposed theory for size effects in metallic thin films are presented.

Keywords: gradient plasticity; non-local effect; size effect; thin films; length scale; interface effect; surface effect; interfacial energy; microscale; nanoscale.

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1 Introduction

The emerging areas of micro and nanotechnologies exhibit important strength differences that result from continuous modification of the material microstructural characteristics with changing size, with smaller being stronger. There are many experimental observations which indicate that, under certain specific conditions, the specimen size may significantly affect deformation and failure of the engineering materials and a length

scale is required for their interpretation. For example, experimental work on particle-reinforced composites has revealed that a substantial increase in the macroscopic flow stress can be achieved by decreasing the particle size while keeping the volume fraction constant (e.g. Lloyd, 1994). A similar strengthening effect associated with decreasing the diameter of thin wires in microtorsion test has been reported by Fleck et al. (1994) and with decreasing the thickness of thin beams in microbending test has been reported by Stolken and Evans (1998). Moreover, micro and nanoindentation tests have shown that the material hardness increases with decreasing indentation size (e.g. Abu Al-Rub and Voyiadjis, 2004a,b; Voyiadjis and Abu Al-Rub, 2002, 2005). Some of these size effects problems have been addressed previously (Aifantis, 1999, 2001, 2003) within a simpler gradient plasticity framework based on the incorporation of gradients in the equivalent plastic strain alone in accordance with the original (first robust) version of gradient plasticity theory (Aifantis, 1984a, 1987).

The aforementioned dependence of mechanical response on size could not be explained by the classical continuum mechanics since no length scale enters the constitutive description. A multiscale continuum theory, therefore, is needed to bridge the gap between the classical continuum theories and micromechanical theories. Since the increase in strength with decreasing scale can be related to proportional increase in the strain gradients in each of the aforementioned experiments, the gradient plasticity theory has been successful in addressing the size effect problem. This success stems out from the incorporation of a microstructural length scale parameter by means of a gradient-dependent yield condition as first illustrated in the aforementioned works (Aifantis, 1984a; Fleck et al., 1994) and a number of follow up papers based on gradient or non-local constitutive equations for plasticity and damage. Over the past decade, a physical basis of the gradient plasticity theory for metals has been founded on the concept of geometrically necessary dislocations (Ashby, 1970). An extensive review of the recent developments in gradient-dependent theory can be found in Abu Al-Rub (2004), Abu Al-Rub and Voyiadjis (2004a) and Voyiadjis and Abu Al-Rub (2002, 2005). An exception to this is the recent work by Aifantis and Willis (2004, 2005, 2006) who presented a gradient plasticity theory incorporating the gradient of the full plastic strain tensor rather than its scalar equivalent plastic strain gradient counterpart, along with an interfacial energy term, and used this theory in the sequel to address size effect problems through a homogenisation procedure.

Numerous variational formulations and thermodynamic frameworks have been proposed since the 1990s in order to extend the classical (local) constitutive theory to a non-local (gradient-dependent) theory. However, these constitutive models are far from being firmly established. Moreover, the various proposals are also quite different with respect to the structure of the equations. Mühlhaus and Aifantis (1991) formulated the classical (local) incremental variational principle by incorporating first-order gradients of the effective (scalar) plastic strain variable. Fleck and Hutchinson (2001) formulated a gradient theory based on the variational principle of Mühlhaus and Aifantis (1991) but with different constitutive structure. Valanis (1996) and Fremond and Nedjar (1996) postulated the dependence of the Helmholtz free energy on scalar variables and on its first-order gradients. Additionally, they checked the consistency of their constitutive equations using the classical (local) form of the Clausius-Duhem inequality. However, Polizzotto and Borino (1998) assumed a non-local form of the Clausius-Duhem inequality based on the concept of non-locality energy residual. Shizawa and Zbib (1999) derived a rigorous thermodynamical theory of finite strain

gradient plasticity by incorporating kinematic gradient hardening effects through the concept of dislocation density tensor. Another thermodynamic approach was developed by Gurtin (2003) who treated the plastic strain gradients as independent variables, which is different than the above frameworks, and introduced the concept of microforce balance. A very similar framework has been proposed by Gudmundson (2004) and in a different way, with a far reaching elaboration on the interfacial energy term and the subsequent homogenisation procedure, in the aforementioned work of Aifantis and Willis (see also Aifantis,[1] 2004/MS Thesis, 2005/PhD Thesis). Thermodynamic approaches to gradient-dependent plasticity and damage models are developed by Voyiadjis et al. (2004) who postulated the dependence of the Helmholtz free energy on scalar and tensorial hardening/softening variables and its second-order gradients. In this connection, it is pointed out that the standard form of Clausius-Duhem inequality prohibits the Helmholtz free energy to depend on strain gradients. This problem was overcome in a previous thermodynamic treatment (Aifantis, 1984b,c) where the energy equation was generalised to include an extra working term due to the strain gradients. In the present paper, the aforementioned difficulty is circumvented by maintaining the standard form of the energy equation but modifying the Clausius-Duhem inequality to reflect the introduction of non-local effects.

The objective of this paper is to develop a consistent thermodynamic framework that extends the J_2 flow theory to include the effects of strain gradients. This is achieved within the extent of small strain/small rotation plasticity and rate-independent material response. The principle of virtual power and the second-law of thermodynamics are used here to extend the well known procedures of classical thermodynamics and local-type constitutive theory to gradient-dependent theory. This theory is a two non-local parameter theory of gradient type that takes into account large variations in the plastic strain tensor and large variations in the accumulated (scalar) plastic strain. Both isotropic and kinematic gradient hardening effects are considered in the present work. In addition, a framework for the formulation of consistent microscopic boundary conditions is presented. Using a simplified version of the proposed theory, the length scale effects are investigated for biaxial tension of a metallic thin film on a thick elastic substrate.

Notation: Hereafter, $\|\ \|$ is the Euclidean norm of second rank tensors, $(:)$ stands for tensor contraction, the superimposed dot $(\cdot)$ indicates the differentiation with respect to time t, and a comma followed by an index i denotes differentiation with respect to x_i. The first-order gradient, divergence, curl and Laplacian of a tensor field $\mathbf{A}$ are defined by $(\mathrm{div}\,\mathbf{A})_i = A_{ij,j}$, $(\mathrm{curl}\,\mathbf{A})_{ij} = e_{ipq}A_{jq,p}$, $(\nabla^2\mathbf{A})_{ij} = A_{ij,kk}$ and $(\boldsymbol{\nabla}\mathbf{A})_{ijk} = A_{ij,k}$, respectively.

2 Crystallographic basis for presence of plastic strain gradients

The classical theory of isotropic plastic solids undergoing small deformations is based on the additive decomposition of the total strain, $\boldsymbol{\varepsilon}$, into elastic and plastic parts, where $\boldsymbol{\varepsilon}^e$ being the elastic component and $\boldsymbol{\varepsilon}^p$ being the corresponding plastic component such that:

$$\varepsilon_{ij} = \varepsilon_{ij}^e + \varepsilon_{ij}^p, \quad \varepsilon_{kk}^p = 0 \tag{1}$$

From the kinematics of dislocation motion, the plastic part of the total strain arising solely from slips, $p^{(\beta)}$, on all systems ($\beta = 1,...,N$) is taken to be

$$\varepsilon_{ij}^{p} = \sum_{\beta} p^{(\beta)} \mu_{ij}^{(\beta)}, \quad \mu_{ij}^{(\beta)} = \tfrac{1}{2}\left(s_i^{(\beta)} n_j^{(\beta)} + n_i^{(\beta)} s_j^{(\beta)} \right) \tag{2}$$

where $\boldsymbol{\mu}^{(\beta)}$ is the symmetric Schmidt orientation tensor, and $\mathbf{s}^{(\beta)}$ and $\mathbf{n}^{(\beta)}$ are the unit vectors characterising the slip direction on the slip plane β and its normal, respectively, such that $\| \mathbf{s}^{(\beta)} \| = \| \mathbf{n}^{(\beta)} \| = 1$ and $\mathbf{s}^{(\beta)} \, \mathbf{n}^{(\beta)} = 0$.

Material deformation in metals enhances the dislocation formation, the dislocation motion, and the dislocation storage. The dislocation storage causes material hardening. The stored dislocations generated by trapping each other in a random way are referred to as Statistically Stored Dislocations (SSDs), while the stored dislocations that maintain the plastic deformation compatibilities within the polycrystal caused by non-uniform dislocation slip are called Geometrically Necessary Dislocations (GNDs). Their presence causes additional storage of defects and increases the deformation resistance by acting as obstacles to the SSDs (Ashby, 1970). In order to account for strain-gradient effects, one makes use of the GNDs concept. The Nye's dislocation density tensor $\boldsymbol{\alpha}$, which is a representation of GNDs such that α_{ij} is the *i*-component of the resultant Burgers vector related to GNDs of line vector *j*, is defined as (Nye, 1953)

$$\alpha_{ij} = \sum_{\xi} \rho_G^{(\xi)} b_i^{(\xi)} t_j^{(\xi)} \tag{3}$$

where $\rho_G^{(\xi)}$ is the density of GNDs along slip system $\xi = 1,...,G$, $\mathbf{b}^{(\xi)}$ is the Burgers vector and $\mathbf{t}^{(\xi)}$ is the unit vector tangent to a GND. The total accumulation of GNDs is obtained as the magnitude of Nye's dislocation density tensor, $\alpha = \| \boldsymbol{\alpha} \|$ such that if one neglects the interaction among different slip systems, α reads

$$\alpha = \rho_G b \tag{4}$$

where $b = \| \mathbf{b}^{(\xi)} \|$ is the magnitude of the Burgers vector, $\rho_G = \sum_{\xi} \rho_G^{(\xi)}$ is the total density of GNDs, and $\| \mathbf{t}^{(\xi)} \| = 1$.

For irrotational plastic flow (i.e. neglecting the plastic spin tensor), $\boldsymbol{\alpha}$ can be related to the gradient of plastic strain tensor, $\nabla \boldsymbol{\varepsilon}^{p}$, by Arsenlis and Parks (1999), Bassani (2001) and Fleck et al. (1994)

$$\alpha_{ij} = e_{jkl} \varepsilon_{il,k}^{p} \tag{5}$$

where e_{irq} is the permutation tensor. By substituting Equation (2), into Equation (5), $\boldsymbol{\alpha}$ can be written as follows:

$$\alpha_{ij} = \sum_{\beta} p_k^{(\beta)} \eta_{ikj}^{(\beta)}, \quad \eta_{ikj}^{(\beta)} = e_{jkl} \mu_{il}^{(\beta)} \tag{6}$$

Therefore, Equation (5) relates the dislocation density tensor to the slip gradients. Equating Equations (3) and (5) give the gradient of plastic strain tensor in terms of GNDs density as

$$e_{jkl} \varepsilon_{il,k}^{p} = \sum_{\xi} \rho_G^{(\xi)} b_i^{(\xi)} t_j^{(\xi)} \tag{7}$$

Neglecting the interaction among different slip systems, one can define the accumulation of GNDs, α from Equation (5) as follows:

$$\alpha = \sum\nolimits_{\beta} \sqrt{p_k^{(\beta)} p_k^{(\beta)}} \tag{8}$$

It can be seen that the accumulation of GNDs is related to the magnitude of plastic slip gradients. Equating Equations (4) and (8) give the magnitude of plastic slip gradients in terms of the GNDs density as

$$\sum\nolimits_{\beta} \sqrt{p_k^{(\beta)} p_k^{(\beta)}} = \rho_G b \tag{9}$$

Therefore, from Equations (7) and (9) one should consider the effects of both $\nabla \boldsymbol{\varepsilon}^p$ and ∇p in the development of scale-dependent plasticity. The third-order tensor $(\nabla \boldsymbol{\varepsilon}^p)_{ijk} = \varepsilon^p_{ij,k}$ is attributed to a non-vanishing net Burgers vector at the microscale, while $\left(\nabla p\right)_k = p_k$ is attributed to the accumulation of the GNDs.

In classical continuum plasticity, the isotropic hardening variable, $\dot{p}$, is defined as the rate of the local effective plastic strain, which is intended to measure the SSD density and is expressed by

$$\dot{p} = \left\| \dot{\varepsilon}^p_{ij} \right\| = \sqrt{\dot{\varepsilon}^p_{ij} \dot{\varepsilon}^p_{ij}} \tag{10}$$

while the unit direction of plastic strain tensor, $\mathbf{N}$, is defined as follows:

$$N_{ij} = \frac{\dot{\varepsilon}^p_{ij}}{\| \dot{\varepsilon}^p_{ij} \|} = \frac{\dot{\varepsilon}^p_{ij}}{\dot{p}} \Rightarrow \dot{\varepsilon}^p_{ij} = \dot{p} N_{ij} \Rightarrow \dot{p} = \dot{\varepsilon}^p_{ij} N_{ij} \tag{11}$$

Substituting Equation (2) into the above expression and neglecting interactions between different slip planes, yields $\dot{p} = \sum_{\beta} \dot{p}^{(\beta)}$. Therefore, in the following variational framework the local kinematic fields $\dot{\varepsilon}^p_{ij}$ and $\dot{p}$ as well as the non-local kinematic fields $\dot{\varepsilon}^p_{ij,k}$ and $\dot{p}_{,k}$ are considered.

3 Thermodynamics of higher-order gradient plasticity

3.1 Principle of virtual power

The principle of virtual power is the assertion that, given any subbody Γ, the virtual power expended on Γ by materials or bodies exterior to Γ (i.e. external power) be equal to the virtual power expended within Γ (i.e. internal power). Let $\mathbf{n}$ denotes the outward unit normal to $\partial\Gamma$. The external expenditure of power is assumed to arise from a macroscopic surface traction $\mathbf{t}$, the microtraction stress tensor conjugate to $\dot{\boldsymbol{\varepsilon}}^p$, and the microtraction force associated with the history plasticity variable $\dot{p}$. One, therefore, can write the external virtual power in the following form

$$P_{\text{ext}} = \int_{\partial\Gamma} \left(t_i \delta v_i + m_{ij} \delta \dot{\varepsilon}^p_{ij} + q \delta \dot{p} \right) \mathrm{d}A \tag{12}$$

The kinematical fields $\delta \mathbf{v}$, $\delta \dot{\boldsymbol{\varepsilon}}^p$ and $\delta \dot{p}$ are considered here as virtual, where δ is the variation parameter and $\mathbf{v}$ is the velocity vector. The tensor $\mathbf{m}$ is the microtraction

stress tensor conjugate to $\boldsymbol{\varepsilon}^p$, defined for each unit vector $\mathbf{n}$ normal on the boundary $\partial\Gamma$ of Γ. The scalar q is the microtraction force associated with the effective plastic strain variable, p.

The external power is balanced by an internal expenditure of power characterised by an elastic stress $\boldsymbol{\sigma}$ defined over Γ for all time, the backstress $\mathbf{X}$ associated with kinematic hardening, and the drag-stress R associated with isotropic hardening. However, since the goal of this paper is a theory that allows for gradients of the plastic strain and effective plastic strain, one also considers power expenditures associated with kinematic variables $\nabla\boldsymbol{\varepsilon}^p$ and ∇p. One, therefore, can assume that additional power is expended internally by the higher-order microstress $\mathbf{S}$ conjugate to $\nabla\boldsymbol{\varepsilon}^p$, and the higher-order microforce vector $\mathbf{Q}$ conjugate to ∇p. Specifically, the internal virtual power is assumed to have the following form

$$P_{\text{int}} = \int_{\Gamma} \left(\sigma_{ij}\delta\dot{\varepsilon}^{e}_{ij} + X_{ij}\delta\dot{\varepsilon}^{p}_{ij} + R\delta\dot{p} + S_{ijk}\delta\dot{\varepsilon}^{p}_{ij,k} + Q_k\delta\dot{p}_k \right) \mathrm{d}V \tag{13}$$

and to balance P_{ext}, Equation (12), in the sense that $P_{\text{ext}} = P_{\text{int}}$.

Operating on $\dot{p} = \| \dot{\boldsymbol{\varepsilon}}^p \|$ by the gradient and substituting the result along with $\dot{\boldsymbol{\varepsilon}}^e = \dot{\boldsymbol{\varepsilon}} - \dot{\boldsymbol{\varepsilon}}^p$ and Equations (11)–(13) into the virtual power balance, $P_{\text{ext}} = P_{\text{int}}$, and then applying the divergence theorem yields, after some lengthy manipulations, the following result

$$\begin{aligned} &\int_{\Gamma} \sigma_{ij,j}\delta v_i \,\mathrm{d}V + \int_{\partial\Gamma} \left(t_i - \sigma_{ij}n_j \right) \delta v_i \,\mathrm{d}A + \int_{\Gamma} \left[\tau_{ij} - X_{ij} + S_{ijk,k} - \left(R - Q_{k,k} \right) N_{ij} \right] \delta\dot{\varepsilon}^{p}_{ij} \,\mathrm{d}V \\ &\qquad + \int_{\partial\Gamma} \left[\left(m_{ij} + qN_{ij} \right) - \left(S_{ijk} + Q_k N_{ij} \right) n_k \right] \delta\dot{\varepsilon}^{p}_{ij} \,\mathrm{d}A = 0 \end{aligned} \tag{14}$$

The fields Γ, $\delta\mathbf{v}$ and $\delta\dot{\boldsymbol{\varepsilon}}^p$ may be arbitrarily specified if and only if

$$\sigma_{ij,j} = 0 \,,\; t_i = \sigma_{ij}n_j \tag{15}$$

$$\tau_{ij} - X_{ij} + S_{ijk,k} - \left(R - Q_{k,k} \right) N_{ij} = 0, \quad \tilde{m}_{ij} = \left(S_{ijk} + Q_k N_{ij} \right) n_k \tag{16}$$

such that $\tilde{\mathbf{m}} = \mathbf{m} + q\mathbf{N}$. Equation $(15)_1$ expresses the *macroforce balance* according to the notion of Gurtin (2003). Equation $(15)_2$ defines the stress vector as the surface density of the forces imposed. It shows that $\boldsymbol{\sigma}$ is really the Cauchy stress tensor, which is a second-order symmetric tensor. It also provides the local *macrotraction boundary conditions* on forces. One can view the *microforce balance* in Equation $(16)_1$ as the plasticity non-local yield condition, which is demonstrated in the following section, and the *microtraction condition* in Equation $(16)_2$ as a higher-order condition (or internal boundary condition) augmented by the interaction of dislocations across interfaces.

3.2 Non-local yield criterion

By taking the Euclidean norm $\| \, \|$ of Equation $(16)_1$, the non-local plasticity loading surface f is given by

$$f = \left\| \tau_{ij} - X_{ij} + S_{ijk,k} \right\| - R + Q_{k,k} = 0 \tag{17}$$

where $\mathbf{N}$ is collinear with $\boldsymbol{\tau} - \mathbf{X} + \operatorname{div}\mathbf{S}$, $\| R - Q_{k,k} \| = R - Q_{k,k}$ and $\| \mathbf{N} \| = 1$. It is obvious that Equation (17) represents a sphere in deviatoric stress-space of radius $R - \operatorname{div}\mathbf{Q}$ centred at $\mathbf{X}$ - $\operatorname{div}\mathbf{S}$. One can also notice that the higher-order stress $\operatorname{div}\mathbf{S}$ is a back-stress quantity giving rise to additional kinematic hardening, while the microstress $\operatorname{div}\mathbf{Q}$ is giving rise to additional isotropic hardening.

Since Equation (17) implies that $\mathbf{N}$ is parallel to $\boldsymbol{\tau} - \mathbf{X} + \operatorname{div}\mathbf{S}$, one can, therefore, express the plasticity flow rule, Equation $(11)_2$, as follows:

$$\dot{\varepsilon}_{ij}^{p} = \frac{\dot{p}\left(\tau_{ij} - X_{ij} + S_{ijk,k}\right)}{\left\|\tau_{ij} - X_{ij} + S_{ijk,k}\right\|} \tag{18}$$

such that the direction of plastic flow $\mathbf{N}$ can be given by

$$N_{ij} = \frac{\partial f}{\partial \sigma_{ij}} = \frac{-\partial f}{\partial X_{ij}} = \frac{\partial f}{\partial S_{ijk,k}} \tag{19}$$

Therefore, the flow rule in Equation (18) asserts that the flow direction $\mathbf{N}$ in Equation (19) is normal to the yield surface and directed outward from the yield surface. Moreover, if the higher-order gradients are neglected, one can easily retrieve from Equations (17)–(19), respectively, the classical yield criterion, flow rule and flow direction.

3.3 Non-local clausius-duhem inequality

Utilising Equation (16) into Equation (13), and replacing the virtual quantities by the actual fields, one can rewrite the expression of the internal power as follows:

$$P_{\text{int}} = \int_{\Gamma} \sigma_{ij}\dot{\varepsilon}_{ij}\,\mathrm{d}V + \int_{\partial\Gamma} \tilde{m}_{ij}\dot{\varepsilon}_{ij}^{p}\,\mathrm{d}A \tag{20}$$

Comparing the above equation with its corresponding local expression $P_{\text{int}} = \int_{\Gamma} \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}\,\mathrm{d}V$, implies that the long-range (non-local) energy interactions can be of non-vanishing within the plastic zone, which is represented by the second term in Equation (20). Hence, according to the notion of Eringen and Edelen (1972), the energy term $\int_{\partial\Gamma} \tilde{\mathbf{m}} : \dot{\boldsymbol{\varepsilon}}^{p}\,\mathrm{d}A$ is called as the *non-locality energy residual* that results from microstructural interactions between the material points in the active plastic zone and at interfaces. Therefore, one can define the density of the non-locality energy residual, $\mathbb{R}$, as follows:

$$\int_{\Gamma} \mathbb{R}\,\mathrm{d}V = \int_{\partial\Gamma} \tilde{m}_{ij}\dot{\varepsilon}_{ij}^{p}\,\mathrm{d}A = \int_{\partial\Gamma} \left(S_{ijk} + Q_k N_{ij}\right) n_k \dot{\varepsilon}_{ij}^{p}\,\mathrm{d}A = \int_{\Gamma} \left[\mathbb{Z}_{ijk}\dot{\varepsilon}_{ij}^{p}\right]_{,k}\,\mathrm{d}V \tag{21}$$

such that $\mathbb{R}$ and $\mathbb{Z}$ are given by, respectively,

$$\mathbb{R} = \left[\mathbb{Z}_{ijk}\dot{\varepsilon}_{ij}^{p}\right]_{,k} = \mathbb{Z}_{ijk,k}\dot{\varepsilon}_{ij}^{p} + \mathbb{Z}_{ijk}\dot{\varepsilon}_{ij,k}^{p}, \quad \mathbb{Z}_{ijk} = S_{ijk} + Q_k N_{ij} \tag{22}$$

where $\mathbb{R} \neq 0$ in Γ^{p}, but $\mathbb{R} = 0$ out of Γ^{p} or for a homogeneous plastic deformation (i.e. $\mathbb{R} = 0$ in the absence of plastic strain gradients).

One considers here a purely mechanical theory (*isothermal conditions* are assumed) based on the requirement that the rate of change in the total free energy should be less than or equal to the power done by external forces (Gurtin, 2003). If one denotes $\rho\Psi$ as the specific free energy, this requirement takes the form of a free energy inequality

$$\int_{\Gamma} \rho\dot{\Psi} \mathrm{d}V \le P_{\text{ext}} \tag{23}$$

By substituting Equations (20) and (21) into Equation (23) along with $P_{\text{ext}} = P_{\text{int}}$, one obtains the following thermodynamic restriction in a point wise form:

$$\sigma_{ij}\dot{\varepsilon}_{ij} - \rho\dot{\Psi} + \mathbb{R} \ge 0 \tag{24}$$

The inequality in Equation (24) is termed here as the *non-local Clausius-Duhem inequality* differing from its classical counterpart only in the presence of $\mathbb{R}$. This inequality holds everywhere in Γ, but $\mathbb{R} = 0$ at material points in the elastic zone. Moreover, it can be noted from Equation (22) that for a homogeneous plastic strain distribution $\mathbb{R} = 0$, one retains the classical Clausius-Duhem inequality. The non-local Clausius-Duhem inequality, Equation (24), can be recognised to be substantially coincident with the analogous inequality presented by Polizzotto and Borino (1998). This result credits the proposed framework as a valid approach to model non-local plasticity.

Assuming a separable material, that is, no coupling between the elastic and plastic free energies (Gurtin, 2003), one can write the Helmholtz free energy potential as

$$\Psi = \Psi^e\left(\varepsilon_{ij}^e\right) + \Psi^p\left(\varepsilon_{ij}^p, p, \varepsilon_{ij,k}^p, p_k\right) \tag{25}$$

Taking the time derivative of Equation (25) with respect to its internal state variables and substituting the result in the non-local Clausius-Duhem inequality yield the following thermodynamic state laws:

$$\sigma_{ij} = \rho\frac{\partial\Psi^e}{\partial\varepsilon_{ij}^e}, X_{ij} = \rho\frac{\partial\Psi^p}{\partial\varepsilon_{ij}^p}, R = \sigma_y + \rho\frac{\partial\Psi^p}{\partial p}, S_{ijk} = \rho\frac{\partial\Psi}{\partial\varepsilon_{ij,k}^p}, Q_k = \rho\frac{\partial\Psi}{\partial p_{,k}} \tag{26}$$

where σ_y is the initial yield strength.

In order to develop equations amenable to the analysis and computation, one now considers a simple example for the definition of the Helmholtz free energy function. One can assume decoupling between the elastic behaviour and plasticity hardening (i.e. separable material) such that both Ψ^e and Ψ^p that appear in Equation (25) can be assumed to have, respectively, the following quadratic analytical form:

$$\rho\Psi^e = \tfrac{1}{2}\varepsilon_{ij}^e E_{ijkl}\varepsilon_{kl}^e, \rho\Psi^p = \tfrac{1}{2}a_1\varepsilon_{ij}^p\varepsilon_{ij}^p + \tfrac{1}{2}a_2 p^2 + \tfrac{1}{2}a_3\varepsilon_{ij,k}^p\varepsilon_{ij,k}^p + \tfrac{1}{2}a_4 p_k p_k \tag{27}$$

where $\mathbf{E}$ is the symmetric fourth-order elastic stiffness tensor and a_i ($i = 1-4$) are material constants. Making use of Equation (27) into Equation (26) the following laws can be obtained

$$\sigma_{ij} = E_{ijkl}\left(\varepsilon_{kl} - \varepsilon_{kl}^p\right), X_{ij} = h\varepsilon_{ij}^p, R = \sigma_y + hp, S_{ijk} = B_{ijmn}\varepsilon_{mn,k}^p, Q_k = h\ell^2 p_k \tag{28}$$

where, for simplicity, it is assumed that $a_1 + a_2 = h$ and $a_3 + a_4 = h\ell^2$ with h being the constant hardening modulus, $B_{ijmn} = a_3\delta_{im}\delta_{jn} + a_4 N_{ij}N_{mn}$.

Substituting Equation (28) into the yield function f, Equation (17), one can then write

$$f = \underbrace{\left\| \tau_{ij} - X_{ij} + S_{ijk,k} \right\|}_{\text{effective von-Mises stress}} - \underbrace{\sigma_y - hp + h\ell^2\nabla^2 p}_{\text{isotropic hardening function}} = 0 \tag{29}$$

with

$$S_{ijk,k} = B_{ijmn,k}\varepsilon^p_{mn,k} + B_{ijmn}\nabla^2\varepsilon^p_{mn} \tag{30}$$

where ∇^2 designates the Laplacian operator and $B_{ijmn,k} = a_4(N_{ij,k}N_{mn} + N_{ij}N_{mn,k})$.

Thus, this theory shows that the Laplacian of the effective plastic strain contributes to the size of the yield surface (isotropic hardening), whereas the first-order gradient and the Laplacian of the plastic strain tensor contribute to the movement of the centre of the yield surface (kinematic hardening). It is noteworthy that the present formulation links hardening to the gradients of plastic strain $\nabla\boldsymbol{\varepsilon}^p$ and the effective plastic strain ∇p and, respectively, not to $\nabla^2\boldsymbol{\varepsilon}^p$ and $\nabla^2 p$, consistent with basic notions of the role of the net Burgers vector and the geometrically necessary dislocations. Instead, $\nabla^2\boldsymbol{\varepsilon}^p$ and $\nabla^2 p$ emerges in the resulting field equations as a byproduct of the more fundamental role of the plastic strain gradients.

3.4 Microscale plastic boundary conditions

Polizzotto and Borino (1998) assumed that the non-locality residual in Equation (21) is equal to zero and called it as the *insulation condition* meaning that non-local energy is not allowed to flow from any point in Γ to the exterior of the body. Therefore, for simplicity, if one neglects the interior surface energy that results from dislocation interactions at the surface/interface boundaries (e.g. internal boundaries at inclusions), which is very difficult to be satisfied in reality, the insulation condition can be expressed as

$$\int_\Gamma \mathbb{R}\mathrm{d}V = 0 \tag{31}$$

Therefore, from the above assumption, one may require the application of internal microboundary conditions in a pointwise format if there is no plastic flow across the interfaces, which results in a null expenditure of micropower in the sense that:

$$\tilde{m}_{ij}\dot{\varepsilon}^p_{ij} = 0 \text{ on } \partial\Gamma^p \tag{32}$$

where $\partial\Gamma^p \subseteq \partial\Gamma$ is the plastic boundary. The above equation renders two conditions according to a split of the plastic subdomain boundary into external and internal parts, $\partial\Gamma^p = \partial\Gamma^p_{\text{int}} \bigcup \partial\Gamma^p_{\text{ext}}$:

1 *Microplastic-clamped* boundary condition that is imposed on the internal plastic boundary $\partial\Gamma^p_{\text{int}}$

$$\dot{\varepsilon}_{ij}^{p} = \mathbf{0} \text{ on } \partial\Gamma_{\text{int}}^{p} \tag{33}$$

and gives the so-called continuity boundary condition of Dirichlet type and is meant to characterise, for example, microscopic behaviour at the boundary of a metallic film perfectly bonded to an elastic substrate or metallic matrix perfectly bonded to an elastic inclusion. Therefore, this boundary condition places a constraint on the plastic flow and could characterise the dislocation blocking at the interface. Moreover, $\partial\Gamma_{\text{int}}^{p}$ could characterise the movable elastic-plastic boundary.

2 *Microtraction-free* boundary condition that is imposed on the external plastic boundary $\partial\Gamma_{\text{ext}}^{p} \subseteq \partial\Gamma$

$$\tilde{m}_{ij} = \mathbf{0} \text{ on } \partial\Gamma_{\text{ext}}^{p} \tag{34}$$

and gives the so-called Neumann type boundary which assumes that the microtractions $\tilde{\mathbf{m}}$ vanish at the external surfaces $\partial\Gamma_{\text{ext}}^{p} = \partial\Gamma^{p} \bigcap \partial\Gamma$ (i.e. unmovable external surfaces). This condition is meant to characterise, for example, the free surface of a void in a material or the free surface of a nano-size structural system. Equation (34) places no constraint on the plastic flow and could characterise free dislocation movements across the boundaries.

The role of interfaces in enhancing the yield strength in composites and polycrystals has been addressed recently by Aifantis and Willis (2005, 2006) by introducing quadratic interfacial energies at the hard boundaries. Therefore, non-trivial boundary conditions can be obtained by adopting the approach of Aifantis and Willis (2005, 2006) by simple setting the thermodynamic conjugate force $\tilde{\mathbf{m}}$ as a function of the interfacial energy.

4 Application to size effects in thin films: biaxial loading of a thin film on a substrate

This section presents some recent application of gradient plasticity to handle size effects observed in metals. The proposed gradient plasticity theory is now used to investigate the size dependent behaviour in biaxial loading of a plastic thin film on an elastic substrate. In the following application, for simplicity, kinematic hardenings introduced by the conjugate forces $\mathbf{X}$ and $\mathbf{S}$ that appear in Equation (29) are neglected.

A biaxially loaded isotropic elasto-plastic thin film of thickness t on a thick semi-infinite elastic substrate is considered as shown in Figure 1(a). Let x_3 be the perpendicular axis to the film and $x_3 = 0$ corresponds to the film-substrate interface. The loading is defined by a monotonically increasing biaxial strain ε_o such that $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_o$. A plane stress situation is assumed such that the non-vanishing stress and plastic strain components are

$$\sigma_{11} = \sigma_{22} = \sigma_o\left(x_3\right), \varepsilon_{11}^{p} = \varepsilon_{22}^{p} = -\tfrac{1}{2}\varepsilon_{33}^{p} = \varepsilon_o^{p}\left(x_3\right) \tag{35}$$

The effective plastic strain $p = \sqrt{\varepsilon_{ij}^{p}\varepsilon_{ij}^{p}}$ and its Laplacian $\nabla^2 p$ are given as

$$p = \sqrt{6}\varepsilon_o^{p} \text{ and } \nabla^2 p = \sqrt{6}\varepsilon_{o,33}^{p} \tag{36}$$

where $\varepsilon^p_{o,33} = \partial\varepsilon^p_o / \partial x_3 \partial x_3$. The stress-strain relationship can be simply obtained from the generalised Hooke's law as

$$\sigma_o\left(x_3\right) = \left[\frac{E}{(1-\nu)}\right]\left[\varepsilon_o - \varepsilon^p_o\left(x_3\right)\right] \tag{37}$$

Substituting Equations (35)–(37) into the yield condition, Equation (29), yields the following ordinary differential equation for $\varepsilon^p_o(x_3)$:

$$\varepsilon^p_{o,33} - \left[\frac{1}{\ell^2} + \frac{E}{3(1-\nu)h\ell^2}\right]\varepsilon^p_o = \frac{\sigma_y}{h\ell^2} - \frac{E}{3(1-\nu)h\ell^2}\varepsilon_o \tag{38}$$

It is convenient to express the above equation in a non-dimensional form with the aid of variable substitution (i.e. $z = x_3 / t$, $\overline{\varepsilon}^p_o = \varepsilon^p_o / \varepsilon_y$ and $\overline{\varepsilon}_o = \varepsilon_o / \varepsilon_y$ with $\varepsilon_y = \sigma_y(1-\nu)/E$ being the in-plane yield strain) such that

$$\overline{\varepsilon}^p_{o,zz} - \lambda^2\overline{\varepsilon}^p_o = -F \tag{39}$$

with constant coefficients λ and F are given through

$$\lambda^2 = \frac{1}{(\ell/t)^2}\left(1 + \frac{E}{3(1-\nu)h}\right) \text{ and } F = \frac{E\left(\overline{\varepsilon}_o - 3\right)}{3(1-\nu)h(\ell/t)^2} \tag{40}$$

The microboundary conditions can now be utilised as presented by Equations (33) and (34). The microtraction-free boundary condition is imposed at the free surface (i.e. $x_3 = t$) and the microplastic-clamped boundary condition is imposed at the film-substrate interface such that one can write, respectively, the following

$$\frac{\partial\overline{\varepsilon}^p_o}{\partial z} = 0 \text{ at } z = 1 \text{ and } \overline{\varepsilon}^p_o = 0 \text{ at } z = 0 \tag{41}$$

Solving the ordinary differential equation, Equation (39), which is subjected to the boundary conditions in Equation (41), one obtains a closed-form expression for $\overline{\varepsilon}^p_o(z)$ as

$$\overline{\varepsilon}^p_o(z) = \frac{F}{\lambda^2}(1 - \cosh\lambda z + \tanh\lambda\ \sinh\lambda z) \tag{42}$$

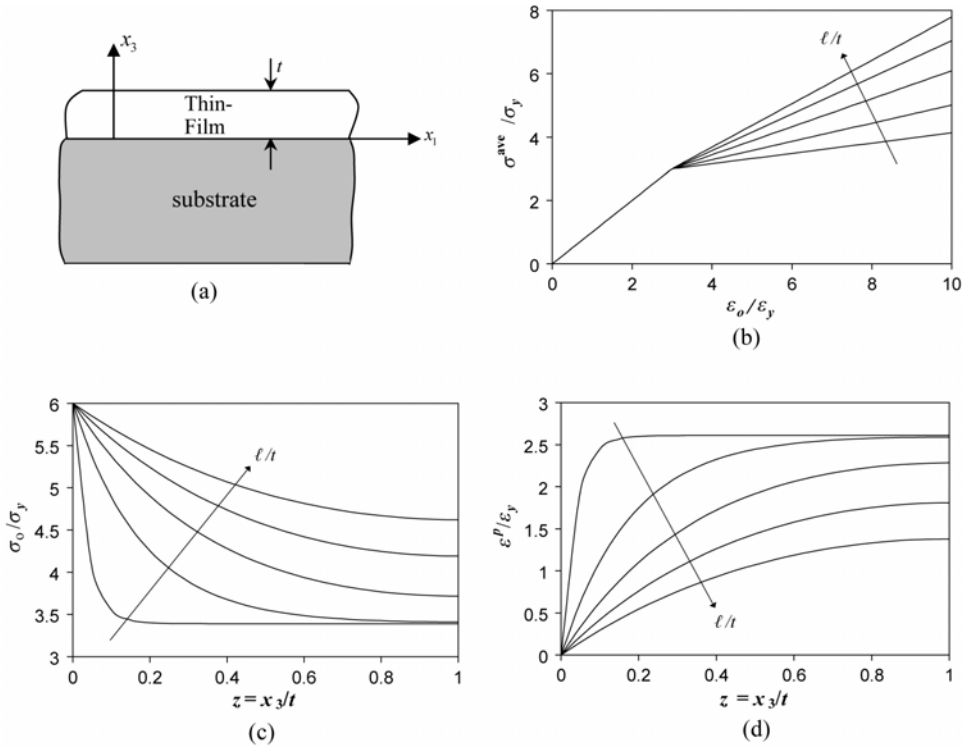
where $\mathrm{H}(\overline{\varepsilon}_o - 3)$ is the Heaviside step function. Substituting Equation (42) back into the normalised expression of Equation (37) (i.e. the in-plane stresses are normalised by the yield stress σ_y such that $\overline{\sigma}_o = \overline{\varepsilon}_o - \overline{\varepsilon}^p_o$), one finds the following

$$\overline{\sigma}_o = \overline{\varepsilon}_o - \frac{E\left(\overline{\varepsilon}_o - 3\right)}{3(1-\nu)h + E}(1 - \cosh\lambda z + \tanh\lambda\ \sinh\lambda z)H\left(\overline{\varepsilon}_o - 3\right) \tag{43}$$

The average stress in the thin-film, $\overline{\sigma}^{\text{ave}}_o$, can be determined from an integration of Equation (43) from 0 to 1, such that:

$$\overline{\sigma}^{\text{ave}}_o = \overline{\varepsilon}_o - \frac{E\left(\overline{\varepsilon}_o - 3\right)}{3(1-\nu)h + E}\left(1 - \frac{\tanh\lambda}{\lambda}\right)H\left(\overline{\varepsilon}_o - 3\right) \tag{44}$$

Figure 1 (a) Biaxial tension of a thin-film on a substrate. Different film thicknesses are represented by ℓ / t = 0.1, 0.5, 1, 1.5, 2 for, (b) average biaxial stress versus biaxial strain, (c) biaxial stress through the thickness at a strain of $\varepsilon_o = 6\varepsilon_y$ and (d) biaxial plastic strain through the thickness



Results in Figure 1 are presented for $h(1-\nu)/E$ = 0.05 and ν = 0.3. Different film thicknesses are represented by ℓ / t = 0.1, 0.5, 1, 1.5, 2. In Figure 1(b), normalis results for average film stress, Equation (44), versus applied biaxial strain ε_o are presented. It is clearly seen that the hardening tangent modulus increases with decreasing the film thickness, which agrees qualitatively with the experimental observations. Figure 1(c) and (d) show the variation of the biaxial stress and plastic strain, Equations (43) and (42), across the film thickness. Instead of a uniform distribution of stress and plastic strain across the film thickness, according to classical local plasticity, the stress increases and the plastic strain decreases as the film-substrate interface is approached. Moreover, the results clearly show that the biaxial stress and plastic strain profiles tend to become homogeneous with increased thickness due to smaller gradient effects. This means that gradients eventually disappear for large thicknesses. As shown in Figure 1(c), the increase of plastic strain at $x_3 = t$ overwhelms the increase in plastic gradients as the substrate is approached due to the distribution of gradients, which is lower as the free boundary is approached and higher at the substrate. The elimination of gradients spreads from the free boundary, propagating through the entire thickness. Finally, the plastic strain becomes uniform across the thickness.

5 Conclusions

The proposed framework is a two non-local parameter theory that takes into account large variations in the plastic strain tensor and large variations in the accumulated (effective) plastic strain. It is argued that the gradient of the plastic strain tensor, $\varepsilon^p_{ij,k}$, and the gradient of the effective plastic strain, p_k, should be incorporated when formulating a gradient plasticity theory since the former accounts for the Nye's dislocation density (or incompatibility) tensor and the later accounts for the accumulation of the geometrically necessary dislocations. The thermodynamic conjugate force associated with $\varepsilon^p_{ij,k}$ introduces kinematic hardening whereas the conjugate force of p_k introduces isotropic hardening.

The formulation of higher-order boundary conditions is very important within strain gradient plasticity theory, especially, at interfaces, grain, or phase boundaries. A framework for the formulation of these additional boundary conditions is presented. The microlevel significance and nature of these boundary conditions is emphasised. It is shown for biaxial loading of a film-substrate system, in which the material is initially homogenous, that in the absence of the higher-order boundary conditions, the material would support uniform fields and hence the constitutive gradient-dependence would have no influence. Therefore, strain gradients come into play if the boundaries are assumed to constrain the plastic flow.

In conclusion, if continuum theories are to be used to predict elastic-plastic behaviour at the micron or submicron length scales, a higher-order theory is likely to be required. Moreover, it would be of particular interest to compare the effects of microtraction stresses at interfaces and free surfaces in the present gradient theory with discrete dislocation dynamics simulations.

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Note

[1]The aim of the MS Thesis, in particular, was to produce a gradient plasticity theory through a variational formulation where the displacement vector and the plastic strain tensor are treated as independent variables on equal footing; in addition to the introduction of an interfacial energy term across which plastic strain is continuous but plastic strain gradients are not, thus necessitating a jump in the respective conjugate stress quantity, that is, the higher-order stress. While the interfacial energy term generalises directly and in a neat way the original plasticity theory (Aifantis, 1984a), the tensorial plastic strain gradients (in contrast to the gradients of the scalar equivalent plastic strain) caused a departure of the resulting gradient plasticity framework from the classical structure based on the concept of a universal flow (equivalent stress versus. equivalent strain) relationship. The need for a compromise between the 1984 original theory and the newly advocated 2004 one was pointed out but it was not pursued further to avoid redundancy in using, in addition to the plastic strain tensor, the equivalent plastic strain as independent variables without a clear physical meaning of the conjugate stress variables and the associated jump conditions at the interface.