ASPECTS OF A FINITE STRAIN CONSTITUTIVE MODEL FOR SEMICRYSTALLINE POLYMERS

Thesis submitted by
David William Holmes BE (Hons) JCU
in October 2007

for the degree of Doctor of Philosophy
in the School of Engineering
James Cook University
Statement of Access

I, the undersigned, author of this work, understand that James Cook University will make this thesis available for use within the University Library and, via the Australian Digital Theses network, for use elsewhere.

I understand that, as an unpublished work, a thesis has significant protection under the Copyright Act and;

I do not wish to place any further restriction on access to this work.

_________________________________________  ______________
Signature                                      Date
Statement of Sources

I declare that this thesis is my own work and has not been submitted in any form for another degree or diploma at any university or other institution of tertiary education. Information derived from the published or unpublished work of others has been acknowledged in the text and a list of references is given.

_________________________  __________________________
Signature                                           Date
Electronic Copy

I, the undersigned, the author of this work, declare that the electronic copy of this thesis provided to the James Cook University Library is an accurate copy of the print thesis submitted, within the limits of the technology available.

____________________________________  _______________________
Signature                                              Date
List of Publications

Throughout the progression of this research there have been several international conference presentations and international journal articles. The chapters of this thesis largely resemble these publications following the plan outline here.

Chapter 2 was presented in part at the Australian and Korean Rheology Conference, 2005 (AKRC05). The chapter has been published in largely unaltered form under the citation


Chapter 3 was presented at the World Congress on Computational Mechanics 2006 (WCCM VII). The chapter has been submitted for journal publication in largely unaltered form under the citation


Chapter 4 has been submitted for journal publication in largely unaltered form under the citation

Chapter 5 has been presented in part at Computational Techniques and Applications Conference 2006 (CTAC-06) and the accompanying paper has been accepted for publication in the conference proceedings under the citation

Statement on the Contribution of Others

I, the undersigned, declare that this thesis is my own work and acknowledge the support and contributions of the following organizations and people:

- Stipend support and research expenses were provided through an industry scholarship from Gough Plastics Pty Ltd.

- Additional research expenses were funded through the James Cook University’s School of Engineering internal research allocation.

- A financial contribution toward travel expenses to the WCCM VII conference held in Los Angeles in 2006 was made by the James Cook University ‘Graduate Research International Travel Awards’.

- My supervisory team of Prof. Jeffrey Loughran and Dr. Harry Suehrcke contributed towards the editorial aspects of the journal papers associated with this research (as per the indicated authorship above) and the subsequent editing of this thesis.

_________________________________________  __________________________
Signature                                           Date
Acknowledgements

I would like to take this opportunity to thank the many people that have made completion of this research possible.

Firstly to my principal supervisor Prof. Jeffrey Loughran, without whose initial belief, continued support and guidance, none of this would have been possible.

To my other supervisor Dr. Harry Suehrcke for sharing his wealth of practical knowledge and for the helpful input with writing.

To the administrative staff in the School of Engineering for their assistance and good humor throughout the day to day workings of university life.

To ICO Courtenay for supply of materials testing samples and to Peter Lyngcoln, Stuart Petersen and Dr. Paul Britton for assistance with preliminary experimental testing.

To my friends whose good humor and support helped maintain my sanity throughout the years of study.

And finally and most significantly to my partner Sarah Butterworth for here love, support and seemingly endless tolerance of the long nights spent writing and to our families, who made us the people we are today and continue to inspire us to learn and grow.

I thank you all
Abstract

In this thesis, the development of a constitutive model for the finite strain deformation of semicrystalline polymers is presented. It reports on the formulation and numerical implementation of the model and the theoretical aspects of the associated experimental testing and parameter estimation.

Within both academia and industry to date, there exists no single constitutive model for semicrystalline polymers that is broadly accepted as representing the general case. This is in spite of the relatively complete scientific understanding of the material’s response and the increasing use of such materials where structural loading can be significant. Numerical representation of such materials conventionally involves over-simplification of response, largely necessitated by the limitations of current experimental testing methods. A complex constitutive theory is only as powerful as the experimental method from which its parameters are fit. As such, the objective of this research was to develop a complete, generalized constitutive theory for semicrystalline polymers with a corresponding testing methodology that enables its practical use within industry.

The constitutive model selected can be characterized by a parallel combination of elastic, viscoelastic and viscoplastic model elements which most closely represents the complete deformation behavior of semicrystalline polymers in the pre-necking region ($\varepsilon < 15\%$). The accompanying mathematics are formulated for 3D, finite strain and are based on thermodynamic dissipation in keeping with conventional continuum mechanics methodology. Strain hardening has been found to be of importance within the viscoplastic element. The parallel configuration of the three model elements facilitates the decoupled algorithmic treatment of each response. This has been carried out in principal space, given the assumption of isotropy, making practical both its numerical implementation and the physical determination of model parameters. A strategy analogous to classical return mapping is used for
solution of the viscoelastic evolution while a new, principal space, closest point projection
return mapping algorithm has been developed for solution of the viscoplastic evolution,
accounting for isotropic strain hardening. The consistent algorithmic tangential modulus is
formulated to ensure quadratic convergence of the whole implicit finite element procedure.

The computational model has been verified through a series of simple finite element
tests involving combinations of large strain normal and shear loadings, and large rigid body
rotations. Several example problems have been solved as demonstration of the models
versatility.

Using the developed model, a study using numerical simulations of uniaxial and biax-
ial tensile testing methods has been carried out. Through this study it has been possi-
ble to develop an experimental methodology to isolate the component stress contributions
from each of the three deforming modes as well as subset separation of viscous, yield, and
isotropic hardening stresses for viscoplasticity. Via conventional optimisation procedures
and an additionally developed iterative procedure for the viscoelastic response, this testing
methodology makes possible the full specification of the model parameter set. Verification
of the testing methodology was done via comparison between the calculated test curves,
and values output directly from the numerical simulations.

The model proposed in this thesis corresponds to a general account of semicrystalline
polymer constitutive response, possessing capabilities not accounted for previously in the-
ories within the literature. Perhaps the most significant outcome from this work is the
experimental data processing methodology that allows such a complex model to be accu-
rately and practically fit to real materials. Being able to better predict the loaded response
of semicrystalline polymers is critical for their continued and increased use in circumstances
where structural loads are possible.
Contents

Statement of Access ii
Statement of Sources iii
Electronic Copy iv
List of Publications v
Statement on the Contribution of Others vii
Acknowledgements viii
Abstract ix
List of Tables xv
List of Figures xvii

1 Introduction 1
  1.1 Motivation ...................................................... 1
  1.2 Objectives .................................................... 2
  1.3 Outline ....................................................... 3

2 Constitutive Model Formulation 4
  2.1 Background .................................................... 4
  2.2 Review of literature ......................................... 6
    2.2.1 Direction from micromechanics ......................... 6
    2.2.2 Macroscopic constitutive theories ..................... 8
2.2.3 Model selection .................................................. 14
2.3 Suitability of the selected rheological form to polymers ............ 16
  2.3.1 Static loading .................................................. 17
  2.3.2 Constant strain rate loading .................................. 18
  2.3.3 Other phenomena .............................................. 23
2.4 Formulation of constitutive mathematics ................................ 24
  2.4.1 Kinematics and thermodynamics ............................... 25
  2.4.2 Stress ........................................................ 28
  2.4.3 Viscoelastic evolution equations .............................. 29
  2.4.4 Viscoplastic evolution equations .............................. 31
2.5 Summary .......................................................... 34

3 Numerical Implementation and Verification ................................. 35
  3.1 Generalized elasticity ............................................. 36
    3.1.1 Volumetric-deviatoric strain separation ...................... 37
    3.1.2 Thermodynamics in tensor space ............................. 38
    3.1.3 Spectral decomposition of strain ........................... 39
    3.1.4 Stress expression in principal stretches ..................... 42
    3.1.5 Closed-form tangential modulus expression in principal stretches .... 44
  3.2 Extension for generalized inelasticity ............................. 48
    3.2.1 Alternate treatment of the deformation gradient ............ 48
    3.2.2 Implications for tensor space thermodynamics ............. 49
    3.2.3 Spectral decomposition of elastic strain .................... 50
    3.2.4 Stress expression in principal stretches .................... 52
    3.2.5 Closed-form tangential modulus expression in principal stretches .... 53
  3.3 Principal space algorithmic development of the three specific cases .. 57
    3.3.1 Elastic element ............................................ 58
    3.3.2 Viscoelastic element ....................................... 59
    3.3.3 Viscoplastic element ....................................... 63
  3.4 Numerical verification ............................................ 69
    3.4.1 Single element normal tests ................................ 69
    3.4.2 Simple shear tests ......................................... 70
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.4.3 Simply supported beam: Creep</td>
<td>73</td>
</tr>
<tr>
<td>3.4.4 Simply supported beam: Relaxation</td>
<td>74</td>
</tr>
<tr>
<td>3.5 Summary</td>
<td>76</td>
</tr>
<tr>
<td>4 Development of Testing Methodology</td>
<td>77</td>
</tr>
<tr>
<td>4.1 3D results from uniaxial testing</td>
<td>78</td>
</tr>
<tr>
<td>4.2 Isolation of viscoelastic component stress</td>
<td>81</td>
</tr>
<tr>
<td>4.3 Separation of elastic and viscoplastic component stresses</td>
<td>84</td>
</tr>
<tr>
<td>4.3.1 Initial elastic stress estimate</td>
<td>86</td>
</tr>
<tr>
<td>4.3.2 Testing regime</td>
<td>87</td>
</tr>
<tr>
<td>4.3.3 The viscoplastic element modulus</td>
<td>89</td>
</tr>
<tr>
<td>4.3.4 Elastic and viscoplastic stress and strain components</td>
<td>92</td>
</tr>
<tr>
<td>4.4 Measurement of subset viscoplastic viscous, yield and hardening stresses</td>
<td>96</td>
</tr>
<tr>
<td>4.5 Summary</td>
<td>100</td>
</tr>
<tr>
<td>5 Parameter Estimation</td>
<td>101</td>
</tr>
<tr>
<td>5.1 Levenberg-Marquardt optimization for hyperelasticity</td>
<td>102</td>
</tr>
<tr>
<td>5.2 A modified Levenberg-Marquardt technique for nonlinear viscoelasticity</td>
<td>107</td>
</tr>
<tr>
<td>5.3 Estimation of viscoplastic element parameters</td>
<td>112</td>
</tr>
<tr>
<td>5.4 Generalization of the modified Levenberg-Marquardt method</td>
<td>115</td>
</tr>
<tr>
<td>5.5 Summary</td>
<td>117</td>
</tr>
<tr>
<td>6 Conclusions</td>
<td>118</td>
</tr>
<tr>
<td>6.1 Discussions on the proposed theories</td>
<td>118</td>
</tr>
<tr>
<td>6.2 Critiques and limitations</td>
<td>120</td>
</tr>
<tr>
<td>6.3 Future directions</td>
<td>121</td>
</tr>
<tr>
<td>6.4 Conclusion</td>
<td>121</td>
</tr>
<tr>
<td>A Derivation of the dissipation requirement</td>
<td>132</td>
</tr>
<tr>
<td>B Viscoplastic constitutive equations in 1D</td>
<td>137</td>
</tr>
<tr>
<td>C Principal Space Differentials</td>
<td>142</td>
</tr>
<tr>
<td>C.1 Modified Lagrangian eigenvalue base definition</td>
<td>142</td>
</tr>
<tr>
<td>C.2 First principal stretch differentials</td>
<td>143</td>
</tr>
</tbody>
</table>
C.3 First and second jacobian differentials ........................................... 146
C.4 Second principal stretch differentials ............................................. 147

D Expansions of $\bar{C}$ ................................................................. 150
D.1 Expansion of $\bar{C}$ for double coalescence of eigenvalues ................. 150
D.2 Expansion of $\bar{C}$ for triple coalescence of eigenvalues ................. 151
List of Tables

2.1 Arbitrarily chosen 1D linear model parameters . . . . . . . . . . . . . . . . 17

3.1 Material parameters for simple shear testing . . . . . . . . . . . . . . . . . 71

5.1 Effect on the nature of the optimization equation set as subsequent data points are included in the calculations . . . . . . . . . . . . . . . . . . . . . . . . . . . 111
## List of Figures

2.1 One-dimensional elasto-viscoelastic-viscoplastic rheological models .... 9  
2.2 Generalized elasto-viscoelastic-plastic-viscoplastic model ............ 13  
2.3 One-dimensional elasto-viscoelastic-viscoplastic rheological model .... 15  
2.4 Theoretical component strain curves ................................ 20  
2.5 Theoretical total stress strain curves ................................ 21  
2.6 Experimental total stress strain curves taken from the literature ....... 22  
2.7 The multiplicative split of the deformation gradient .................. 26  
3.1 1D rheological representation of the viscoelastic element ............ 60  
3.2 1D rheological representation of the viscoplastic element ............ 63  
3.3 Single element geometry for normal testing ........................... 69  
3.4 Four element simple shear test geometry .............................. 70  
3.5 Shear stress vs shear strain for cyclic simple shear test ............ 71  
3.6 Isotropic hardening variable during cyclic simple shear test ........ 72  
3.7 Load applied to simply supported beam during creep test ............ 73  
3.8 The creep over time of the simply supported beam .................... 74  
3.9 Displacement applied to simply supported beam during relaxation test 75  
3.10 Relaxation of stress at the center of the simply supported beam .... 75  
4.1 3D test data from uniaxial experiments ................................ 80  
4.2 Strain profile for cyclic load-unload test ............................. 82  
4.3 Stress strain curve for a single conditioning loop .................... 83  
4.4 Cyclic stress strain curve for viscoelastic isolation ................... 84  
4.5 Deviatoric strain during stress relaxation ............................ 85  
4.6 Elastic stress curve ranges ........................................... 87
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.7</td>
<td>Load-unload-recovery testing</td>
<td>88</td>
</tr>
<tr>
<td>4.8</td>
<td>Loading-unloading-relaxation testing</td>
<td>89</td>
</tr>
<tr>
<td>4.9</td>
<td>$X$ diagram</td>
<td>90</td>
</tr>
<tr>
<td>4.10</td>
<td>Calculation of the viscoplastic stress component</td>
<td>91</td>
</tr>
<tr>
<td>4.11</td>
<td>Calculation of the change in viscoplastic stress</td>
<td>92</td>
</tr>
<tr>
<td>4.12</td>
<td>Calculation of multiple changes in viscoplastic stress</td>
<td>93</td>
</tr>
<tr>
<td>4.13</td>
<td>Average viscoplastic modulus</td>
<td>93</td>
</tr>
<tr>
<td>4.14</td>
<td>Calculation of the plastic component of viscoplastic strain</td>
<td>95</td>
</tr>
<tr>
<td>4.15</td>
<td>Comparison between the calculated and the actual stress curves</td>
<td>97</td>
</tr>
<tr>
<td>4.16</td>
<td>Subset components of viscoplastic stress</td>
<td>98</td>
</tr>
<tr>
<td>B.1</td>
<td>1D elasto-viscoplastic rheological element</td>
<td>137</td>
</tr>
<tr>
<td>B.2</td>
<td>Representation of the elastic limit</td>
<td>140</td>
</tr>
</tbody>
</table>