On Cold-Rolling (Bonding) of Polymeric Films: Numerical Simulations and Experimental Results

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Abstract

Recently a new phenomenon for bonding of polymeric films in solid-state, at ambient temperatures ($\approx 20^\circ$C) well below the glass transition temperature ($T_g \approx 78^\circ$C) of the polymer, has been reported. This is achieved by bulk plastic compression of polymeric films held in contact. Here we analyze the process of cold-rolling of polymeric films via finite element simulations, and illustrate a flexible and modular experimental rolling-apparatus that is capable of achieving the bonding of polymeric films through cold-rolling. Firstly, the classical theory of rolling a rigid-plastic thin-strip is utilized to estimate various deformation fields such as strain-rates, velocities, loads etc. in rolling the polymeric films at the specified feed-rates and desired levels of thickness-reduction(s). Predicted magnitudes of slow strain-rates, particularly at ambient temperatures during rolling, and moderate levels of plastic deformation (at which bauschinger effect can be neglected for the particular class of polymeric materials studied here), greatly simplifies the task of material modeling and allows us to deploy a computationally efficient, yet accurate, finite deformation rate-independent elastic-plastic material behavior model (with inclusion of isotropic-hardening) for analyzing the rolling of these polymeric films. The interfacial behavior between the roller and polymer surfaces is modeled using Coulombic friction; consistent with the rate-independent behavior. The finite deformation elastic-plastic material behavior based on (i) the additive decomposition of stretching tensor ($D = D^e + D^p$, i.e. a hypoelastic formulation) with incrementally objective time integration and, (ii) multiplicative decomposition of deformation gradient ($F = F^e F^p$) into elastic and plastic parts, are programmed and carried out for cold-rolling within ABAQUS Explicit. Predictions from both the formulations, i.e., hypoelastic and multiplicative decomposition, exhibit a close match. We find that no specialized hyperlastic/visco-plastic model is required to describe the behavior of the particular blend of polymeric films, under the conditions described here, thereby speeding up the computation process.

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for steady-state rolling predictions. Rolling simulations reveal that elasticity of the polymeric sheets, undergoing bulk plastic deformation at finite strains, gives rise to large rolling loads which cannot be captured by the classical rolling theories. Finite element predictions thus enable the development of the rolling-apparatus and particularly help achieve two key desirable features: (i) through-thickness (and homogeneous) plastic deformation of the polymer film-stack, and (ii) negligible unwanted deflections (less than 0.003 degrees in shaft-axis orientation at load levels of 4 kN, to avoid non-uniform compression of the films). The developed rolling apparatus (which comprises of symmetric ‘C’ shaped vertical roller-stands, appropriately-sized compression rollers, frame elements, shafts, bearings, drive systems, etc.) is able to exert appropriate levels of line-loading to cause deformation induced roll-bonding in time on the order of seconds. Finally, the experimentally observed rolling loads during actual operation are also compared and found to match closely with the proposed finite element analyses. Deformation aspects of solid-state polymeric sheets presented here are expected to facilitate the development of processes and rolling-apparatuses involving (or related to) cold roll-bonding of polymers.

Notation

- $a$: half contact width in rolling;
- $da$: differential area in current configuration;
- $B$: reference body;
- $b_0$: body force (per unit volume) in current configuration;
- $b$: body force (per unit volume) in current configuration with inertia effect included;
- $c$: dilatational wave speed;
- $C^e$: elastic right Cauchy–Green strain;
- $\mathbf{C}$: Fourth order elasticity tensor;
- $C^{ep}$: Elasto-plastic tangent modulus for hypoelasticity;
- $2d$: total compression in the film-stack;
- $dv$: differential volume in current configuration;
- $dv^*$: differential volume of current configuration with respect to a rotated frame;
- $dv_R$: differential volume in the reference configuration;
- $\mathbf{D}$: stretching-rate tensor, symmetric part of velocity gradient $\mathbf{L}$;
- $\mathbf{D}^e$: elastic stretching-rate tensor, symmetric part of $\mathbf{L}^e$;
- $\mathbf{D}^p$: plastic stretching-rate tensor, symmetric part of $\mathbf{L}^p$;
- $\bar{\mathbf{D}}^p$: plastic stretching-rate tensor in current configuration for hypoelasticity;
- $\dot{e}_i$: principal components of the spatial strain-rate with $i = 1, 2, 3$;
- $\delta \epsilon_i$: incremental strain in principal directions with $i = 1, 2, 3$;
- $\epsilon_z$: nominal strain in z-direction; $E$: Young’s modulus;
$E_{\text{roller}}$ Young’s modulus of roller;

$E_{\text{film}}$ Young’s modulus of polymer film;

$E_h$ Hencky strain given by $ln(U^r)$;

$E_i$ eigen values of Hencky strain given by $ln(\lambda^i)$ with $i = 1, 2, 3$;

$\Delta E$ spatial incremental strain in hypoelastic formulation;

$\Delta E^e$ spatial incremental elastic strain in hypoelastic formulation;

$\Delta E^p$ spatial incremental plastic strain in hypoelastic formulation;

$E_{\text{roller}}$ Young’s modulus of the rollers;

$\Delta \varepsilon_{\text{pl}}^s$ incremental equivalent plastic strain;

$\varepsilon_{\text{pl}}^s$ equivalent plastic strain;

$\varepsilon_{\text{pl}}^p$ accumulated plastic strain;

$f$ yield function for multiplicative plasticity;

$f_h$ yield function for hypoelasticity;

$\mathcal{F}$ vector of external nodal point forces for the finite element model;

$F$ deformation gradient;

$F^e$ elastic part of deformation gradient;

$F^p$ plastic part of deformation gradient;

$F_{\text{friction}}$ measured force during measurement of coefficient of friction;

$G$ elastic shear modulus;

$h_1$ thickness of film-stack at the inlet;

$h_0$ thickness of film-stack at the outlet;

$\bar{h}$ average of the inlet and outlet thicknesses ($h_1, h_o$);

$h$ thickness of film-stack (at a distance $x$ from origin) in the roller bite;

$h_i$ thickness of the stock at the inlet;

$H$ hardening modulus;

$I$ second order identity tensor;

$I$ fourth order identity tensor;

$\mathcal{I}$ vector of internal nodal point forces for finite element model;

$J$ determinant of the deformation gradient $F$;

$J^+$ determinant of deformation gradient $F^+$;

$J^+$ determinant of deformation gradient $F^+$;

$k$ yield strength in shear;

$K$ elastic bulk modulus;

$l_e$ smallest element dimension in finite element mesh;

$L$ total load in rolling;
L spatial velocity gradient;
L^e \text{ elastic part of velocity gradient; }
L^p \text{ plastic part of velocity gradient; }
\tilde{L}^e \text{ virtual elastic velocity gradient; }
\tilde{L}^p \text{ virtual plastic velocity gradient; }

m'g \text{ weight of the steel block used in friction measurement experiment; }
m \text{ velocity-dependent friction factor in range between 0 and 1; }
M \text{ moment per-unit width during rolling; }
M^e \text{ Mandell stress, work conjugate to } L^p ;
\mathcal{M} \text{ diagonal lumped mass matrix for global finite elements; }
n \text{ unit vector } n \text{ in the deformed configuration; }
N^p \text{ direction of plastic flow (determining } D^p \text{); }
\bar{N}^p \text{ direction of plastic flow transformed into the current configuration (determining } \bar{D}^p \text{); }
P \text{ material region in the reference configuration; }
P_t \text{ material region in the current configuration; }
p \text{ radial pressure due to roller; }
P_l \text{ line loading in rolling; }
\bar{p} \text{ mean normal pressure; }
q \text{ frictional traction due to roller; }
Q \text{ frame-rotation; }
r_i^e \text{ orthonormal eigen-vectors of } \mathbf{C}^e \text{ and } \mathbf{U}^e \text{ with } i = 1, 2, 3;
R \text{ radius of Roller; }
R_a \text{ surface roughness of rollers; }
R \text{ rotation tensor obtained by the decomposition } \mathbf{F} = \mathbf{R} \mathbf{U} ;
R^e \text{ rotation tensor obtained by the decomposition } \mathbf{F}^e = \mathbf{R}^e \mathbf{U}^e ;
R^p \text{ rotation tensor obtained by the decomposition } \mathbf{F}^p = \mathbf{R}^p \mathbf{U}^p ;
S^e \text{ microstress that is work conjugate to elastic velocity gradient } L^e ;
T \text{ Cauchy stress; }
T^e \text{ microstress that is work conjugate to plastic velocity gradient } L^p ;
T_g \text{ Glass transition temperature; }
t(n) \text{ Traction field (associated with } n \text{); }
T^J \text{ Jaumann rate of Cauchy stress; }
U_{x,rel} \text{ relative displacement of shaft center point with respect to base point in x-direction; }
U_{y,rel} \text{ relative displacement of shaft center point with respect to base point in y-direction; }
U \text{ symmetric and positive-definite stretch tensor tensor from right polar decomposition of } \mathbf{F} ;
symmetric and positive-definite right elastic stretch tensor from right polar decomposition of $F^e$; 
$U^e$ symmetric and positive-definite right plastic stretch tensor from right polar decomposition of $F^p$; 
$\dot{U}$ vector of nodal point velocities for the finite element model; 
$V_1$ incoming speed of the film-stack into the rollers; 
$V_2$ tangential speed of the rollers; 
$v_o$ exit speed of the film-stack; 
$v$ spatial velocity; 
$v_v$ virtual spatial velocity; 
$v_x$ velocity at any section (at a distance of $x$ units from the exit); 
$V$ symmetric and positive-definite stretch tensor tensor from left polar decomposition of $F$; 
$V^e$ symmetric and positive-definite elastic stretch tensor from left polar decomposition of $F^e$; 
$V^p$ symmetric and positive-definite plastic stretch tensor from left polar decomposition of $F^p$; 
$\mathcal{V}$ set of virtual velocity fields; 
w width of the film; 
$W$ continuum spin, the anti-symmetric part of $L$; 
$W^e$ elastic spin, the anti-symmetric part of $L^e$; 
$W^p$ plastic spin, the anti-symmetric part of $L^p$; 
x, $y$, $z$ coordinate variables; 
x position of a material point in current configuration; 
X position of a material of a body in reference configuration; 
$Y$ scalar yield-strength and radius of the spherical yield strength; 
$Y^'$ hardening modulus; 
$\tau_{\text{shear}}$ tangential traction due to friction; 
$\sigma_v$ equivalent (von Mises) stress; 
$\sigma_{y,\text{film}}$ yield-strength of film; 
$\sigma_{y,\text{roller}}$ yield-strength of roller; 
$\sigma_1, \sigma_2, \sigma_3$ principal stress components; 
$\sigma'_1, \sigma'_2, \sigma'_3$ deviatoric principal stress components; 
$\bar{T}_z$ average transverse stress in thin-strip plastic rolling; 
$T_x$ average longitudinal stress in thin-strip plastic rolling; 
$T_y$ average out-of-plane stress in thin-strip plastic rolling; 
$T_{xz}$ normal stress in x-direction; 
$T_{yz}$ normal stress in y-direction; 
$T_{xy}$ shear stress in x-y plane; 
$\Psi$ Nodal point displacements in the finite element solution;
\( \phi \) angular position of a differential element; 
\( \nu \) Poisson’s ratio; 
\( \tau \) duration of contact in rollers during rolling; 
\( \mu \) coefficient of dry friction; 
\( (\cdot) \) time rate of a quantity; 
\( ||(\cdot)|| \) norm of the quantity; 
\( (\cdot)'' \) double derivative w.r.t. time; 
\( A : B \) inner product of two second order tensors \( A \) and \( B \); 
\( \text{Sym}(\cdot) \) symmetric part of the operand; 
\( \nabla \) gradient with respect to material coordinates \( X \); 
\( (\cdot)^{-1} \) inverse of the tensor operand; 
\( (\cdot)^{\top} \) transpose of the tensor operand; 
\( \text{det}(\cdot) \) determinant of the tensor operand; 
\( \text{tr}(\cdot) \) trace of the tensor operand; 
\( \text{grad}(\cdot) \) gradient of the quantity with respect to spatial coordinates \( x \); 
\( (\cdot)^* \) transformed quantity in rotated-frame; 
\( \text{div}(\cdot) \) divergence of the quantity with respect to spatial coordinates \( x \); 
\( (\cdot)_0 \) deviatoric part of the tensor quantity; 
\( \nu \) Poisson’s ratio; 
\( \chi \) deformation, mapping a material point to a spatial point; 
\( \rho \) density at a material point in current configuration; 
\( \Psi(P_t) \) external power delivered to a body part \( P_t \) in current configuration; 
\( \mathcal{I}(P_t) \) internal power expended inside a body part \( P_t \) in current configuration; 
\( \mathcal{W}_{\text{int}} \) Virtual internal power; 
\( \mathcal{W}_{\text{ext}} \) Virtual external power; 
\( \omega \) angular velocity of rolls; 
\( \varphi \) free energy per-unit intermediate volume; 
\( \tilde{\varphi} \) free energy function expressed in terms of invariants of \( C^e \); 
\( \hat{\varphi} \) free energy function expressed in terms of invariants of \( U^e \); 
\( \delta \) dissipation per-unit intermediate volume; 
\( \Omega \) frame-spin; 
\( \lambda \) consistency parameter for the yield condition; 
\( \dot{\lambda} \) equivalent plastic strain rate; 
\( \lambda_i^e \) positive eigen values of \( U^e \) with \( i = 1, 2, 3 \); 
\( \omega_i^e \) positive eigen values of \( C^e \) with \( i = 1, 2, 3 \);
\(\psi\) angular deflection of the shaft;
\(\omega\) angular speed of the rollers.

1 Introduction

An ongoing activity at the NVS-MIT Center for Continuous Manufacturing (NVS-MITCCM) envisions continuous manufacturing of pharmaceutical tablets from thin polymeric films (approximately 100 microns in thickness). Such a manufacturing process is intended to operate in a minimum number of steps, and the final drug-product to be made from starting to completion in a continuous manner at one facility. This process involves the following steps which are to be operated in a continuous mode: (i) preparation of polymeric films through solvent casting, (ii) folding of the solvent-cast films, (iii) bonding of the folded film layers, and (iv) shaping (or cutting) of the tablets from the bonded film stock. Figure 1 schematically shows the key steps involved in this process. For a detailed account of this methodology see [90, 114]. These polymeric films are engineered to exhibit unique mechanical and physical properties, and can be bonded in the solid-state at ambient temperatures by subjecting them to active plastic deformation [92, 91], and thereby alleviating the need of adhesives, surface modifications, heat treatments, etc. In order to achieve bonding of these films in a continuous mode, a rolling-like operation is conceived as a natural option.

Several methods, usually referred to as calendering, are already in practice for continuously processing flat sheet-like materials such as paper, polymers, metals, blankets, cardboard laminates, etc. The central idea in calendering-like methods is to subject the incoming stock of material under compression through rollers with a desired processing step. For centuries the metal industries have also been producing flat products such as metal sheets, plates, strips, bars etc. through rolling processes. These processes fall in the category of hot and/or cold forming (indicating net change in the shape of input stock material), and the machinery to carry out these processes have traditionally been referred to as rolling mills. Examples of continuous processes for cold bonding and welding of metals or cladding processes can be found in [70, 23, 39, 125, 69]. Some applications related to cold-rolling of polymeric sheets, slabs, etc. can also be found in the literature: for e.g., studying the time-dependent evolution of micro-structural properties after plastic deformation during cold-rolling of polycarbonate (PC) and polystyrene (PS) [15], improving the tensile strength and toughness of certain thermoplastics by unidirectional (and biaxial) rolling [17, 19, 74], enhancing the yield strength for producing cold-forged products from the rolled-rods of HDPE, PP, POM, ABS and PVC [72], and analyzing the effect of tensile properties of ABS (with rubber inclusions) [10] and poly(butylene succinate) (PBS) based nanocomposites [94] after rolling. Even calendaring of thermoplastic materials above \(T_g\) has been studied [13]. However, a continuous process or a rolling-apparatus/process for bonding of polymeric-films in solid-state well below the glass-transition
temperature is a new application, and has never been reported nor attempted before. Given the rich history of rolling and calendering processes, it is worth asking the following question: if rolling mills are in existence for such a long time [15, 12, 122, 48, 111], can one not utilize an off-the-shelf rolling-mill for this application? and, why is there a need to really develop a new rolling-apparatus? Firstly, for decades the majority of rolling mills have been designed with their roller-axes in the horizontal orientation. Only a handful of rolling mills have been reported in the literature with vertically oriented roller-axes (or rolling-mill stands), and to our best knowledge no detailed analysis or evaluation of deflection of critical mechanical-parts during the operation of these vertically-oriented rolling mills has been made [53, 88, 52, 87, 73, 73, 98, 98]. Furthermore, the mechanics of material deformation in a rolling mill, resulting properties of the product, and machine deflections are closely tied to the machine specifications and material properties. Therefore, unless a realistic deflection analysis of the machine-parts (or rollers) is made for rolling the desired material, the overall performance of the rolling-apparatus will be questionable.

As shall become abundantly clear during the course of this study, the vertical orientation of roller-axes, the roll-gap adjustment mechanism in the vertical layout, and uniform and homogeneous compression of the incoming film-stack are key requirements for successful roll-bonding of polymeric films. We find no detailed or quantifiable investigation of these factors in the literature, particularly, for rolling solid-state polymeric films that require (homogeneous) through-thickness plastic deformation. Deflections occurring in any commercial rolling mill, obviously, will entirely depend on the material properties of each machine-element, part layouts, assembly details, etc. and unless they are provided by the vendor it is difficult to accurately predict them. Different mechanisms for adjusting the roll-gap and suggestions to minimize or counteract roller/shaft deflections during rolling operations are also available (for example [115]), however a thorough analysis on the deflections incurred in different parts of commercially available rolling-mills (particularly in the vertical orientation), to the degree that we seek, is not available. Availing a high capacity off-the-shelf commercial rolling mill (with a vertical stand) for this application may be possible, but it could very well be an overkill and not guaranteed to meet all our requirements, let alone the compromise on flexibility, modularity and adaptability of the rolling-apparatus if later modifications are desired. Therefore conceptualizing and building a rolling-apparatus from scratch, while taking into account key functional requirements of the new cold-roll-bonding process, is a reliable path. Such a study will enable practitioners to derive key principles and functionalities targeted towards this (or related) applications, and custom design rolling-apparatuses for a commercial production.

Drawing inspiration from the existing manufacturing technologies related to rolling, we propose a new scheme for roll-bonding polymers and a rolling-apparatus for the same. To summarize, the main requirements for such a rolling-apparatus (and process) are as follows:

(i) It should be able to exert sufficient loads, so as to induce through-thickness (and preferably
homogeneous) plastic deformation up to 20% (or more) on the incoming stack of polymeric films. The roller-axes should be oriented vertically (since the incoming films emerge from an upstream step with their initial thickness-wise dimension oriented in the horizontal direction, Figure 1).

(ii) The desired levels of plastic strains should be achieved in the incoming stack of folded films; being fed at linear rate of $5 - 30$ mm/min.

(iii) The rolled stock should be uniform in width and thickness, and free from defects (such as warping, wrinkles, undulations, etc.) during and after the rolling operation.

(iv) The rollers should provide sufficient traction and necessary drive for the incoming films.

This paper is structured as follows: Section 2 reviews several attempts in the literature that have analyzed rolling processes. In Section 3 we describe the proposed cold-roll-bonding of polymeric films and utilize a classical rolling model to study various deformation fields. This enables us to set up an appropriate material model for polymeric films during finite elastic-plastic deformation in rolling, and also the frictional model between rollers and films. Time integration of the material models in the finite element setting are also described. Section 4 describes our fabricated ‘C’ shaped vertical-stand roller assembly and utilizes elastic-plastic model of polymeric films during static compression. Detailed development of the rolling-apparatus is presented here and selection criteria for several mechanical components along with their fabrication and assembly details are also given. Section 5 discusses the performance of the rolling-apparatus and compares the experimentally observed rolling loads with classical theory and plane strain steady-state rolling finite element simulations. The finite element simulations reveal key features of the deformation of polymer films in the roller bite, and simulation-based predictions match with those reported experimentally. The accuracy of finite element simulations are also verified. Finally, conclusions and directions for future work are summarized in Section 6.

2 Related Work on Rolling

Over the course of the last century there has been a continual interest and extensive theoretical, experimental and computational efforts to study deformations in rolling processes for predictions of the forming loads and torques, and distribution of pressures along the arc of contact in the roller bite. Major efforts
have been spent in understanding the mechanics of deformation in rolling since it is critical for predictions of (i) material velocities, strain rates, and strains, (ii) temperatures and heat transfer, (iii) variation in material strength or the flow stress, and (iv) stresses, pressure, and energy. Once the mechanics of deformation is known then it is possible to derive how a desired geometry can be obtained by rolling, and what would be the expected properties and micro-structures of the final part. Since cold and hot rolling processes have been active areas of research, several excellent references are available. Some of these references are listed as follows: introductory level material in [62], books dealing with detailed treatment of traditional rolling processes [100, 101, 38, 121], design of rolling mills [37, 105], critical review of classical rolling theories [80], overview of numerical methods and non-linear finite element methods in rolling [61, 82].

Broadly speaking, the methods of analysis for rolling, both analytical and numerical, can be categorized into: (i) slab methods, (ii) slip line analysis, (iii) upper bound analysis and (iv) computational methods. We review these approaches next.

(a) Slab methods: The object of deformation is assumed to be divided into multiple slabs, and for each slab simplifying assumptions are made for the stress distributions. Then, for the resulting system the approximate equilibrium equations are solved with imposition of strain compatibility between the neighboring slabs and boundary tractions, and approximate loads and stress distributions are derived. The pioneering work on strip rolling dates back to Siebel and Leug [106, 107] and Kármán [117], where several simplifications and physical assumptions were employed in modeling the strip rolling via quasi-static equilibrium treatment. Specifically, homogeneous compression, constant yield strength, and kinetic friction between the roller and strip were assumed. Several mathematical approximations were also involved in solving the resulting equilibrium differential equation. In subsequent attempts, Kármán-like models were employed by Nadai (1939) [83], Tselikov (1939) [115] and Ekelund (1927) [32], where manageable analytical descriptions of the strip rolling were derived.

Orowan (1943) considered a more comprehensive treatment of strip rolling by discarding a majority of previously-employed physical assumptions and mathematical approximations [89]. In particular, homogeneous compression, constant yield strength, kinetic friction due to slipping between the roller and stock interface, etc. were not assumed like in earlier attempts. In this manner, a differential equation was set up, and in order to proceed towards a solution of this differential equation, without making mathematical approximations and in the absence of digital computers, Orowan resorted to a graphical scheme that could provide an exact solution based on his theory numerically. It should be noted that these formulations were based on the equilibrium differential equations and therefore represented the quasi-static and steady-state solution. Transient or dynamic effects were not included. Bland and Ford (1948) proposed an approximate theory based on Orowan’s model while accounting front and back tensions in the strip [13]. Later, an extensive experimental work, evaluation and comparison of earlier theories on
cold-rolling of metals was presented by Ford in [35], along with a new simpler yet accurate theory. Reasonable predictions were found to be made, by each of the theory, in majority of the experimental cases. Subsequently, several rolling theories were also studied and compared experimentally in [110, 36]. It is worth highlighting that earlier theories ignored elastic strains of the stock during rolling deformations, which can be considered as a reasonable approximation for rolling of metals, and therefore in spite of several assumptions involved in analytically modeling a flat-rolling operation, satisfactory predictions (in terms of ‘order of magnitude estimates for rolling loads’) could often be noted. In a noteworthy attempt, pressure distribution in rolling along the contact arc were measured experimentally [3], and comparisons were made with predictions based on earlier theories. Agreements were noted in certain circumstances, but there were also some notable exceptions. For recent methods and accurate measurements of pressure distributions, along with a critical review, see [43] and [113]. Summarily, the slab-methods have several limitations in terms of their prediction capabilities for general non-linear rolling problems (non-linear/time-dependent material behavior, arbitrary strains, complex frictional conditions, temperature effects, etc.), but under certain simplified circumstances they can show some usefulness. In this study, we shall utilize the predictions from rigid-plastic rolling analysis to guide our finite-element analysis set-up and rolling-apparatus.

(b) Slip-line theory: The slip-line field method is used when the deformation is assumed to be plane strain with rigid perfectly-plastic materials, i.e., materials with a constant yield strength and rate-independent behavior. Here, elasticity is not included and the loading has to be quasi-static. This approach utilizes the hyperbolic properties of the equilibrium equations and can describe inhomogeneous deformations. Construction of slip-line fields can predict the exact corresponding stress distribution, but in context to rolling they have not yielded satisfactory predictions; even for metal deformation. Apart from underlying assumptions in the slip-line method, a particular difficulty arises in incorporating a satisfactory frictional/traction condition between the stock and the roller, see [1]. [5], [24], [27]. Nowadays this approach has been largely superseded by the finite element modeling.

(c) Upper-bound method: According to this approach, first kinematically admissible velocity fields are assumed (while permitting for discontinuities) and the material is treated as rigid-plastic. Assumed velocity fields are derived from experimental evidence, or say experience. Then, a work-energy balance principle is employed for the deformation process at hand such that the externally applied load is expressed in terms of total energy expenditure during a forming process with the kinematically assumed velocity (or deformation field). Usually forging operations are analyzed by Hill’s method [45]. An upper bound approach to cold strip rolling was given by Avitzur [10]. Although strain-hardening can be included in this approach (unlike the slip-line method), yet accuracy in prediction relies on how closely the assumed velocity field matches the actual velocity field for a particular rolling operation. Owing to
these limitations, a complete and realistic description of deformation in rolling is rarely noted, though it may again serve as a quick means to make an estimation of the rolling loads.

(d) Finite Element Analysis: Finite Element Methods (FEM) have emerged as a powerful and useful computational tool for a variety of purposes. It is fair to say that nowadays, for most complex problems, where exact analytical solutions are not available, FEM offer a reliable route to arrive at accurate solutions for boundary value problems involving large-deformations and material non-linearity.

In the earliest FEM approaches for analyzing rolling (and other metal-forming operations), the material behavior was idealized to be rigid-perfectly plastic or visco-plastic, and ‘Eulerian’ flow-type formulations, with velocity field as an unknown, were adopted [22, 124]. Subsequently, several similar finite element procedures for rolling appeared [25, 125, 77, 54, 47] to estimate the stresses, deformation rates, forming loads in the plastic zone, roll deformation, etc. in steady state rolling. An approximate three-dimensional analysis for rolling, using Hill’s extremum procedure for rigid-plastic material, was done in [57].

Although satisfactory predictions could be noted with these approaches, but difficulties occurred for describing free boundaries, non steady-state behavior and elastic effects. Another source of challenge in the rigid-plastic material idealization arose from achieving the volume preserving constraint on the velocity field, i.e. plastic flow incompressiblity. This was largely attempted by use of the penalty based approaches or mixed formulations (using pressure and velocity fields), see [31], however only approximate results have been noted. Consideration of elastic effects at the entry and exit during rolling, within the Eulerian framework, was also done in [26, 112] by employing an ‘initial stress method’. In such approaches, the spatial strain-rate was expressed as sum of inelastic and elastic part and constitutively related to the deviatoric stress and material derivative of the stress, respectively.

A major challenge faced in earlier finite element analyses of rolling has been modeling of the interfacial friction between the roller and stock. While, in general, the coefficient of friction or friction factor is reported to depend on the relative velocity, interfacial pressure and surface roughness, a reliable model describing the relationship of these parameters to the frictional events has still not been provided. Often the coefficient of friction has been treated as a free parameter whose magnitude is determined by inverse calculations, i.e., by matching the experimental and simulation results. Hartley et al. [42] pointed out the importance of appropriately modeling the friction in numerical simulations and used a layer of elements at the contact surface (whose stiffness could be altered) to model friction; but usefulness of such an approach for any realistic or complex frictional behavior remained unclear. Other approaches to model frictional behavior included: (i) the use of a velocity-dependent friction factor \( m \) (which relates the interfacial frictional shear traction to the yield stress of the material as \( \tau_{\text{shear}} = mk \), where \( k \) is the shear yield strength and \( m \) lies between 0 and 1) by Kobayashi et. al [55], and (ii) a velocity-dependent
friction model by Zienkiewicz et al. (1978) [125]. In [64] Li and Kobayashi (1982) expressed the frictional shear stress at the roll/rolled metal contact as a function of the relative velocity. The shear stress, thus, depended on a friction factor, the relative velocity, the yield strength of the rolled metal, a coefficient of friction and two positive constants, defined for both the forward and the backward slip zones. They analyzed the problem of cold rolling of rigid-plastic strips between rigid work rolls and compared their predicted roll pressure distributions to the data of Al-Salehi et al. (1973) [3]. They observed discrepancies and attributed them to, among other factors, the manner in which friction was modeled.

Notable contributions on the development of ‘Total Lagrangian’ [44] and ‘Updated Lagrangian’ [75] formulations for the displacement based finite elements, addressing issues related to large deformations/rotations and material non-linearities, greatly facilitated the study of metal-forming problems in the subsequent era, and rolling in particular; see [66, 102, 60, 70, 29, 30]. The use of objective stress rate measure was widely noted for nonlinear problems, and plasticity was incorporated through additive decomposition of spatial strain-rate or multiplicative decomposition of the deformation gradient into the elastic and plastic parts. ‘Locking phenomenon’ exhibited by the displacement based finite elements due to volume preserving plastic flow constraint, particularly, in fully plastic regimes were avoided through special arrangements of the elements [84] or by usage of the selective reduced integration method [126]. Incorporation of realistic frictional interaction was made possible through development of robust computational contact algorithms [51] that employed penalty or Lagrange multiplier methods, and thereby largely overcoming the issues associated with earlier ad-hoc treatments for friction. Finally, developments such as arbitrary Lagrange Eulerian re-meshing, to over difficulties associated with excessive mesh distortion in fem, further aided the deployment of FEM for metal forming problems. For a detailed overview of the numerical solutions in context to rolling and forming methods can be found in [55, 103, 28].

Majority of the aforementioned finite element procedures were based on the incremental kinematics, and deployed (implicit) iterative schemes for solving the equilibrium/momentum equations. Methods based on explicit time integration of global equilibrium equations also gained popularity for a variety of metal forming applications [96, 50, 71]. Unlike implicit schemes, explicit integration of equations of motion is only conditionally stable and they have smaller memory footprint as storage of tangent stiffness matrix is not required. In explicit procedures, the critical time step for stability purposes is approximated as \( \frac{\ell_e}{c} \), where \( \ell_e \) is the smallest element size and \( c \) is the dilatational wave speed, i.e. it depends upon the fastest frequency of the discretized finite element mesh (which in turn depends on smallest element size, density and material moduli). This is also commonly known as the CFL limit. Explicit dynamics have been widely and successfully used for problems where inertial effects are negligible, and mass scaling is often adopted to increase the stable time increments. Alternately, for explicit time integration where stable time increments are small, effective sub-cycling procedures have been proposed according to which larger time steps can be taken [57]. In contrast, incremental time steps for implicit schemes are only
limited by the accuracy considerations and not constrained by any stability limit.

The choice of explicit method has proven to be useful for handling contact conditions (impenetrability and slip/stick), since contact tangent stiffness matrix, as required for implicit methods, is not needed during the solution procedure. Moreover the size of the stiffness matrix can change depending upon the contact state, and the stiffness matrix may lose symmetry due to friction. In presence of complex loading and contact conditions, the implicit methods can also face convergence difficulties. However, for simple contact conditions, such as those occurring in quasi-static rolling, methods of Lagrange multipliers and augmented Lagrangian can exactly satisfy the contact conditions [120]. For explicit methods, predictor-corrector or penalty based methods are commonly employed to handle contact.

In this work, we are interested in rolling of glassy polymers and it is well known that polymers exhibit a time-dependent visco-plastic mechanical behavior. Thus, quasi-static rolling simulations adopting a finite deformation time-dependent behavior, with sufficiently fine mesh, would require large physical computational times to arrive at a steady-state solution when carried out on standard computing platforms (i.e., without using sophisticated methods of GPUs or parallel computing etc.). However, thermoplastics are also known to exhibit rate-independent behavior at slow strain-rates and ambient temperatures [81, 6]. This is precisely the fact that we utilize for describing our polymer’s behavior – we first estimate the strain-rates associated with our rolling simulations at ambient temperatures and find them to be small. This allows us to work with a rate-independent material model. Based on earlier discussions, we choose to work with an explicit time stepping algorithm to simulate quasi-static rolling. For quasi-static rolling scenarios mass-scaling is used to speed-up the computations. The frictional interaction is also modeled using the rate-independent Coulombic friction and contact is handled through the kinematic-constraint algorithm.

To best of our knowledge, we find no work on cold-rolling/bonding of polymers, and resort to a commercial finite element software (ABAQUS) to set up a reasonable simulation model to make predictions for rolling-loads and deflections so that an apparatus can be designed cold-roll-bonding of polymeric films. The experimentally observed rolling loads also match those based on our prediction and thereby verify our procedure.

3 Mechanics and Modeling of Cold Roll-Bonding of Solid-State Polymeric Films

Figure 2 shows synthesis of polymeric films through solvent casting. The films contain a base polymer METHOCEL E15 and a compatible plasticizer PEG-400. METHOCEL is the trade name for

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1In our opinion, for rate-independent polymer deformation and simple contact conditions; as those occurring in this slow rolling process, implicit schemes could be equally useful. Newton Raphson type of scheme, with rate-independent material model, can give quadratic convergence during the equilibrium iterations and contact conditions can be satisfied exactly.
Hydroxypropyl-Methyl-cellulose (HPMC) and PEG-400 is polyethylene glycol with a molecular weight of 400. E15, PEG, ethanol and water were mixed in a weight ratio of 1.32 : 4.36 : 4.36 : 1, respectively, and a homogeneous solution was obtained through mixing with an electric stirrer for over 24 hours. After completion of the blending process, the solution was carefully stored in glass bottles at rest for 12 hours so as to get rid of any air bubbles. Solvent casting was carried out using a casting knife applicator from Elcometer on a Heat-resistant Borosilicate Glass. All steps were carried out in a chemical laboratory where ambient conditions of 20°C ± 2°C and R.H. 20% ± 5% were noted. After drying, amorphous and glassy polymeric films of E15 were obtained (containing 42.3% of PEG by weight). The glass transition temperature of the polymeric films was found to be 78 °C. As an illustration, Figure 4 shows bonding of multiple layers of films when subjected to plastic deformation in simple compression. The goal in cold-roll-bonding process is to achieve such bonding via inducing through-thickness plastic deformation. For further details on synthesis, preparation and characterization of polymeric films see [90, 92].

Figure 2: Solution making, film casting and peeling.

Figure 3: Bonding of polymeric films in compression due to bulk plastic deformation.

A schematic for the desired cold-roll bonding of multiple polymeric layers is shown in Figure 4. A key requirement of the rolling-apparatus is to apply sufficient levels of loads to induce through-thickness plastic deformation in the incoming stack of polymer films. In other words, the rolling apparatus should achieve desired levels of plastic-strain on the stack of incoming films over the interval of time (τ) spent
Figure 4: Schematic of a rolling scheme, where multiple layers are fed through rollers and bonded via induced plastic deformation.

under the compression rollers. The time ($\tau$) is set by size of the rollers and the specified production rates at which the films are fed. For the sake of illustration, a stack of polymer films (initially non-bonded sheets with a total thickness of $h_1$) is fed through the rollers, and shown to undergo net reduction in thickness. During active plastic deformation in the roller bite, the polymer molecules interpenetrate across respective film interfaces and cause bonding. If the stack thickness is small compared to the roller radius (R), i.e., $h << R$, an element in the roller bite will experience almost homogeneous straining through its thickness and negligible shear stresses will be developed between the film interfaces. On the other hand, if there is a non-uniform strain in the thickness direction then the interfaces (other than the symmetry plane of the film-stack in thickness direction) will have a tendency to exhibit tangential relative motion which hinders the molecular interpenetration of polymer chains and diminishes bonding.

In an extreme limit if the film-stack is quite thick compared to the radius of the rollers, then no through-thickness (plastic) deformation will incur and only local indentation will occur during roller compression. The current deformation induced bonding is a multi-scale process, since polymer mobility and interpenetration occurs at a molecular scale when plastic deformation occurs at a macroscopic continuum scale, thus predicting the exact role of strain gradient in the thickness direction on bonding is all-together another detailed modeling task and beyond the scope of this current work. In order to achieve homogeneous through-thickness plastic strain, rollers of radius $R$ such that $R/h \gg 1$, can be considered sufficiently large. We shall verify that under this assumption with finite element simulations and show that under such circumstances shear stress can be neglected and rolling loads are accurately predicted.

The polymer films of interest have a yield-strength ($\sigma_{y,films}$) $\sim$ 4 – 6 MPa and modulus ($E_{film}$) $\sim$ 50 – 200 MPa. Stainless steel rollers, with a typical yield-strength $\sigma_{y,roller}$ $\sim$ 600 MPa and a modulus $E_{roller}$ $\sim$ 180 – 200 GPa, can be employed and essentially treated as rigid in comparison to these ductile polymer films. Thus flattening of rollers, which may be an issue in context of rolling of metals, is unlikely.
here. Rotatory motion to the rollers can be imparted in a variety of manner, but the rollers and drive mechanism for our application need to have a vertical-axis orientation. Additionally, the roller-assembly should have sufficient flexibility to impose a desired level of plastic thickness reduction (typically in the range of 10-20%) and therefore the gap between the rollers should be adjustable. Now we resort to an elementary analytical model based on rigid-plastic analysis to estimate various fields associated with rolling deformation and accordingly make simplifying assumptions to carry out the analysis through finite element simulations.

3.1 Analysis Based on Rigid Perfectly-Plastic Material Model

According to rigid-plastic rolling \[49\], the estimates of line-loading \(P_l\) and torque \(M\) (per-unit width of the strip) are given as follows:

\[
\frac{P_l}{\sigma_{y, film}/\sqrt{6}a} = 2 + \frac{a}{\bar{h}} \left( \frac{1}{2} - \frac{1}{3} \frac{a}{R} \right); \quad (1)
\]

\[
\frac{M}{\sigma_{y, film}/\sqrt{6}a^2} = 1 + \frac{a}{4h} \left( 1 - \frac{a}{R} \right). \quad (2)
\]

In the above equations, \(\sigma_{y, film}\) is taken to be the tensile yield strength of the material, \(a\) is the half contact width, \(\bar{h} = \frac{h_1 + h_o}{2}\) is the mean film thickness during rolling, and \(R\) is the radius of the rollers. The detailed features of the theory and derivation of a rigid-plastic rolling scheme are given in the Appendix Section \[8.1\] and the rigid-plastic rolling scheme is shown in Figure \[5\]. Good bonding is expected if there is a uniform compression of the film-stack across along the width direction (normal to the plane of Figure \[5\]). Therefore, the parallel between the rollers in the final assembly is critical. For a stack thickness of 1 mm, an accuracy in roller-parallel during compression up to 0.02 mm (corresponding to a 2.0% thickness-reduction), over the stack width, can be regarded as sufficiently good for bonding purposes and serves as a guideline for our rolling-apparatus \[2\].

From the geometry of deformation, if we assume that there is no slippage or sliding between the rolled-stock and the rollers at the exit, then the exit velocity \(v_o\) of the rolled-stock is equal to tangential velocity of the rollers at the exit \(V_2 = R\omega\), i.e., \(v_o = V_2\). In this rolling scheme, the time of compression \(\tau\) under the rollers can be estimated as:

\[
\tau = \frac{\sqrt{R(h_1 - h_o)}}{V_2}. \quad (3)
\]

For the purpose of illustration, we choose \(\sigma_{y, film} = 6.0 \text{ MPa}, h_1 = 0.6 \text{ mm}, h_o = 0.48 \text{ mm}\), so that \(2d = (h_1 - h_o) = 0.12 \text{ mm}\) (indicating 20% plastic compression). If \(R\) is chosen to be 100 mm then \(a(= \sqrt{Rd}) = 2.45 \text{ mm}\) (see Section \[8.1\] for derivation of various quantities). Now substituting these

\[2\]In a later section we report that the standard deviation in thickness of roll-bonded stacks, through our apparatus, is less than 2.0% of initial thickness.
values in equation 1, we estimate \( P_l = 16.7 \times 10^3 \text{ N/m} \). If we assume the width \((w)\) of the strip to be 10 mm then the total compressive load \( L = P_l w \) is approximately 167.8 N. The moment per unit width \((M)\) according to equation 2 is 20.492 N, and therefore the total torque for 10 mm wide strip is 0.204 Nm. For the outlet speed 10 mm/min, the residence time \( \tau \) for this case is 20.78 seconds. The rigid plastic analysis suggests that it is possible to achieve active plastic deformation in seconds with rollers of size \( R = 100 \text{ mm} \).

In accordance with the rigid-plastic model, we can also roughly estimate the variation of strain-rate and strain in the rolling-bite under assumption of homogeneous compression (and neglecting shearing). The nominal compressive strain in the z-principal (thickness reduction or transverse) direction is given as

\[
e_z = (1 - h/h_i)
\]

where \( h_i \) is the thickness of the stock at the inlet, and \( h \) can be approximated from the roll profile (see equation 197 Appendix Section 8.1) as \( h = h_o + R\phi^2 \approx h_o + \frac{a^2}{R} \). The corresponding nominal strain rate, in the roller bite, is given as

\[
|\dot{e}_z| = \frac{1}{h_i} \frac{dh}{dt}
\]
and, from the conversation of mass we can write

\[ v_x h = v_o h_o \]  \hfill (6)

where, \( v_x \) and \( v_o \) represent the velocities at any section (at a distance of \( x \) units from the exit) and at the exit, respectively. Similarly, \( h \) and \( h_o \) are the height at any section (at a distance \( x \) from the origin) and at the exit, respectively. Using the above equations, we can re-write the expression for strain rate as

\[ |\varepsilon'_{x}| = \frac{2xh_o v_o}{R h_i h} \]  \hfill (7)

By choosing \( R = 100 \) cm, \( h_1 = 0.6 \) mm, \( h_o = 0.54 \) mm (indicating 10% nominal strain in thickness reduction), and \( v_o \approx 10.0 \) mm/min, and utilizing the above expressions, plots of strain rate and strain in the roller-bite as a function of distance from the exit (considered to be origin), are shown in Figures 6 and 7 respectively. The time spent in the roller bite according to equation 3 is 14.69 seconds.

Since we have employed the model of rigid-plastic rolling in the above calculations, the strain-rates predicted here are overestimates (as we have neglected the elastic deformation) of the actual strain-rates that would occur. The average nominal strain-rate in the roller bite for this case can be estimated as 0.10/16.69=0.006 sec\(^{-1}\), which itself is small. It is important to note that the over-estimated strain rates in the roller bite are themselves not very large, and indicate that deformation in the rollers occurs at slow rates. This is a useful observation because solid-state polymers, when deformed at slow to moderate rates, show small rate-sensitivity in their mechanical response and, this simplifies the task by enabling us to use a rate-independent material model in the finite element simulations. As pointed out in [6], at strain-rates less than 0.01, hardly any rate sensitivity is noted. Moreover, even the rate-dependent models at slow deformation rates reduce to their rate-independent counterparts. If strain-rate effects are
significant, such as in calendering of elastic-viscous materials [93], then appropriate material behavior models must be considered. At a molecular level, well below the $T_g$, the resistance to plastic flow in polymers is mostly dominated by the resistance to molecular level re-orientations. The backstress due to entropic chain alignment is also dormant well below $T_g$, especially when only moderately large deformation regimes are considered. Thus, effects due to network elasticity of the polymer matrix can be ignored altogether. The plastic flow can be modeled using $J^2$ flow criterion and normality flow rule. Pressure sensitivity can also be ignored well below $T_g$ for stress states occurring in rolling. For comprehensive details on mechanical behavior of polymers see [8]. For popular sophisticated polymer models, see for e.g., [14, 7, 9]. Finally, we also take note of the fact that polymers can exhibit noticeable elastic deformations which cannot be ignored with respect to plastic strains, and our experimental results indeed reveal this behavior.

### 3.2 Finite Strain Rate-Independent Elastic-Plastic Material Model for Polymeric Films

Based on the discussions presented in the previous section, we now work towards deriving a rate-independent deformation model to describe the polymer behavior at finite strains. We present two approaches to model this behavior, along with the necessary preliminaries, based on: (a) multiplicative decomposition of deformation gradient into elastic and plastic parts, and (b) hypoelastic formulation that employs an objective measure of stress-rate and utilizes additive decomposition of spatial strain-rate into elastic and plastic parts.
Kinematics

A homogeneous body $B$ is identified with the region of space that it occupies with respect to a fixed reference configuration. $X$ denotes an arbitrary material point of $B$. The motion of $B$ is then described as a smooth one-to-one mapping $x = \chi(X, t)$, and the deformation gradient is defined as

$$F = \nabla \chi = \frac{\partial \chi}{\partial X}. \quad (8)$$

The velocity gradient $L$, i.e., gradient of the spatial velocity with respect to spatial coordinates, is related to the deformation gradient $F$ through the identity

$$L = \text{grad} \dot{\chi} = \dot{F}F^{-1}. \quad (9)$$

The velocity gradient $L$ can be decomposed into the symmetric part $D$ and anti-symmetric part $W$ as

$$D = \frac{1}{2}[L + L^\top], \quad (10)$$

and

$$W = \frac{1}{2}[L - L^\top]. \quad (11)$$

$D$ is the stretching rate tensor and $W$ is the continuum spin. Within the framework of large defor-
mations, the multiplicative decomposition \[59\] of the deformation gradient \( \mathbf{F} \) can be written as

\[
\mathbf{F} = \mathbf{F}^e \mathbf{F}^p. \tag{12}
\]

where \( \mathbf{F}^e \) represents the local elastic deformation deformation of material in an infinitesimal neighborhood of \( \mathbf{X} \) due to stretch and rotation. \( \mathbf{F}^p \) is the plastic distortion and represents the local deformation of material \( \mathbf{X} \) in an infinitesimal neighborhood due to the irreversible/plastic deformation. By substituting \( \mathbf{F} \) from equation 12 into equation 9, we can obtain the following expression for \( \mathbf{L} \):

\[
\mathbf{L} = \dot{\mathbf{F}}^e \mathbf{F}^e^{-1} + \mathbf{F}^e \dot{\mathbf{F}}^p \mathbf{F}^p \mathbf{F}^e^{-1}. \tag{13}
\]

We define the elastic and plastic velocity gradients, \( \mathbf{L}^e \) and \( \mathbf{L}^p \), as follows

\[
\mathbf{L}^e = \dot{\mathbf{F}}^e \mathbf{F}^e^{-1}, \tag{14}
\]

and

\[
\mathbf{L}^p = \dot{\mathbf{F}}^p \mathbf{F}^p^{-1}. \tag{15}
\]

Using equations 14 and 15 we can rewrite equation 13 as

\[
\mathbf{L} = \mathbf{L}^e + \mathbf{F}^e \mathbf{L}^p \mathbf{F}^e^{-1}. \tag{16}
\]

Now the elastic stretching \( \mathbf{D}^e \) and the elastic spin \( \mathbf{W}^e \) are defined as

\[
\mathbf{D}^e = \frac{1}{2} \left[ \mathbf{L}^e + \mathbf{L}^e^T \right], \tag{17}
\]

and

\[
\mathbf{W}^e = \frac{1}{2} \left[ \mathbf{L}^e - \mathbf{L}^e^T \right]. \tag{18}
\]

Similarly, the plastic stretching \( \mathbf{D}^p \) and the plastic spin \( \mathbf{W}^p \) are defined as

\[
\mathbf{D}^p = \frac{1}{2} \left[ \mathbf{L}^p + \mathbf{L}^p^T \right], \tag{19}
\]

and

\[
\mathbf{W}^p = \frac{1}{2} \left[ \mathbf{L}^p - \mathbf{L}^p^T \right]. \tag{20}
\]

We shall utilize two commonly employed kinematical assumptions concerning the plastic flow. First, the plastic flow is incompressible, i.e., plastic flow does not induce changes in volume. This is achieved
by assuming that $L^p$ and hence $D^p$ are deviatoric, i.e., $\text{tr}(L^p)=\text{tr}(D^p)=0$. This can be expressed as:

$$J^p = \det F^p = 1 \quad \text{and} \quad \text{tr}L^p = 0. \quad (21)$$

We can also decompose $J_\text{p}=\det F^p$ into elastic and plastic components as $J = \det F^e \det F^p$. Using $J^p = \det F^p$ and $J^p = \det F^p = 1$, we have $J = J^e J^p = J^p$. Secondly, if we idealize the material to be isotropic, then plastic flow can be assumed to be irrotational [41], i.e.,

$$W^p = 0. \quad (22)$$

Accordingly, $L^p$ is symmetric and

$$D^p = L^p. \quad (23)$$

From equation [15] we can write

$$D^p = L^p = \dot{F}^p F^p - 1 \quad (24)$$

Rearranging the terms in equation [24] we get

$$\dot{F}^p = D^p F^p. \quad (25)$$

The deformation gradient $F$ admits a right and left polar decomposition given as

$$F = RU = VR. \quad (26)$$

The polar decomposition for the elastic and plastic part of deformation gradient can also be done as

$$F^e = R^e U^e = V^e R^e, \quad (27)$$

and

$$F^p = R^p U^p = V^p R^p. \quad (28)$$

In what follows, we assume that the plastic part of the deformation gradient, i.e., $F^p$ (and therefore $L^p$) is invariant under frame transformation. This is a common approach [2] [11] and simply implies that the fictitious intermediate configuration, which is obtained by elastic destressing, of a material point neighborhood, is invariant. $F^p$ is a point wise map of each material point neighborhood from the reference configuration to the intermediate configuration (due to plastic distortion alone), and its invariance under the change of the observer/reference can be assumed without the loss of generality; granted that the
remaining kinematics of deformation is then described consistently under this assumption.

3.3 Rate-Independent Deformation Model Based on Multiplicative Decomposition of the Deformation Gradient

Multiplicative decomposition of deformation gradient into elastic and plastic parts was first introduced in [56] and [59], and has gained wide popularity in modeling constitutive responses of variety of materials. In this section we summarize a deformation model based on the multiplicative decomposition, list the constitutive equations and also provide the time integration algorithm.

Consider B to be the reference body. \( P \) be an arbitrary part of the reference body which is represented by \( P_t \) at any time \( t \). \( n \) is the outward unit normal on the boundary of \( P_t \). Let the traction field be represented by \( t \) (with the associated unit vector \( n \)) and the body force by \( b_0 \). Accordingly, the body force including the effect of intertia can be written as

\[
b = b_0 - \rho \ddot{\chi}.
\]  
(29)

Both \( t \) and \( b \) expend power over the velocity \( \dot{\chi} \), and the rate of external work can be written as

\[
\mathcal{W}(P_t) = \int_{\partial P_t} t(n) \cdot \dot{\chi} \, da + \int_{P_t} b \cdot \dot{\chi} \, dv.
\]  
(30)

Now we assume that the internal power inside the body is expended against two internal kinematical processes during deformation, elastic and plastic, and corresponding “micro-stresses” arising due to these processes are:

- An elastic stress \( S^e \) (defined in the current configuration) that is power-conjugate to \( L^e \). Thus, the power expended per unit volume of the current configuration is \( S^e : L^e \).

- A plastic stress \( T^p \) (defined in the intermediate configuration) which is power-conjugate to \( L^p \). Thus, the power expended per unit volume in the intermediate configuration is \( T^p : L^p \). Since \( L^p \) is deviatoric, we assume that \( T^p \) is also deviatoric.

We note that the fictitious intermediate configuration and the reference configuration have same volume (due to plastic incompressibility). Thus, total internal power expended in the body can be expressed as a volume integral over the current configuration as

\[
\mathcal{I}(P_t) = \int_{P_t} (S^e : L^e + J^{-1} T^p : L^p) \, dv.
\]  
(31)

3.3.1 Principal of virtual power

Let us consider that at any given fixed time, the deformation fields \( \chi \) and \( F^e \) (hence \( F \) and \( F^p \)) are given. Now the virtual velocities corresponding to the fields \( v \), \( L^e \) and \( L^p \) be represented as the set
\( \mathcal{V} = (\mathbf{v}, \mathbf{L}^e, \mathbf{L}^p) \). According to equation 16, the kinematically admissible virtual quantities must satisfy the following constraint

\[
\text{grad} \mathbf{v} = \mathbf{L}^e + \mathbf{F}^p \mathbf{L}^p \mathbf{F}^p^{-1}.
\] (32)

Then, according to equations 30 and 31 we can write the external virtual power as:

\[
\Psi(P_t, \mathcal{V}) = \int_{\partial P_t} \mathbf{t}(\mathbf{n}) \mathbf{\tilde{v}} d\mathbf{a} + \int_{P_t} \mathbf{b} \mathbf{\tilde{v}} d\mathbf{v},
\] (33)

and the internal virtual work as:

\[
\mathcal{J}(P_t, \mathcal{V}) = \int_{P_t} (\mathbf{S}^e : \mathbf{\tilde{L}}^e + \mathbf{J}^{-1} \mathbf{T}^p : \mathbf{L}^p) d\mathbf{v}.
\] (34)

According to the principle of virtual work, given any part \( P \), \( \Psi(P_t, \mathcal{V}) = \mathcal{J}(P_t, \mathcal{V}) \) for all generalized and kinematically admissible virtual velocities \( \mathcal{V} \), i.e.

\[
\int_{\partial P_t} \mathbf{t}(\mathbf{n}) \mathbf{\tilde{v}} d\mathbf{a} + \int_{P_t} \mathbf{b} \mathbf{\tilde{v}} d\mathbf{v} = \int_{P_t} (\mathbf{S}^e : \mathbf{\tilde{L}}^e + \mathbf{J}^{-1} \mathbf{T}^p : \mathbf{L}^p) d\mathbf{v}.
\] (35)

In other words, the assumption (or choice) of work conjugate stress measures \( \mathbf{S}^e \) and \( \mathbf{T}^p \) is justified, if and only if, they satisfy equation 35 for all kinematically admissible virtual velocities \( \mathcal{V} \), i.e.

3.3.2 Consequences of frame indifference

Based on the physical grounds, we require that the internal (virtual) work \( \mathcal{J}(P_t, \mathcal{V}) \) (equation 34), which is a scalar quantity, must be invariant under a change of reference frame. Given a change in frame, if \( P_t^* \) and \( \mathcal{J}^*(P_t^*, \mathcal{V}^*) \) represent the region and the internal work in the new frame, then the invariance of the internal work requires

\[
\mathcal{J}(P_t, \mathcal{V}) = \mathcal{J}^*(P_t^*, \mathcal{V}^*),
\] (36)

where \( \mathcal{V}^* \) is the kinematically admissible virtual velocity in the new frame. Also transformation of the virtual fields upon the change of reference follows identically their nonvirtual counterparts. Further from standard transformation laws under change of reference frame, we can write

\[
\mathbf{\tilde{L}}^e = \mathbf{Q} \mathbf{\tilde{L}}^e \mathbf{Q}^\top + \mathbf{\Omega},
\] (37)

where \( \mathbf{\tilde{L}}^e \) is the elastic distortion rate in the new frame, \( \mathbf{Q} \) is the frame-rotation, and \( \mathbf{\Omega} = \dot{\mathbf{Q}} \mathbf{Q}^\top \) is the frame-spin. Since, we have taken \( \mathbf{F}^p \) to be frame invariant under change of frame, thus, according to equation 15 \( \mathbf{L}^p \) is frame invariant, and so is \( \mathbf{\tilde{L}}^p \).
If we consider a virtual field with $\tilde{L}^p = 0$, then the invariance of internal virtual work equations \[34\] and \[36\] reduces to:

$$
\int_{P_t} (S^e : \tilde{L}^e) dv = \int_{P_t^*} (S^{e*} : \tilde{L}^{e*}) dv^*,
$$

(38)

i.e. internal virtual work due to $S^e$ must be invariant. In equation \[38\], $dv = dv^*$ (since differential volume is unaffected upon change of reference), the integral on the right hand side can be transformed over $P_t$, and since $P_t$ is arbitrary the following should be satisfied pointwise:

$$
S^e : \tilde{L}^e = S^{e*} : \tilde{L}^{e*}.
$$

(39)

Substituting equation $\tilde{L}^{e*}$ from \[37\] into the right hand side of equation \[39\] we obtain

$$
S^e : \tilde{L}^e = S^{e*} : QL^e Q^\top + S^{e*} : \Omega.
$$

(40)

If we choose a frame whose spin $\Omega$ is zero then above equation reduces to

$$
S^e : \tilde{L}^e = S^{e*} : QL^e Q^\top,
$$

(41)

or

$$
S^e : \tilde{L}^e = Q^\top S^{e*} Q : \tilde{L}^e.
$$

(42)

Since equation \[42\] must hold true for all kinematically admissible $\tilde{L}^e$, we conclude that

$$
S^{e*} = QS^e Q^\top.
$$

(43)

Further, if we choose a rotating frame with $Q = I$, i.e., a frame that instantaneously coincides with the reference frame but with a non-zero spin then $S^{e*} = S^e$ at that instant, and equation \[41\] reduces to

$$
S^e : \Omega = 0.
$$

(44)

Since $\Omega$ is arbitrary and skew, therefore equation \[44\] is satisfied for all $\Omega$, if and only if, $S^e = S^{e\top}$. So far we utilized the fact that internal virtual work due to elastic stretching must be invariant by choosing $\tilde{L}^p = 0$, which is justified in an elastic-plastic constitutive model if one imagines deformation processes in the elastic regime only. The invariance of elastic part of the internal virtual work implies that plastic part of the internal virtual work must also be invariant, and since $\tilde{L}^p$ is invariant so should $T^p$ be.
3.3.3 Macroscopic force balance

We again consider a macroscopic virtual velocity \( \mathbf{\bar{v}} \) for which \( \mathbf{\bar{v}} \) is arbitrary and \( \mathbf{\bar{L}} = \text{grad} \mathbf{\bar{v}} \), with \( \mathbf{\bar{L}} = 0 \). By substituting \( \mathbf{\bar{L}} = 0 \) in equation 34 and equating equations 33 and 34 we obtain

\[
\int_{\partial P_t} \mathbf{t}(\mathbf{n}) \cdot \mathbf{\bar{v}} d\mathbf{a} + \int_{P_t} \mathbf{b} \cdot \mathbf{\bar{v}} d\mathbf{v} = \int_{P_t} (\mathbf{S}^e : \mathbf{\bar{L}}) d\mathbf{v}. \tag{45}
\]

By using divergence theorem, we can rewrite the right hand term of the equation 45 as

\[
\int_{P_t} (\mathbf{S}^e : \mathbf{\bar{L}}) d\mathbf{v} = -\int_{P_t} \text{div} \mathbf{S}^e \cdot \mathbf{\bar{v}} d\mathbf{v} + \int_{\partial P_t} (\mathbf{S}^e \cdot \mathbf{n}) \mathbf{\bar{v}} d\mathbf{a}. \tag{46}
\]

Therefore, equation 45 becomes

\[
\int_{\partial P_t} \mathbf{t}(\mathbf{n}) \cdot \mathbf{\bar{v}} d\mathbf{a} + \int_{P_t} \mathbf{b} \cdot \mathbf{\bar{v}} d\mathbf{v} = -\int_{P_t} \text{div} \mathbf{S}^e \cdot \mathbf{\bar{v}} d\mathbf{v} + \int_{\partial P_t} (\mathbf{S}^e \cdot \mathbf{n}) \mathbf{\bar{v}} d\mathbf{a}, \tag{47}
\]

upon re-writing

\[
\int_{\partial P_t} (\mathbf{t}(\mathbf{n}) - \mathbf{S}^e \cdot \mathbf{n}) \cdot \mathbf{\bar{v}} d\mathbf{a} + \int_{P_t} (\text{div} \mathbf{S}^e + \mathbf{b}) \cdot \mathbf{\bar{v}} d\mathbf{v} = 0. \tag{48}
\]

Since, equation 48 must hold for all \( P \) and all \( \mathbf{\bar{v}} \), the traction condition yields to

\[
\mathbf{t}(\mathbf{n}) = \mathbf{S}^e \mathbf{n}, \tag{49}
\]

and the local force balance

\[
\text{div} \mathbf{S}^e + \mathbf{b} = 0. \tag{50}
\]

This traction condition and the force balance and the symmetry and frame-indifference of \( \mathbf{S}^e \) are classical conditions satisfied by the Cauchy stress \( \mathbf{T} \), an observation that allow us to write

\[
\mathbf{T} \overset{\text{def}}{=} \mathbf{S}^e \tag{51}
\]

and to view

\[
\mathbf{T} = \mathbf{T}^\top \tag{52}
\]

as the macroscopic stress and as the local macroscopic force balance. Granted that we are working in an inertial frame, so that equation 50 reduces to the local balance law for linear momentum

\[
\text{div} \mathbf{T} + \mathbf{b}_0 = \rho \mathbf{\dot{v}} \tag{53}
\]

where \( \mathbf{b}_0 \) is the non-inertial body force.
3.3.4 Microscopic force balance

Now we assume virtual fields $\mathcal{V}$ such that $\tilde{\mathcal{V}} = 0$. Then according to equation 32 we have

$$
\tilde{\mathbf{L}}^e = -\mathbf{F}^e \tilde{\mathbf{L}}^p \mathbf{F}^{e^{-1}}.
$$

(54)

Similarly, equation 35 reduces to

$$
\int_{P_1} (\mathbf{T} : \tilde{\mathbf{L}}^e + J^{-1} \mathbf{T}^p : \tilde{\mathbf{L}}^p) dv = 0.
$$

(55)

for all $P_1$. This implies that quantity inside the integral is identically zero, i.e.,

$$
J^{-1} \mathbf{T}^p : \tilde{\mathbf{L}}^p = -\mathbf{T} : \tilde{\mathbf{L}}^e.
$$

(56)

Now substituting $\tilde{\mathbf{L}}^e$ from equation 54 into equation 56 we obtain

$$
J^{-1} \mathbf{T}^p : \tilde{\mathbf{L}}^p = \mathbf{T} : (\mathbf{F}^e \tilde{\mathbf{L}}^p \mathbf{F}^{-1} - \mathbf{T}^e).
$$

(57)

By performing some algebraic manipulation and utilizing the fact that $\tilde{\mathbf{L}}^p$ is deviatoric and $\mathbf{T}$ is symmetric, we find that

$$
J^{-1} \mathbf{T}^p : \tilde{\mathbf{L}}^p = (\mathbf{F}^e \mathbf{T}_0 \mathbf{F}^{e^{-1}}) : \tilde{\mathbf{L}}^p.
$$

(58)

Since $\tilde{\mathbf{L}}^p$ is arbitrary, the microscopic force balance leads to

$$
J\mathbf{F}^e \mathbf{T}_0 \mathbf{F}^{e^{-1}} = \mathbf{T}^p.
$$

(59)

Next, we define the Mandel stress as

$$
\mathbf{M}^e = \int_{P_1} \mathbf{F}^e \mathbf{T}_0 \mathbf{F}^{e^{-1}} dv.
$$

(60)

We note that

$$
\mathbf{T}^p = \mathbf{M}_0^e.
$$

(61)

3.3.5 Rate-independent elastic-plastic constitutive response

We denote the free energy per unit volume of the intermediate configuration as $\varphi$, and make a constitutive hypothesis that it only depends on the elastic part of the deformation, $\mathbf{F}^e$. Further, if we invoke the requirement of the frame invariance of free energy (which it must satisfy due to being a scalar quantity), then we can deduce
\[ \varphi = \dot{\varphi}(C^e). \]  

(62)

Following the standard and well accepted principle of free-energy imbalance in solids [21], during the deformation, where \( \mathcal{W}(P_t) = \mathcal{F}(P_t) \), we require that

\[ \int_{P_t} \dot{\varphi} J^{-1} dv - \int_{P_t} \left( T : L^e + J^{-1} T^p : L^p \right) dv \leq 0. \]  

(63)

Since \( P_t \) is arbitrary the above equation implies

\[ \dot{\varphi} = J T : L^e - T^p : L^p = -\delta \leq 0, \]  

(64)

where \( \delta \) is the dissipation magnitude. We note that \( T \) is symmetric, thus

\[ T : L^e = T : D^e. \]  

(65)

By defining

\[ T^e \overset{def}{=} JF^{e-1} T F^{e-\top}, \]  

(66)

we can show that

\[ J T : D^e = \frac{1}{2} T^e : \dot{C}^e. \]  

(67)

Also, using the definition of \( M^e \) from equation 60 and that of \( T^e \) from equation 66 we obtain

\[ M^e = C^e T^e. \]  

(68)

Using equation 62 we can write

\[ \dot{\varphi} = \frac{\partial \dot{\varphi}(C^e)}{\partial C^e} : \dot{C}^e \]  

(69)

and, now using equations 65, 67 and 69 in equation 64 the free-energy imbalance can be written as

\[ \left[ \frac{1}{2} T^e - \frac{\partial \dot{\varphi}(C^e)}{\partial C^e} \right] : C^e + T^p : L^p \geq 0. \]  

(70)

If we consider kinematical processes in which \( L^p = 0 \) and

\[ \dot{C}^e = -\left[ \frac{1}{2} T^e - \frac{\partial \dot{\varphi}(C^e)}{\partial C^e} \right], \]  

(71)

then equation 70 reduces to
\[ -\left[ \frac{1}{2} \mathbf{T}^e - \frac{\partial \hat{\varphi} (\mathbf{C}^e)}{\partial \mathbf{C}^e} \right] : \left[ \frac{1}{2} \mathbf{T}^e - \frac{\partial \hat{\varphi} (\mathbf{C}^e)}{\partial \mathbf{C}^e} \right] \geq 0. \tag{72} \]

The only possible way to satisfy equation 72 for arbitrary pair of nonzero \( \mathbf{T}^e \) and \( \frac{\partial \hat{\varphi} (\mathbf{C}^e)}{\partial \mathbf{C}^e} \), is to have

\[ \mathbf{T}^e = 2 \frac{\partial \hat{\varphi} (\mathbf{C}^e)}{\partial \mathbf{C}^e}. \tag{73} \]

An immediate consequence of equation 73 in conjunction with equation 70 is that

\[ \mathbf{T}^p : \mathbf{L}^p \geq 0. \tag{74} \]

Equation 74 is satisfied for all possible combinations of \( \mathbf{T}^p \) and \( \mathbf{L}^p \), if and only if, the deviatoric plastic velocity gradient is co-linear with \( \mathbf{T}^p \).

To proceed further we need to specify the form of free-energy function. If we assume that the material remains isotropic during deformation, then the dependence of the free energy \( \varphi \) reduces to only the invariants of \( \mathbf{C}^e \), i.e.

\[ \varphi = \hat{\varphi} (\mathcal{I}^e), \tag{75} \]

where \( \mathcal{I}^e = (I_1(\mathbf{C}^e), I_2(\mathbf{C}^e), I_3(\mathbf{C}^e)) \) is set of invariants of \( \mathbf{C}^e \). The spectral decomposition of \( \mathbf{C}^e \)

\[ \mathbf{C}^e = \sum_{i=1}^{3} \omega_i^e \mathbf{r}_i^e \otimes \mathbf{r}_i^e, \quad \text{with} \quad \omega_i^e = \lambda_i^{e2}. \tag{76} \]

where \((\mathbf{r}_1^e, \mathbf{r}_2^e, \mathbf{r}_3^e)\) are the orthonormal eigenvectors of \( \mathbf{C}^e \) and \( \mathbf{U}^e \), and \((\lambda_1^e, \lambda_2^e, \lambda_3^e)\) are the positive eigen values of \( \mathbf{U}^e \). The free energy function 75 can then be written as a function of \( \lambda_1^e, \lambda_2^e \) and \( \lambda_3^e \), i.e.,

\[ \varphi = \hat{\psi} (\lambda_1^e, \lambda_2^e, \lambda_3^e). \tag{77} \]

Using equation 73 and chain rule of differential we can write

\[ \mathbf{T}^e = 2 \frac{\partial \hat{\psi} (\lambda_1^e, \lambda_2^e, \lambda_3^e)}{\partial \mathbf{C}^e} = 2 \sum_{i=1}^{3} \frac{\partial \hat{\psi} (\lambda_1^e, \lambda_2^e, \lambda_3^e)}{\partial \lambda_i^e} \frac{\partial \lambda_i^e}{\partial \mathbf{C}^e} = \sum_{i=1}^{3} \frac{1}{\lambda_i^e} \frac{\partial \hat{\psi} (\lambda_1^e, \lambda_2^e, \lambda_3^e)}{\partial \lambda_i^e} \frac{\partial \omega_i^e}{\partial \mathbf{C}^e}. \tag{78} \]

Using equation 76 into 78 it can be proved that

\[ \frac{\partial \omega_i^e}{\partial \mathbf{C}^e} = \mathbf{r}_i^e \otimes \mathbf{r}_i^e, \tag{79} \]

and now substituting 79 in equation 78 we obtain

\[ \mathbf{T}^e = \sum_{i=1}^{3} \frac{1}{\lambda_i^e} \frac{\partial \hat{\psi} (\lambda_1^e, \lambda_2^e, \lambda_3^e)}{\partial \lambda_i^e} \mathbf{r}_i^e \otimes \mathbf{r}_i^e. \tag{80} \]
Using equation 66, we can now find the Cauchy stress $T$ as

$$T = J^{-1} F^e T^e F^{eT} = J^{-1} R^e U^e T U^e R^{eT} = J^{-1} R^e \left( \sum_{i=1}^{3} \lambda_i \frac{\partial \psi}{\partial \lambda_i} r_i^e \otimes r_i^e \right) R^{eT}. \quad (81)$$

Similarly, using equation 68 we can obtain an expression for $M^e$ as

$$M^e = 3 \sum_{i=1}^{3} \lambda_i \frac{\partial \psi}{\partial \lambda_i} r_i^e \otimes r_i^e. \quad (82)$$

Next, we define a measure of elastic strain $E^e$ as

$$E^e \triangleq \ln U^e = \sum_{i=1}^{3} E_i^e r_i^e \otimes r_i^e \quad (83)$$

where,

$$E_i^e \triangleq \ln \lambda_i^e. \quad (84)$$

The free energy function can now be re-written in terms of $E_1^e, E_2^e$ and $E_3^e$ as

$$\varphi = \hat{\psi}(\lambda_1^e, \lambda_2^e, \lambda_3^e) = \psi(E_1^e, E_2^e, E_3^e). \quad (85)$$

$$M^e = \sum_{i=1}^{3} \frac{\partial \psi(E_1^e, E_2^e, E_3^e)}{\partial E_i^e} r_i^e \otimes r_i^e. \quad (86)$$

A popular choice for free energy form that is valid for moderately large elastic deformation is as follows:

$$\psi(E^e) = G|E_0^e|^2 + \frac{1}{2} K(\text{tr}E^e)^2 \quad (87)$$

In the above equation $G > 0$ and $K > 0$, and the free energy is an isotropic function of $E^e$ (satisfying frame invariance and material isotropy). Utilizing $87$ in equation $86$ we obtain an expression for Mandel stress in terms of $E^e$ as

$$M^{e} = 2GE_0^e + K(\text{tr}E^e)I. \quad (88)$$

Using the relation in equation 66 now we can obtain the Cauchy stress in the current configuration as:

$$T = J^{-1} R^e M^e R^{eT}. \quad (89)$$

We take caution of the fact that free energy, equation $87$ based on Henkcy strain measure does not satisfy the property of polyconvexity for extremely large elastic strains, see [76] [85] [108]. However, for
small to moderately large strains its use has proven to be extremely effective.

3.3.6 Plastic flow: yield and consistency condition

Recall that earlier in Section 3.3.6 we defined \( T^p \) as the work conjugate to plastic flow which is characterized by \( L^p \) (or \( D^p \)). Now we require a condition to determine the onset of yielding or active plastic flow. To this end, the yield surface is represented with a spherical surface of radius \( Y(e^p) > 0 \) in the space of symmetric and deviatoric tensors. Here \( e^p \) indicates the accumulated plastic strain, and radius of the the yield surface depends on it. The elastic range then identified as the closed ball with the radius \( Y(e^p) \). Plastic flow can only happen when \( T^p \) lies on the yield surface. From equation 61 we have \(|M_0^p| = |T^p|\) and thus condition for yielding can be written as

\[
|M_0^p| = Y(e^p) \quad \text{for} \quad D^p \neq 0,
\]

and the condition

\[
D^p = 0 \quad \text{for} \quad |M_0^p| < Y(e^p).
\]

We introduce a scalar yield function

\[
f = |M_0^p| - Y(e^p),
\]

and \( f \) follows the following constraint

\[
-Y(e^p) \leq f \leq 0.
\]

The yield condition in equation (90) is then equivalent to the requirement that

\[
f = 0 \quad \text{for} \quad D^p \neq 0,
\]

and equation (91) takes the form

\[
D^p = 0 \quad \text{for} \quad f < 0.
\]

The yield function \( f \) obeys the following additional restriction:

\[
\text{if} \quad f = 0 \quad \text{then} \quad \dot{f} \leq 0
\]

This leads to the no-flow condition

\[\text{In our notation, the } |A| \text{ of a tensor } A \text{ is equal to } \sqrt{\frac{2}{3} A : A} \]

32
and the consistency condition

\[ D^p = 0 \text{ if } f < 0 \text{ or if } f = 0 \text{ and } \dot{f} < 0 \quad (97) \]

Next, let

\[ Y'(e^p) = \frac{dY(e^p)}{de} \quad (99) \]

so that by equation \[94\]

\[ \bar{Y}(e^p) = Y'(e^p)D^p; \quad (100) \]

then, letting

\[ H(e^p) \overset{def}{=} Y'(e^p), \quad (101) \]

we arrive at

\[ \bar{Y}(e^p) = H(e^p)|D^p|. \quad (102) \]

Thus if we assume that \( D^p \neq 0 \), then by equation \[90\] \( |M_0^0| - Y(e^p) = 0 \), hence equation \[102\] yields

\[ |M_0^0| = H(e^p)|D^p|. \quad (103) \]

First of all, a consequence of flow rule is that

\[ N^p = \frac{M_0^0}{|M_0^0|} \quad (104) \]

i.e., the deviatoric Mandel tensor points in the direction of plastic flow.

\[ \dot{f} = \frac{M_0^0}{|M_0^0|} - \bar{Y}(e^p) \quad (105) \]

\[ \dot{f} = \frac{M_0^0}{|M_0^0|} : M_0^0 - H(e^p)|D^p| \quad (106) \]

\[ \dot{f} = N^p : M_0^0 - H(e^p)|D^p|. \quad (107) \]

Our next step is to compute the term \( N^p : M_0^0 \). Clearly,
\[ N^p : M^e = N^p : M^e, \]  
(108)

because \( N^p \) is deviatoric.

### 3.3.7 Time Integration for Rate-Independent Multiplicative Plasticity

Now we describe the time integration for the rate-independent and isotropic hardening deformation model with the multiplicative decomposition of the deformation gradient. The structure adopted here is that of a deformation driven problem, i.e., at each integration point of the finite element an updated deformation gradient is provided, and we are required to compute all the desired quantities. Most quantities are updated explicitly, however, the yield condition (and associated consistency parameter) are solved iteratively.

**Initialization:** At time step \( n = 0 \),

Compute the volume ratio \( J_{n+1} \) using

\[ J_{n+1} = det(F_{n+1}). \]  
(109)

Initialize \( N^p = 0, \Delta \tau^p = 0, \) and

\[ F^p_{n+1} = I \]  
(110)

where \( I \) is the second order identity tensor. Then,

\[ F^e_{n+1} = F_{n+1}F^{p-1}_{n+1} = F_{n+1}. \]  
(111)

Decompose \( F^e_{n+1} \) from equation (111) into \( R^e_{n+1} \) and \( U^e_{n+1} \) as

\[ F^e_{n+1} = R^e_{n+1}U^e_{n+1}. \]  
(112)

Compute the Hencky strain \( E^e_{n+1} \) as

\[ E^e_{n+1} = ln(U^e_{n+1}). \]  
(113)

Compute the Mandel stress as

\[ M^e_{n+1} = 2G E^e_{n+1} + K \left[ tr(E^e_{n+1}) \right] I. \]  
(114)

Now Cauchy stress \( T \) can be calculated by transforming the Mandel stress into the current configuration as

\[ T = J^{-1} F^{-1} M^e_{n+1} F M^e_{n+1} J. \]
\[ T_{n+1} = \frac{1}{J_{n+1}} R_{n+1} \varepsilon_{n+1}^e R_{n+1}^T. \]  

(115)

For any time step \( n > 0 \), assume the following quantities are known from previous step \( \{ F_n^e, F_n^p, Y_n, M_n^e, \varepsilon_{n+1}^d, h_n \} \). Given \( F_{n+1}^e \), our goal is to find \( \{ F_{n+1}^e, F_{n+1}^p, Y_{n+1}, M_{n+1}^e, \varepsilon_{n+1}^d, h_{n+1} \} \). We carry out explicit integration for all the quantities except for the consistency parameter, which we solve through numerical iterations so as to accurately satisfy the consistency condition during yielding. Following steps elaborate the procedure (trial quantities are denoted by \( \ast \ast \)):

**Step 1:** Compute the volume ratio \( J_{n+1} \) using

\[ J_{n+1} = \text{det}(F_{n+1}). \]  

(116)

**Step 2:** Compute

\[ F_{n+1}^e = F_{n+1}^e F_n^{-1}. \]  

(117)

**Step 2.1:**

\[ F_{n+1}^e = R_{n+1} U_{n+1}^e. \]  

(118)

**Step 2.2:** Compute the Hencky strain \( E_{n+1}^e \) as

\[ E_{n+1}^e = \ln(U_{n+1}^e). \]  

(119)

**Step 2.3:** Compute the Mandel stress as

\[ M_{n+1}^e = 2G E_{n+1}^e + K \left[ \text{tr}(E_{n+1}^e) \right] I. \]  

(120)

**Step 2.4:** Compute the equivalent (von mises) stress in the intermediate configuration using the deviatoric part of the Mandel stress:

\[ (\sigma_v^e)_{n+1} = \sqrt{\frac{3}{2} (M_{n+1}^e)^2_{0} : (M_{n+1}^e)^2_{0}}. \]  

(121)

In the above equation \( (M_{n+1}^e)_{0} \) denotes the deviatoric part of the Mandel stress \( M_{n+1}^e \). \( (M_{n+1}^e)_{0} \) is given by

\[ (M_{n+1}^e)_{0} = M_{n+1}^e + \bar{p}_{n+1} \cdot I, \]  

(122)

where

\[ \bar{p}_{n+1} = -\frac{1}{3} \text{tr}(M_{n+1}^e). \]  

(123)
and I is the second order identity tensor. Here we have not included any kinematic hardening (or back stress). Mandel stress is work conjugate to the plastic velocity gradient, and we have assumed plastic spin to be zero (on grounds of material isotropy), and plastic velocity gradient is essentially plastic stretching.

**Step 3:** Now we check for yielding by comparing the equivalent stress \((\sigma_{\nu}^{**})_{n+1}\), given by equation 133 and current yield strength \(Y_n\). If \((\sigma_{\nu}^{**})_{n+1} - Y_n < 0\), then the current increment is only elastic, set equivalent plastic strain increment to be zero \((\Delta \varepsilon_{\nu}^{pl} = 0)\), and goto **Step 6**.

**Step 4:** If the yield condition is satisfied and plasticity has to occur, then we calculate the direction of plastic flow \((N_n^p)\) and take it to be co-linear with the deviatoric part of Mandel stress.

\[
N_n^p = \frac{(M_n^e)_0}{\sqrt{2/3 (M_n^e)_0 : (M_n^e)_0}}.
\]  

**Step 5:** The plastic stretching \(D_n^p\) is given as

\[
D_n^p = \lambda N_n^p,
\]  

where \(\lambda\) is the equivalent plastic strain rate. If we assume that the current time step is small then \(\dot{\lambda} dt = \Delta \varepsilon_{\nu}^{pl} = d\lambda\). Now the goal is to find the incremental equivalent plastic strain such that consistency condition for the yield locus is satisfied.

**Step 5:** Here we demonstrate how to solve for the consistency parameter \((d\lambda)\) through iterations. During this iterative search we use \((**)\) to denote trial quantities. We start with some trial value for consistency parameter as \(d\lambda^{**}\).

**Step 5.1:** Compute trial plastic deformation gradient (recall that in a continuum setting we have \(\dot{F}^p = D^p F^p\))

\[
F_{n+1}^{**} = F_n^p + d\lambda^{**} N_n^p F_n^p.
\]  

**Step 5.2:** Compute trial elastic deformation gradient

\[
F_{n+1}^{**} = F_{n+1}^{p}\cdot F_{n+1}^{**} - 1.
\]  

**Step 5.3:** Decompose the obtained \(F_{n+1}^{**}\) from equation 127 into \(R_{n+1}^{**}\) and \(U_{n+1}^{**}\) as

\[
F_{n+1}^{**} = R_{n+1}^{**} U_{n+1}^{**}.
\]
Step 5.4: Compute the trial Hencky strain $E_{n+1}^{***}$ as

$$E_{n+1}^{***} = \ln(U_{n+1}^{***}). \quad (129)$$

Step 5.5: Compute the trial Mandel stress as

$$M_{n+1}^{***} = 2G E_{n+1}^{***} + K \left[ tr(E_{n+1}^{***}) \right] I, \quad (130)$$

where $G$ is the shear modulus and $K$ is the bulk modulus. If Youngs modulus $E$ and Poisson’s ratio $\nu$ are known, then bulk and shear modulus are obtained from following relations

$$G = \frac{E}{2(1 + \nu)}. \quad (131)$$

$$K = \frac{2G\nu}{1 - 2\nu}. \quad (132)$$

Step 5.6: Compute the trial equivalent (von Mises) stress in the intermediate configuration using the deviatoric part of the Mandel stress.

$$(\sigma_v)^{***}_{n+1} = \sqrt{\frac{3}{2}(M^{***}_{n+1})_0 : (M^{***}_{n+1})_0}, \quad (133)$$

where

$$(M^{***}_{n+1})_0 = M^{***}_{n+1} + \bar{\sigma}_{n+1} I, \quad (134)$$

and

$$\bar{\sigma}_{n+1} = -\frac{1}{3} tr(M^{***}_{n+1}). \quad (135)$$

Step 5.7: Compute trial value of the new yield locus:

$$Y^{***}_n = Y_n + h^{***}_{n+1} d\lambda^{***}. \quad (136)$$

In the above $h^{***}_{n+1}$ is the hardening modulus (and depends upon the accumulated plastic strain).

Step 5.8: Check whether the consistency condition is satisfied:

$$(\sigma_v)^{***}_{n+1} - Y_n - h^{***}_{n+1} d\lambda^{***} = 0 \quad (137)$$

If the above equation is not satisfied (according to some predefined numerical tolerance), then choose the different value of $d\lambda^{***}$ and go to Step 5.1. We performed this iterative procedure using a Bisection method.
**Step 5.9:** The solution \(d\lambda\) satisfying the equation \(137\) is taken as the incremental equivalent plastic strain \((\Delta \varepsilon_{pl}^{n})\)

\[
\Delta \varepsilon_{pl}^{n} = d\lambda.
\]  

Also, the value of the hardening modulus satisfying the consistency condition is updated as \(h_{n+1}\).

**Step 6:** Compute the plastic part of the deformation gradient at time \(t_{n+1}\)

\[
F_{n+1}^{p} = F_{n}^{p} + \Delta \varepsilon_{pl}^{n} N_{p} F_{n}^{p}.
\]  

**Step 7:** Compute the elastic part of the deformation gradient at time \(t_{n+1}\)

\[
F_{n+1}^{e} = F_{n+1}^{p-1}.
\]  

**Step 8:** Decompose \(F_{n+1}^{e}\) from equation \(140\) into \(R_{n+1}^{e}\) and \(U_{n+1}^{e}\) as

\[
F_{n+1}^{e} = R_{n+1}^{e} U_{n+1}^{e}.
\]  

**Step 9:** Compute the Hencky strain \(E_{n+1}^{e}\) as

\[
E_{n+1}^{e} = ln(U_{n+1}^{e}).
\]  

**Step 10:** Compute the Mandel stress at time \(t_{n+1}\)

\[
M_{n+1}^{e} = 2G E_{n+1}^{e} + K \left[ tr(E_{n+1}^{e}) \right] I.
\]  

**Step 10:**

Now Cauchy stress \(T_{n+1}\) can be calculated by transforming the Mandel stress into the current configuration as

\[
T_{n+1} = \frac{1}{J_{n+1}} R_{n+1}^{e} M_{n+1}^{e} R_{n+1}^{eT}.
\]  

**Step 11:**

Updated yield strength

\[
Y_{n+1} = Y_{n} + h_{n+1} \Delta \varepsilon_{pl}^{n}.
\]  

Update the total equivalent plastic strain

\[
\bar{\varepsilon}_{n+1}^{pl} = \bar{\varepsilon}_{n}^{pl} + \Delta \varepsilon_{pl}^{n}.
\]
3.4 Hypoelasticity: Additive Decomposition of Spatial Strain Rate

In a hypoelastic constitutive law, the time rate change in stress is expressed as some function of time derivatives of strains, and the integration procedure utilized should satisfy the criterion of “incremental objectivity”, see [46, 97]. The stresses are obtained by integration of these rate-type equations, see [95, 34, 119, 86, 65, 118].

Using equation 10 we can write

\[ D = \frac{1}{2} [L + L^\top] = \text{Sym}(L), \quad (147) \]

where \( \text{Sym} \) yields the symmetric part of the operand. Substituting the expression for \( L \) from equation 16 in equation 147, we obtain

\[ D = \text{Sym} \left[ L^e + F^e (D^p + W^p) F^{-1} e \right]. \quad (148) \]

By setting plastic spin \( W^p = 0 \), substituting \( F^e = R e U^e \), and utilizing \( D^e = \text{Sym}(L^e) \) we can re-write equation 148 as

\[ D = D^e + \text{Sym} \left[ R^e U^e (D^p) U^{-1} e R^{-1} e \right]. \quad (149) \]

Further if elastic stretches are small, i.e. \( U^e \approx 1 \), then above equation can be reduced to

\[ D = D^e + \text{Sym} \left[ R^e (D^p) R^{-e} \right]. \quad (150) \]

It can be shown that \( R^e (D^p) R^{-e} \) is symmetric and hence we can re-write

\[ D = D^e + \bar{D}^p, \quad (151) \]

with

\[ \bar{D}^p = R^e D^p R^{-e} = R^e D^p R^\top e. \quad (152) \]

Equation 151 is commonly known as the additive decomposition of the spatial strain rate. Material models based on above decomposition of require evolution rule for \( D^e \) and \( D^p \) (or \( \bar{D}^p \)).

One of the commonly utilized measure of objective rate is known as the Jaumman rate, and the Jaumman rate of cauchy stress (denoted by \( T^J \)) is given as

\[ T^J = \dot{T} - WT + TW, \quad (153) \]

where \( \dot{T} \) is the material derivative of the Cauchy stress with respect to undeformed (fixed) basis/configuration. \( T^J \) is also known co-rotational stress rate as it corresponds to the time rate of change with respect to the observer-frame attached and rigidly-rotating with the material point of interest. It
should be noted that the hypoelastic relation given by (153) is not known to be thermodynamically consistent, in the sense that constitutive equation for $T$ is not derived from a free energy function, i.e., path independence aspect is missing from its definition. Issues with rate-type formulations have been well-known for some time [109, 63], however, given their simplicity, efficiency and applicability for problems with small elastic strains they are still practiced [68, 33, 78]. The constitutive equation for small strain elasticity is

$$T = C : E^e$$  \hspace{1cm} (154)

where $C$ is the fourth order elasticity tensor

$$C = \left( K - \frac{2}{3} G \right) I \otimes I + 2G I.$$  \hspace{1cm} (155)

Equation (154) can also be written in a Jaumann (corotational) rate form as

$$T^J = C : D^e.$$  \hspace{1cm} (156)

This finally reduces to

$$T^J = (K - \frac{2}{3} G) (I : D^e) I + 2G D^e.$$  \hspace{1cm} (157)

It should be noted that the above constitutive law describing the elastic deformation satisfies the need of “frame-indifference” or objectivity. Now we need to specify the evolution law for the plastic flow $\dot{D}^p$. As shown in the previous section, equation (104) (the normality flow rule) can be used to write

$$\dot{D}^p = \dot{\lambda} N^p = \dot{\lambda} \frac{M^p}{|M_0^p|}$$  \hspace{1cm} (158)

where $\dot{\lambda}$ is the consistency parameter, and we have used equation (104) for the direction of plastic flow.

From equation (90) we know that during yielding $|M_0^p| = Y(e^p)$. In hypoelasticity the relation between Cauchy stress and Mandel stress (equation (89)) can be reduced to (by choosing $U^e \approx 1$ and $J \approx 1$):

$$T = R^e M^e R^{eT}.$$  \hspace{1cm} (159)

Accordingly, it can be shown that $T_0 = R^e M_0^e R$, and substituting Mandel stress in terms of Cauchy stress, and $|M_0^p| = Y(e^p)$ in equation (158) we can obtain

$$\dot{D}^p = \dot{\lambda} \left( \frac{R^e T_0 R^{eT}}{Y(e^p)} \right).$$  \hspace{1cm} (160)

Using equation (162) equation (160) can be re-written as
\[
\dot{D}^p = \dot{\lambda} \left( \frac{T_0}{Y(e^p)} \right).
\] (161)

Thus, we have obtained the flow rule in terms of Cauchy stress. Also, \(\dot{\lambda} = e^p = |\dot{D}|\). In case of hypoelasticity, it is easy to show that \(|M_0^p| = |T_0|\), thus the yield function given in equation 92 can be re-written as:

\[
f_h = |T_0| - Y(e^p).
\] (162)

where \(f_h\) indicates specialization of the yield function in case of hypoelasticity. Now during plastic yielding (\(f_h = 0\) and \(\dot{f}_h = 0\)), i.e.,

\[
\dot{f}_h = |\dot{T}_0| - |Y(e^p)|,
\] (163)

\[
\dot{f}_h = \frac{T_0}{|T_0|} : T_0 - H(e^p)|\dot{D}^p|.
\] (164)

Now we define the direction of plastic flow in current configuration as

\[
\bar{N}^p = \frac{T_0}{|T_0|},
\] (165)

and accordingly re-write the equation 166 as

\[
\dot{f}_h = \bar{N}^p : T_0 - H(e^p)|\dot{D}^p|.
\] (166)

Since \(\bar{N}^p\) is deviatoric and \(T\) is symmetric, therefore \(\bar{N}^p : T_0 = \bar{N}^p : T\). Thus during active yielding, we have \(\dot{D}^p \neq 0\), and \(\dot{f}_h = 0\), therefore equation 166 becomes

\[
\bar{N}^p : T - H(e^p)|\dot{D}^p| = 0.
\] (167)

From equation 153 we can substitute \(T\) in terms of \(T^J\), \(T\) and \(W\), in equation 167 to finally get

\[
\bar{N}^p : T^J - H(e^p)|\dot{D}^p| = 0.
\] (168)

Equation 156 can be re-written as

\[
T^J = C : (D - \bar{D}^p),
\] (169)

and substituting \(\bar{D} = \dot{\lambda}\bar{N}^p\), we have

\[
T^J = C : D - \dot{\lambda}C : \bar{N}^p,
\] (170)
Substituting $T'$ from equation 170 into equation 168 and writing $|D^p| = \dot{\lambda}$, we can solve for $\dot{\lambda}$ as

$$\dot{\lambda} = \frac{\bar{N}^\nu : C : D}{H(e^\nu) + \bar{N}^\nu : C : \bar{N}^\nu}$$

(171)

Finally back substituting $\dot{\lambda}$ from equation 171 in equation 170, we obtain

$$T' = C : D - \frac{\bar{N}^\nu : C : D}{H(e^\nu) + \bar{N}^\nu : C : \bar{N}^\nu} \bar{N}^\nu,$$

(172)

and upon rearranging we can write above as

$$T' = \left( C - \frac{C : \bar{N}^p \otimes \bar{N}^p : C}{H(e^\nu) + \bar{N}^p : C : \bar{N}^p} \right) : D.$$  

(173)

Accordingly, the elasto-plastic tangent modulus is given as

$$C^{ep} = \left( C - \frac{C : \bar{N}^p \otimes \bar{N}^p : C}{H(e^\nu) + \bar{N}^p : C : \bar{N}^p} \right).$$

(174)

Equation 174 provides Jaumann stress rate in terms of spatial stretching, and now we need to integrate these equations with respect to time to obtain stresses etc.

### 3.4.1 Time Integration Rate-Independent Hypoeelasticity

We again follow a deformation driven problem structure and the computation of strain increment for the co-rotational updates is computed using mid-point rule as proposed by Hughes and Winget [46].

**Initialization:** For $n = 0$,

Initialize $N_{n+1}^p = 0$, $\Delta \varepsilon_{n+1}^{pl} = 0$, $T_{n+1} = 0$, $T_{n+1}' = 0$.

**At any time** $n + 1 > 0$

We assume that quantities at time the beginning of the time increment $t_{n+1}$ (i.e. at the end of $t_n$) are known: Cauchy stress $(T_n)$, co-rotational stress $(T_n')$, yield strength $Y_n$, $\varepsilon_{n}^{pl}$, $h_n$. Now given $F_{n+1}$, $\Delta t_{n+1} (= t_{n+1} - t_n)$, the goal is to find the updated quantities as $T_{n+1}$, co-rotational stress $T_{n+1}'$, yield strength $Y_{n+1}$, $\varepsilon_{n+1}^{pl}$, $h_{n+1}$. A radial-return algorithm is used to satisfy the yield condition. In what

---

4 We assume that we have the solution to the problem up to time $t_n$ (when $x_n$ is the spatial location), and at time $t_{n+1}$ the prescribed deformation takes a material element to the spatial point $x_{n+1}$. This is equivalent way of saying that solution up to $F_n$ is known, and we need to find the solution corresponding to $F_{n+1}$. We denote the incremental displacement as $u = x_{n+1} - x_n$, and a mid-point configuration $x_{n+1} = \frac{1}{2}(x_{n+1} + x_n)$. Now the increment in the strain, from $t_n$ to $t_{n+1}$, are computed with respect to this mid-point configuration as:

$$\Delta \varepsilon_{n+1} = \frac{1}{2} \left( \frac{\partial u}{\partial x_{n+1}} + \left( \frac{\partial u}{\partial x_{n+1}} \right)^T \right).$$

This maintains second order accuracy as long as time steps are small.
follows, the trial quantities are denoted **. For a given strain increment $\Delta E_{n+1}$, the goal is to find the additive split of the strain increment into elastic and plastic parts:

$$\Delta E_{n+1} = \Delta E_e^{n+1} + \Delta E_p^{n+1}$$ (175)

**Step 1:** Compute the trial corrotational stress using the strain increment as

$$T_{n+1}^{J**} = T_n^J + (K - \frac{2}{3}G)(\Delta E_{n+1}) + 2G(\Delta E_{n+1})_0.$$ (176)

**Step 2:** Compute the trial equivalent von-mises stress

$$(\sigma_v)^{**}_{n+1} = \sqrt{\frac{3}{2}(T_{n+1}^{J**})_0 : (T_{n+1}^{J**})_0}.$$ (177)

**Step 3:** If $(\sigma_v)^{**}_{n+1}$ computed in equation [177] is less than $Y_n$ then it is an elastic increment, set $\Delta E_p^{n+1} = 0$, and go to Step 4.5, else continue with following.

**Step 4:** Radial return algorithm is employed to compute the incremental plastic strain. Following steps describe how this is achieved.

**Step 4.1:** The direction of plastic flow is computed as

$$N_{p}^{n+1} = \frac{(T_{n+1}^{J**})_0}{\sqrt{\frac{3}{2}(T_{n+1}^{J**})_0 : (T_{n+1}^{J**})_0}}.$$ (178)

**Step 4.2:** The magnitude of the incremental equivalent plastic strain $(\Delta \bar{\varepsilon}_{pl})$ is obtained by consistently linearizing and satisfying the yield condition, which yields

$$(\Delta \bar{\varepsilon}_{pl})_{n+1} = \frac{(\sigma_v)^{**}_{n+1} - Y_n}{3\mu + h_n}.$$ (179)

**Step 4.3:** The incremental plastic strain is obtained as

$$\Delta E_p^{n+1} = (\Delta \bar{\varepsilon}_{pl})_{n+1} N_{p}^{n+1}.$$ (180)

**Step 4.4:** By using equation [179] the yield strength at $t_{n+1}$ is updated as

$$Y_{n+1} = Y_n + h_n \times (\Delta \bar{\varepsilon}_{pl})_{n+1}.$$ (181)

The hardening modulus is also updated accordingly from the experimental data.

**Step 4.5:** Compute the elastic strain by subtracting the plastic strain increment given by equation [180] from the total strain increment as

$$\Delta E_{n+1}^{e} = \Delta E_{n+1} - \Delta E_{n+1}^{p}.$$ (182)
Step 5: Compute the updated corrotational stress using the incremental elastic strains from equation 182.

\[
T^j_{n+1} = T^j_n + \left(K - \frac{2}{3}G\right) \text{tr}(\Delta E^e_{n+1}) + 2G(\Delta E^e_{n+1})_0 \tag{183}
\]

Step 6: Obtain the Cauchy stress by transforming the corrotational stress into the current frame as:

\[
T_{n+1} = R_{n+1}T_n R^T_{n+1}, \tag{184}
\]

where \(R_{n+1}\) is obtained from polar decomposition of \(F_{n+1}\) as

\[
F_{n+1} = R_{n+1}U_{n+1}. \tag{185}
\]

3.5 Solution to Global Equilibrium Equations Using Abaqus Explicit Dynamics

The Explicit dynamic solver used by Abaqus is based on the central difference formulas for the velocity and the acceleration, and the use of diagonal or lumped element mass matrices is adopted. The quantities such as displacement, velocities and accelerations are specified at nodal points. Let \(\mathcal{F}_n\) and \(\mathcal{I}_n\) denote the external and internal (due to resultant of internal stresses) nodal point force vectors. Then nodal point acceleration vector (\(\ddot{\mathcal{U}}_n\)) is given as:

\[
\ddot{\mathcal{U}}_n = \mathcal{M}^{-1}(\mathcal{F}_n - \mathcal{I}_n), \tag{186}
\]

where \(\mathcal{M}\) is the diagonal lumped mass matrix. \(\mathcal{I}\) is obtained from the VUMAT implementation of the material model. The nodal velocity vector, at mid-point time, is then computed as:

\[
\dot{\mathcal{U}}_{n+\frac{1}{2}} = \dot{\mathcal{U}}_{n-\frac{1}{2}} + \frac{\Delta t_{n+1} + \Delta t_n}{2} \ddot{\mathcal{U}}_n, \tag{187}
\]

and the updated nodal point displacement vector is then computed as

\[
\mathcal{U}_{n+1} = \mathcal{U}_n + \Delta t_{n+1} \dot{\mathcal{U}}_{n+\frac{1}{2}}, \tag{188}
\]

where \(\Delta t\) is defined as

\[
\Delta t_{n+1} = t_{n+\frac{1}{2}} - t_{n-\frac{1}{2}}. \tag{189}
\]

\(^{5}\text{Nodal values are interpolated at the Gauss integration points of the element via shape functions of the used element. Obtained interpolated quantities at integration points are passed either to VUMAT or Abaqus material library to obtain stresses at the integration points which are further integrated for the whole element. This integrated stresses for the element is the internal force } \mathcal{I} \text{ which is assembled for all the elements.}\)
As stated earlier, the explicit procedure does not require any tangent stiffness, and for quasi-static problems (where acceleration terms are negligible) the $M$ matrix can be scaled. However, caution must be taken that mass scaling does not lead to excessively high and unrealistic kinetic energies.

The performance of the multiplicative plasticity based model and hypoelastic formulation, calibrated against tensile test, is shown in Figure 9. We shall use this material model in rolling simulation.

### 3.6 Contact Interaction and Friction

To set up the interaction behavior, we estimated the coefficient of friction between a stainless steel block (made of same material and surface properties as the rollers) and polymer films using a friction-fixture on Instron mechanical tester. Stainless steel block of approximately 500 g was used, and the load vs. displacement during sliding motion of the block on the film was recorded. This is shown in Figure 10. The coefficient of friction ($\mu$) was estimated to be 0.4. In the finite element simulations we used the isotropic Coulomb friction model, with coefficient of friction of 0.4, and limited the maximum value of the shear-traction equal to the shear yield strength of the material. The Coulombic friction model is schematically shown in Figure 11 and according to this the tangential traction is dependent on the normal traction but limited by the material’s yield strength.

We employed the default kinematic constraint algorithm for the contact interaction. In particular, the tangential behavior was modeled through a penalty formulation (with a very large penalty factor of $10^{10}$), and in absence of slip the relative tangential displacement is computed by dividing the frictional force by the chosen penalty factor. This can be helpful when friction can cause local excessive distortion.
Figure 10: Force versus displacement curve during measurement of coefficient of friction on Instron mechanical tester. The coefficient of friction is estimated based upon the average force \( F_{\text{friction}} \), in the steady state, during sliding as \( \mu = \frac{F_{\text{friction}}}{m'g} \), where \( m'g \) is the weight of the block. For \( m' = 0.5 \) kg and \( F_{\text{friction}} = 2N \) (based on this graph), \( \mu_k \) is estimated to be 0.4.

The contact algorithm in a pure master-slave surface formulation, detects contact when the slave node penetrates into the master segment, therefore a sufficiently fine mesh for both bodies will be chosen so that no spurious interpenetration of rigid-roller occur into the ductile film substrate. Additionally, choice of larger roller radii, slow angular speeds, and sufficiently fine mesh are favorable conditions for accurate function of the contact algorithm. Although not necessarily required, we chose to work with a finite sliding formulation; i.e., detect a change in master segment for every slave node (through a global search) after a set number of time increments. A summary of the contact algorithm for rigid (master) and soft surface (slave) is given in Algorithm 1.

![Diagram of contact algorithm](image)

Figure 11: Example of Coulombic friction (rate-independent friction model).
Algorithm 1 Pseudo code for Abaqus/Explicit Contact for rigid (master) segment and soft (slave) node interaction. Assumed that master surface follows a displacement specified motion.

```plaintext
1: procedure Contact
2:   Time → t_n
3:   S^{t_{n+1}}
4:   M^{t_{n+1}}
5: if (no penetration) then
   ▷ Check penetration of slave node into the current master segment
6:   return false;
7: else
   ▷ There is penetration and corrector phase starts
8:   N^{t_{n}}
9:   τ^{t_{n}}
10: RP^{t_{n}}
11: +S^{t_{n+1}}
12: F^{t_{n}}
13: if (no friction) then
   ▷ Check if friction is specified for the problem
14:   return false;
15: else
   ▷ Friction is specified
16:   δU
17: if (|δU| > 0) then
   ▷ Slip will occur
18:   F^{t_{n}}_{fr}
19: if (F^{t_{n}}_{fr} > μF^{t_{n}}_{N}) then
   ▷ Slip will occur
20:   +S^{t_{n+1}} ← +S^{t_{n+1}}
21:   F^{t_{n}}_{fr} ← μF^{t_{n}}_{N}
22: else
   ▷ No slip (if penalty formulation compute small tangential motion)
23: return false;
24: else
   ▷ No relative displacement
25: return false;
26: t_{n} ← t_{n+1}
27: if specified, update master segment for the slave node
28: end procedure
```

▷ All quantities known, need to find explicit updates from
▷ Perform trial kinematic update of the slave node
▷ Perform the kinematic update of the current master segment
▷ No contact and proceed to the next slave node
▷ Check penetration of slave node into the current master segment
▷ No tangential force and proceed for the next slave node
▷ No tangential force and proceed for the next slave node
▷ Friction is specified
▷ Compute relative displacement in tangential direction
▷ Compute tangential force $F^{t_{n}}_{fr}$ assuming no slip
▷ Update the projected location of the slave node
▷ Maximum value of $F^{t_{n}}_{fr}$ can be $μF^{t_{n}}_{N}$
▷ No relative displacement
▷ No tangential force and proceed for the next slave node
▷ At the end of time increment
4 Rolling-Apparatus

We proposed a symmetric ‘C’ shaped roller-stand on which rollers can be mounted. Figure 12 shows a single roller-stand assembly comprising of a roller-carrying shaft, where the shaft is attached to the roller-stand with an appropriate choice of the bearings. The bearings themselves can be press-fitted in the ‘C’ shaped stages. Two such two roller-stands can potentially be attached onto a linear slide for controlling the roll-gap. The ‘C’ shaped stages are proposed to be made out of Aluminum so as to minimize the weight of the assembly. During the rolling operation it is critical that the thickness reduction of film-stack should also be uniform along the width direction (Figure 4). Compression of film-stack will generate roll-separating force, and it is desired that machine parts (roller-stand, shafts, rollers, etc.) have sufficient stiffness against this force while causing homogeneous and uniform compression of the film-stack. Although high stiffness against the roll-separating force is a primary requirement so as to minimize structural deflections in the machine, additional factors such as strength and fatigue life of rollers and/or other machine parts are critical and must be considered for a commercial scale machine.

Since uniform compression of film-stack is a major requirement to achieve high quality bonding, we analyze our proposed ‘C’ shaped roller-stand design for deflections incurred during the compression. Figure 13 shows a 3D finite element model (following the CAD model of a single roller-stand assembly as shown before). The rollers, position handles and shafts are considered to be made of stainless steel with an elastic modulus of 190 GPa (and yield strength of 600 MPa). The ‘C’ shaped stand is made of Aluminum with an elastic modulus of 79 GPa (and yield strength of 240 MPa). For simplification, we model all the contact conditions (between roller and shaft, shaft and roller stand, and position handle and roller stand) with tie-constraints. The interface between rollers and films is modeled using Coulombic friction as described before. The radius of the rollers was chosen as 100 mm and thickness of a single stack as 0.6 mm and width as 10 mm.
Elastic-plastic finite element simulations, as discussed in previous sections, are employed (see [90]). Since 3D finite element simulations are computationally expensive and time consuming, here we only analyze the static compression of the film-stack and study deformation behavior of the roller frame and film-stack so as to first evaluate if our proposed rolling-apparatus is able to at-least meet the necessary functional requirements. In the next section, we shall employ plane-strain rolling simulations to study steady-state rolling loads and deformation patterns in the roller bite. Figure 14 shows the through-thickness plastic deformation in the direction of compression at various levels of load. Although we have treated films as a single stock (an assumption which we shall verify holds pretty good in steady state rolling), the proposed rolling-apparatus is able to deform the film-stack plastically through its thickness and that the plastic strain across the thickness is almost homogeneous. It is also worthwhile noting that load levels predicted by the rigid-perfect-plastic model for 20% nominal thickness reduction strains are smaller than those appearing in this simulation. During the static compression of the film-stack in the rollers, negligible elastic strains were noted in other parts of the finite-element model, suggesting that parts are safe from any yielding failure (we omit to show those details for the sake of brevity).

To study the effect of roll-separating force on the roller assembly, we study the shaft deflection by calculating the angle $\psi$ (in degrees) which gives an estimate on how much the shaft axis deflects with respect to the base during compression. The calculation of $\psi$ is schematically shown in Figure 15. Two points on the shaft axis, ‘Center RP’ and ‘Base RP’, are chosen. The ‘Center RP’ corresponds to the middle point in the vertical direction on the shaft axis, and the ‘Base RP’ corresponds to the point on the shaft axis where the shaft first encounters coupling with the bottom of the frame. $U_{x,rel}$ and $U_{y,rel}$ indicate the relative displacement of ‘Center RP’ with respect to ‘Base RP’, from which the angular
Figure 14: Static compression of the film-stack in the ‘C’ shaped roller assembly. At different levels of compression loads, through-thickness plastic strain (natural log of the plastic strain) in x-direction (PE11) in the film-stack is shown. The shown section corresponds to the one where gap between the rollers is minimum and maximum compression in thickness occurs. See Supplementary Video S1.

Figure 15: Calculation of $\psi$ for the analysis of shaft deflection during stack compression.
The deflection ($\psi$) can be calculated. These quantities are computed during the finite element simulations at different levels of compression loads and shown in Figure 16.

Clearly, the proposed symmetric ‘C’ shaped roller stand design provides a desirably high stiffness against the roll-separating force and very small angular deflections are incurred at loads up to 4 kN. Although bearings are not modeled in this simulation, but if appropriately stiff bearings are chosen (along with other parts), we expect a successfully operating roll-bonding apparatus at these levels of loads.

4.1 Selection of other parts in the rolling-apparatus

Once we have estimated the roll separating forces and torques, and also considered a factor of safety, we can safely select the remaining frame structure of the machine, bearings, drive systems, etc., so that system can safely operate at least up to a few kN of loads. It is worthwhile to note that the absolute levels of loads expected in roll-bonding of thin polymeric films of current interest are quite moderate (up to a few kilo-Newton) compared to those involved in metal rolling operations (up to several Mega-Newton), and therefore a need to deploy any specialized load bearing machine elements is not seen at this stage.

Figure 17 shows the complete CAD model of roll-bonding machine. Table 1 lists the main components of this machine along with their functionalities. There were several choices of bearings available to us, for example, spherical, roller, sliding bearings (also known as journal bearings), etc. Both spherical and roller bearings come with pre-loaded mechanisms and in particular roller bearings can endure very large loads due to line contact. The slide bearings rely on hydrodynamic lubrication and are somewhat cheaper. For slide bearings there will be a fluid layer between the shaft and the bearings in the inner race. Due to current application with pharmaceutical films, we discarded the journal bearings to avoid any potential
Table 1: Major components of roll-bonding system and their functionality. Functional requirements of each element were carefully analyzed before making design choices.

<table>
<thead>
<tr>
<th>Part</th>
<th>Functionality</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Rollers</strong></td>
<td>Compression rollers capable of applying several Kilo-Newton of line-loading were machined out of stainless steel with a surface finish of $R_a \sim 6 \mu m$. The diameter of each roller is 200 mm and height is 100 mm.</td>
</tr>
<tr>
<td><strong>Shafts</strong></td>
<td>Drive the compression rollers and mounted using bearings. Shafts were made of 304 stainless steel, with a diameter of 31.75 mm.</td>
</tr>
<tr>
<td><strong>Linear Slides</strong></td>
<td>Carry the stage of the compression rollers, and enable setting the relative gap between the rollers. The slides were purchased from Stelron Components Inc.</td>
</tr>
<tr>
<td><strong>Micrometer</strong></td>
<td>Measures the distance between compression rollers which are mounted on the linear-slides.</td>
</tr>
<tr>
<td><strong>Load Cell</strong></td>
<td>Purchased from Omega (model LC 305-1K, approximate capacity of 4.5 kN) to measure the compression load (N). It is mounted on the left roller-stage.</td>
</tr>
<tr>
<td><strong>Springs</strong></td>
<td>Extension springs with stiffness approximately 5.8 N/mm are used to secure the left roller-stage and load cell against the position handle.</td>
</tr>
<tr>
<td><strong>Position Handle</strong></td>
<td>Used to drive the left roller-stage through a threaded-rod and set its position.</td>
</tr>
<tr>
<td><strong>Threaded Rod (Right)</strong></td>
<td>Used to set the position of the right roller. Locking nuts secure it in the desired position.</td>
</tr>
<tr>
<td><strong>Pulley System</strong></td>
<td>Attached to the shafts of the rollers, and interconnected by a double-sided timing belt (not shown so as to avoid clutter).</td>
</tr>
<tr>
<td><strong>Stepper Motor</strong></td>
<td>Purchased from Applied Motion Products (Model STM-23 QN). An appropriate gear train was selected to provide a minimum angular speed of 0.08 rpm and maximum torque of 50 Nm to drive compression rollers.</td>
</tr>
</tbody>
</table>
danger of oil seeping. Also, the roller bearings can wear out over time, and therefore we selected cost-effective spherical bearings. The selected bearings were rated to take up to 7 kN radial loads.

The stages carrying the rollers are attached on the anti-friction linear slides such that distance between them can be adjusted. The anti-friction linear slides were procured from Stelron to bear the desired weight of the assembly. Based on the material properties, we estimated the weight of each bonding stage to be 70 kgs, and the chosen Stelron slides were capable of carrying this weight while incurring lateral deflections on the order of a micron. The distance between the rollers is measured by a micrometer. The position of the roller-carrying-stages attached on the linear-slides is set using the two threaded-rods. A load cell is mounted on the left roller-carrying-stage to measure the compression loads. The two threaded rods are secured to the main frame by the action of locking-nuts. During operation the position of the right-roller-stage is fixed, and the position of the left roller-stage is adjusted through threaded-rods and a position handle. The rotatory motion of the position-handle that drives the left-threaded-rod causes the sliding motion of its roller-stage. Two extensional springs are mounted on the frame and the left roller-carrying-stage such that load cell is well-secured against the left threaded rod. A stepper motor with 50 Nm torque capacity was chosen, and this is sufficiently larger than the estimated torque in the previous section. Other machine elements, not discussed in detail here due to sake of conciseness, were sized and selected appropriately. The designed and fabricated roll-bonding machine is shown in Figure 18.

During the operation, the position of the left roller-carrying-stage is adjusted until a desired level of force is read by the load cell; the corresponding distance between the rollers is read in the micrometer. The rotary shafts are driven by a system of pulleys and double-sided timing belts using a stepper-motor. The stepper motor is attached to the main-frame of the machine, and during operation an appropriate level of tension is maintained in the timing belt which drives the pulleys on the rotary shafts. To ensure that the timing belt does not slip over the pulleys when driving the rotary shafts, an idler pulley is present to uptake the slack in the timing belt which occurs as rollers are brought closer. For fixed roller radius, the angular speed of the motor dictates the exit speed \( V_2 \) of the film-stock from the rollers.

We emphasize that in the current application, a thin stack of low modulus and ductile polymer films is fed through the rollers and due to its compliant nature the stack is susceptible to wrinkling or buckling-like defects in presence of any longitudinal (along the feed-direction) compressive stresses. Speed mismatch between the rollers is one such source, and therefore precise functioning of the drive mechanism for the rollers is quite important.

In principle one can include details of bearings, nuts, bolts, etc. in the 3D finite element model to make more detailed predictions, but this will significantly increase the computational cost. We found that route unnecessary. Furthermore, details of certain mechanical components, such as assembly information and construction of the bearings along with its constituent material properties were not available. Next,
Figure 17: The CAD model of roll-bonding machine. The cylindrical rollers provide adequate line-loading to achieve plastic-deformation on incoming films over a desired interval of time. See Supplementary Video S3.

Figure 18: A view of roll-bonding machine. Roller-radii are 100 mm. The parallel between the rollers, in the final assembly over the entire height of the rollers (100 mm), using metal feeler gauze was found accurate up to ±0.003".
we experimentally verify the performance of our roll-bonding machine and present more analyses.

5 Experimental and Simulation Results

As stated earlier, the overall success of the roll-bonding machine lies in precisely applying a line-loading without introducing any unwanted deflection, stretching of the frame, or failure of any machine element during compression between the rollers. In order to evaluate the accuracy of our actual system, we first test the stand-by performance (i.e. compression without the presence of films) in the displacement-controlled mode by imposing different levels of compression using the micrometer and measuring the loads on load-cell as rollers are brought into the contact. The exercise was repeated multiple times up to a maximum load around 1.5 kN, and a load vs. deflection curve (with error-bars) is shown in the Figure 19. From these results it can be concluded that the load follows the displacement in essentially a linear fashion, with good repeatability. The stiffness of the roll-bonding machine based on the experimental load vs displacement curve is estimated to be $5.3 \times 10^6$ N/m.

Figure 23 shows a snapshot of roll-bonding of multiple layers through our devised machine. Several specimens comprising of film-stack were rolled through the machine at different levels of compression loads, leading to different levels of plastic strain. The initial widths of specimens (which were approximately 15 mm) before rolling, were found to be measurably unchanged after rolling, and therefore rolling passes were consistent with the plane-strain scenario. The initial thickness of film-stacks were approximately 0.6 mm, and the average final thickness of the rolled stock depended on the level of plastic-strain imposed. A detailed correlation between imposed plastic strain and degree of bonding see [92, 90]. For several rolled-specimens, we measured the variation in final-thickness of the rolled-stock (at 10 randomly
chosen points on the stock along the length and width) and plotted the standard deviation in final-thickness with respect to plastic strain. This is shown in Figure 20. The uniformity in thickness of films after rolling is greater than 2%, and demonstrates the successful operation of the designed hardware.

Next, we wish to study the deformation of polymer films in the roller-bite and associated rolling loads. A film-stack of initial thickness 0.6 mm was modeled in plane strain compression under the rigid rollers. We utilized the symmetry and therefore carried simulation for one-half of the model, as shown in Figure 21. Properties of the film and friction between the stack and rollers was used as before. Since the coefficient of friction (both kinetic and static) between polymer/polymer interface is usually greater than the steel/polymer interface, the incoming stack of films has a very small thickness compared to the radius of the rollers and the polymer interfaces bond during plastic deformation, the incoming films were modeled as a single stack and therefore slipping and/or sliding between the film interfaces were neglected. The assumption of modeling the incoming stock as a single piece is validated later in this section. The rolling simulations were carried out until steady state force was noted. Rolling loads based on rigid-plastic analysis, for a desired level of plastic strain, were also computed.

The experimental loads, and those predicted by finite element simulations and perfect-plastic model are shown in Figure 22. The assumption of rigid-perfectly plastic model, assuming a constant yield stress, is found to greatly underestimate the actual compression loads to achieve a desired level of plastic-strain. The material films have a yield strength of $\sigma_{y,film} = 4$ MPa and modulus $E = 78$ MPa, thus allowing elastic strains ($\epsilon_e = \sigma_{y,film}/E_{film}$) up to 5%, which is a major contrast with respect to rigid-plastic model.
Figures 25 and 26 show the plots of von Mises stress and equivalent plastic strain, for a scenario in which approximately 13.5% nominal thickness reduction was achieved. Clearly, the plots indicate through-thickness (and almost homogeneous) plastic deformation. The plots of nominal strain and plastic strain in thickness reduction direction are also shown in Figures 27 and 28 respectively.

In the finite element simulation[6], we have treated and assumed the incoming stock as a single piece, however, in reality the stack is composed of several layers and in principle there can be slipping or sliding between the film layers. In order to verify the ‘no-slip’ assumption between the film layers, we plotted the variation of normal stress and shear stresses along the rolling direction, at the roller-film interface (where the shear stresses had maximum values). This is shown in Figure 30. The shear stresses were found to be significantly lower than the normal stresses, and much lower than the shear yield strength of the material. The coefficient of self and dry friction between polymers is usually high (0.5 – 0.8) and therefore our assumption of ignoring slip or sliding between layers is quite justified. Furthermore, during rolling and active plastic deformation, as bonding occurs in the rolling bite, the resistance to slip or sliding will increase.

To compare the overall accuracy of our simulations, we also compare the plots of total energy and kinetic energy, for the whole model, as function of the simulation time. Total energy should remain constant. Initially the kinetic energy of the model and total energy are both zero, and their magnitudes after long times, when steady states have been achieved, should be close to zero. The plots of the total energy and kinetic energy, normalized with respect to strain energy, shown in Figure 24. This excellent energy behavior suggests that explicit integration scheme based on central difference (using automatic time increments) and kinematic constraint algorithm are working accurately over long times. Loss of energy during contact/impact in dynamic problems can be significant [20, 104], but for our rolling cases the material velocities are negligible and therefore concerns related to dissipation during contact or explicit integration over long times do not arise.

It is worth emphasizing that the plastic rolling of a strip is an ideal model, and valid when the incoming strip is passed through rollers to produce appreciable reduction in thickness, such that the plastic deformations are large compared to the elastic deformations. The assumption of totally ignoring elastic strains, is an often well-suited idealization for metals, since elastic strains typically amount to only 0.5% or so. However, solid state polymers can exhibit elastic strains up to (or greater than) 5% and, therefore such an idealization is expected to yield unsatisfactory results. In the rigid-plastic model, the maximum nominal strain in thickness is observed at the location where the roller-gap is minimum. The contact zone in this case extends from the point where stack encounters the roller bite up to this minimum roller-gap location. In contrast, if the material has sufficient elasticity then for the same level...
of nominal plastic thickness reduction, the contact zone is much larger on the entry side and also extends further beyond the minimum roller-gap location on the exit side. For the above case, where a nominal plastic strain of 13.5% is achieved, we plot the normalized contact pressure based on the finite element simulation and rigid-plastic model, as shown in Figure 29. The minimum rolling gap location is at the zero coordinate value. Clearly the contact zone and normalized contact pressure for elasto-plastic analysis are significantly larger than the rigid-plastic scheme. This is the primary reason for large rolling loads encountered, which cannot be captured by the classical rolling theories. This aspect is necessarily relevant if polymer films, as those exhibiting hyper-elasticity and plasticity, are to be roll-bonded in a similar fashion.
Figure 21: The undeformed mesh in elastic-plastic rolling.
Figure 22: Line loading (N/mm) with respect to % plastic strain curves based on experimental data, and predictions from Kármán model and finite strain elastic plastic FEA analysis (both hypoelasticity and multiplicative plasticity).

Figure 23: Roll-bonding of polymeric films through the developed machine. The roller radius (R) is 100 mm. An initial stack of films 0.6 mm undergoing 13.5% nominal thickness reduction.
Figure 24: Normalization of energies has been done with respect to the strain energy of the model at any given time. Both kinetic energy and total energy were initially zero and after long time when steady-state has been reached they are still orders of magnitude smaller compared to the dominant strain energy in the model.
Figure 25: Von Mises stress during elastic-plastic rolling at steady state for approximately 13.5% nominal strain. The peak $S$, Mises is 8.611 MPa.
Figure 26: Equivalent plastic strain (PEEQ) during elastic-plastic rolling at steady state for approximately 13.5% nominal strain. See Supplementary Video S4.
Figure 27: Nominal strain (NE22) in direction of thickness reduction during elastic-plastic rolling at steady state for approximately 13.5% nominal strain.
Figure 28: Plastic strain (PE22) in direction of thickness reduction during elastic-plastic rolling at steady state for approximately 13.5% nominal strain.
Figure 29: Variation of normalized shear \( (T_{xy}) \) and normal stress \( (T_{xx}) \) along the rolling direction, at steady state for approximately 13.5\% nominal strain, between the roller and the film surface. The stresses have been normalized with respect to the yield strength of the film \( \sigma_{y,\text{film}} \). The shear stresses are maximum between the roller and the film interface, and zero on the symmetry plane in the thickness direction. The normal stresses were found to be almost uniform across the stock thickness.

Figure 30: Variation of contact pressure in the roller bite for 13.5\% nominal strain based on elasto-plastic finite deformation in ABAQUS and rigid-perfect-plastic model. The contact pressure has been normalized by yield strength of the material.
6 Conclusions

In this paper, we have presented a methodology for modeling roll-bonding of polymeric films in solid-state, at ambient temperatures, by subjecting them to active plastic deformation. An appropriate finite element analysis is set up for the rolling process for polymeric films, and this task is simplified by using a rate-independent model with isotropic hardening. For thickness reductions up to 20% we found that formulations based on a hypoelastic model and multiplicative decomposition of the deformation gradient into elastic and plastic part were appropriate to represent the rolling deformations. This indicates that at desired levels of strains, at which bonding occurs, elastic stretches are still small and no specialized hyperelastic/visco-plastic polymer behavior is required. The frictional behavior can also be captured appropriately by using an isotropic Coulombic friction in a rate-independent setting. The practical significance of our results are that, under the experimental conditions as presented, one can set up a simplified yet accurate and computationally faster finite element model for rolling analyses. In principle, advanced polymer models can also be calibrated and applied, however, the computational cost involved in various steps during the time-integration of these sophisticated material-models can be significant, without any foreseeable benefits in predictive capability, especially when we are interested in long-time steady-state rolling.

We used the predictions from finite element analysis to fabricate an experimental rolling-apparatus. Rolling apparatus is capable of triggering through-thickness (and approximately homogeneous) plastic deformation on the incoming stack of polymeric films, while incurring minimum deflections in the machine-parts. Homogeneous and through-thickness plastic deformation is achieved by choosing a large roll-radii compared to the stack thickness. Large roll-radii also minimize the shearing between the polymeric interfaces across which bonding is desired, and macroscopic rolling-loads can be predicted accurately without detailed modeling of the interfacial behavior. The proposed ‘C’ shaped symmetric roller-stand is shown to offer high-stiffness against the roll-separating force in the vertical-configuration. The compliant behavior of thin polymeric sheets, makes them susceptible to defects such as wrinkling or buckling in presence of a speed mismatch between the rollers, and therefore usage of belt-driven roller assembly was employed and found to work successfully. Several other key considerations were also taken into account in selecting other components of the rolling-apparatus.

Although analyses of rolling processes have been extensively carried out over the last century ever since the earliest work of Kármán in 1925, majority of these efforts have focused on applications related to metal deformation. A key distinctive feature of large deformation processes in metals lies in relatively small elastic strains compared to the overall strain, and thus the classical theories based on completely ignoring the elastic deformations for prediction of forming loads are often applicable. In the current case of polymeric films, the elastic deformations are non-negligible and lead to higher forming loads than those predicted by the classical theories. In such a scenario, the contact-width and contact-pressure in
the roller bite, comprising of both elastic and elastic-plastic regions, are substantially larger than those predicted by the rigid-plastic rolling scheme. Finite element simulations, taking elastic deformations into account, give an accurate prediction of the actual rolling loads.

It is expected that this study shall facilitate the design and fabrication of machines for deformation induced roll-bonding of polymers and/or (related) processes for a variety of polymer films (based on different formulations and mechanical properties). Development of advanced constitutive models for roll-bonding of polymers, prediction of accurate rolling-loads and evolution of micro-structure in rolling while taking into account rate and thermal effects, etc., can be undertaken as the next step. Multi-scale models from deformation induced diffusion and molecular dynamics can be attempted to develop an integrated framework for making predictions in cold-roll-bonding of polymers.

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Appendix

8.1 Rigid Plastic Analysis

The choice of compression rollers was considered to be the most critical component of our rolling-machine, since the rollers must be sized appropriately to apply desired levels of loads and induce (homogeneous) through-thickness plastic deformation to cause bonding. In general, during deformation the total strain comprises of both elastic and plastic strains, but in rigid-plastic rolling, we ignore the elastic strains assuming that the plastic strains will dominate. Such an idealization may be appropriate for rolling thin-strips of metals (which exhibit small elastic strains) but for polymers exhibiting noticeable elastic strains such a model may not accurately predict rolling loads. Furthermore, polymeric materials can also exhibit strong rate-sensitivity which can contribute to rolling loads and torques. Despite these limitations of the rigid-plastic model, it has a simple and analytical result and we have found it useful to set up our finite element simulations. The key elements of the classic rolling model due to Kármán are:

1. Plane strain: If the thickness of the rolled strip is much smaller than its width then conditions of plane strain prevail, and under these conditions the lateral spread (or increase in the width of the specimen is negligible).

2. Friction at roller and strip interface: It is assumed that the rolling stock slips between the rolls, and conditions of kinetic or dry frictional force exist at all locations on the periphery where the contact exists. At the inlet the speed of the rolling stock is smaller than the tangential velocity of the rollers; thus there is a forward drive due to the action of friction. As the stock proceeds forward it is compressed, due to which it gains speed (conservation of mass). At some point the relative speeds between the roller and stock is zero, and that point is known as neutral point. Due to the frictional drag, the equilibrium consideration implies that the longitudinal stress increases until the neutral point, where the relative velocity is zero, and from there, on when the friction acts in the opposite direction, the longitudinal stress decreases.

3. Elastic and plastic deformations: We have already stated that elastic (recoverable) deformation is neglected compared to the plastic deformation. It is also assumed that plastic deformation occurs at a constant yield or flow stress. The plastic flow is taken to be incompressible.

4. Rollers are rigid: The elastic deformation of the rollers is neglected, and hence rollers are taken to be rigid with constant radius. Any elastic deflection of the other tooling part is ignored.

5. Isotropic deformation: It is assumed that accumulation of plastic strain has no effect on isotropy of the material.

6. Homogeneous compression: It is assumed that plane sections remain plane, i.e., vertical segments only shorten in height and the material deformation is compensated by an increase in longitudinal length, in other words the role of shear stresses in deformation is ignored.
7. Rate insensitivity: The effects of strain rate on the yield stress of the material are neglected.

It should be noted that homogeneous deformation across the thickness does not necessarily imply that plane sections remain plane. The assumption of plane sections remaining plane, explicitly, ignores the effect of shear stresses.

As discussed earlier, Figure 5 exhibits a rigid-plastic rolling scheme in accordance with the von Kármán model, with a differential element, where the mean longitudinal (compressive) stress in the strip is denoted by $\bar{\sigma}_x$ and the transverse stress at the surface by $\bar{\sigma}_z$. From the geometry we have

$$d = \frac{a^2}{R},$$

(190)

where

$$2d = h_1 - h_o.$$  

(191)

The equilibrium of the differential element in vertical and horizontal direction gives:

$$\bar{\sigma}_z \, dx = (pcos\phi + qsin\phi)2Rd\phi$$

(192)

and,

$$d(h\bar{\sigma}_x) = (psin\phi - qcos\phi)2Rd\phi.$$  

(193)

The yield of most ductile materials is usually taken to be governed either by von Mises’ shear strain-energy criterion i.e.

$$\frac{1}{6}(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2$$

$$= k^2 = \sigma_y^2/3.$$  

Since we have assumed a rigid-plastic model, the von Mises yield criterion is satisfied all along the bite of the roller. Once the yield criterion is satisfied under multi-axial loading, the behavior of ductile materials can be described by the Levy-Mises equations, which relate the principal components of strain increments during plastic deformation to the principal applied stresses as

Since we have neglected the effect of shear stresses, and therefore $x$, $y$ and $z$ directions become the three principal directions. Due to plane strain condition along $y$-direction, $e_y = \delta e_y = \dot{\epsilon}_y = 0$. This in accordance with the aforementioned Levy-Mises flow rule leads to:

$$\bar{\sigma}_y = \frac{1}{2}(\bar{\sigma}_x + \bar{\sigma}_z).$$  

(194)

We remind the reader that bar quantities represent the average estimates under assumed homogeneous
deformation in rolling. This implies that stress in y-direction is the average of those in x and z directions. According to the von Mises yield criterion we can say that in plastic flow zone we have

\[ \sqrt{(\bar{\sigma}_x - \bar{\sigma}_y)^2 + (\bar{\sigma}_x - \frac{\bar{\sigma}_x + \bar{\sigma}_z}{2})^2 + (\bar{\sigma}_z - \frac{\bar{\sigma}_x + \bar{\sigma}_z}{2})^2} = Y, \]

\[ |\bar{\sigma}_z - \bar{\sigma}_x| = \sqrt{\frac{2}{3}} Y = \sqrt{2k} = 2k^*. \] (195)

Here, \( k^* \) is the effective shear yield strain ( \( k^* = k/\sqrt{2} \) ). Although we have assumed homogeneous state of stress in the element which is not the case at the surface, yet combining equations 192, 193 and 195 we get

\[ \frac{d}{d\phi} h(p + qtan\phi - 2k^*) = 2R(psin\phi - qcos\phi). \] (196)

This is also the exact form of von Kármán equation [117]. An analytical solution to this equation is not known without further simplifications. However an approximate scheme can be adopted as summarized in [49].

For relatively large rolls we assume \( sin\phi \approx \phi \) and \( cos\phi \approx 1 \) etc. and retain only first order terms in \( \phi \). The roll profile is then approximated by

\[ h \approx h_o + R\phi^2 \approx h_o + x^2/R. \] (197)

Making these approximations in (196) and neglecting the term \( qtan\phi \) compared with \( p \), and changing the position variable from \( \phi \) to \( x \) we get

\[ h \frac{dp}{dx} = 4k^* \frac{x}{R} + 2q. \] (198)

Typically when the coefficient of friction between rollers and strip is large, and/or the strip has low yield strength, the frictional traction at the interface exceeds the yield strength of the strip in shear, so that there is no slip in the conventional sense at the surface. Plastic shear will take place in the rolled stock, while the surface will “stick” to the rolls with static friction. It is worth emphasizing that the above procedure has incorporated no-slip assumption and homogeneous deformation.

As an approximation we can replace \( h \) by the mean thickness \( \bar{h} = \frac{1}{2}(h_o + h_i) \) and on the basis of no-slip or stick assumption we assume \( q \) reaches the shear yield stress throughout the contact arc. Then, equation 198 becomes

\[ \text{we have also used } h_1 \text{ for inlet height at some places (which is same as } h_i) \]
\[
\frac{dh}{dx} = 2k^* \left( \frac{2x}{R} \pm 1 \right). 
\]  
(199)

The positive sign applies to the entry region where the strip is moving slower than the rolls and the negative sign applies to the exit. Equation (199) can be integrated, with boundary conditions that \( \bar{\sigma}_x = 0 \) at entry and exit, to give the pressure distribution as:

At entry

\[
\frac{\bar{h}}{a} \left( \frac{p}{2k^*} - 1 \right) = (1 - x/a) - \frac{a}{R} \left( 1 - x^2/a^2 \right),
\]
and at exit

\[
\frac{\bar{h}}{a} \left( \frac{p}{2k^*} - 1 \right) = -x/a - \frac{ax^2}{Ra^2}.
\]

The pressure at the neutral point is common to both these equations, which locates that point at

\[
\frac{x_n}{a} = -\frac{1}{2} + \frac{a}{2R}.
\]

The line loading per unit width is then found to be

\[
\frac{P_l}{k^*a} = \frac{1}{k^*a} \int_{-a}^{0} p(x)dx \approx 2 + \frac{a}{h} \left( \frac{1}{2} - \frac{1}{3} \frac{a}{R} \right),
\]

and the moment applied to the rolls is found to be

\[
\frac{M}{k^*a^2} = \frac{1}{k^*a^2} \int_{-a}^{0} xp(x)dx \approx 1 + \frac{a}{4h} \left( 1 - \frac{a}{R} \right).
\]

References


