Thermomechanical Modelling of Microstructure Evolution in Solder Alloys

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Thermomechanical Modelling of Microstructure Evolution in Solder Alloys

PROEFSCHRIFT

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The reliability of soldered connections is a very important issue in electronics industry. This project aims to better understand the various factors influencing the lifetime of solder joints through numerical modelling. Because of the high homologous temperatures at which they operate, the joints exhibit high temperature deformation mechanisms associated with creep and relaxation and are susceptible to low cycle fatigue. Another issue is that due to the ongoing miniaturisation, microstructural sizes influence the material properties greatly.

The project was initiated with the commercial eutectic tin–lead solder in mind. For this alloy the microstructure evolves, coarsens, significantly over time. To accurately capture the various, and sometimes very large, time scales that come into play, accelerated test methods are not suitable and real time testing is too time consuming. This is where numerical simulations can provide more insight and permit a significant reduction in cost and time of the design of highly reliable soldered connections in electronic packages and components.

The first part of this thesis deals with the microstructure evolution of tin–lead due to diffusion. In the following two parts the proposed model is extended with respectively viscoplastic material behaviour and a nonlocal damage approach. In the final part attention is given to the tin–silver–copper system, which is one of the most likely replacement candidates of tin–lead in the strive towards a lead-free electronics industry.

The evolving microstructure has been taken into account using a phase field model which is solved using the finite element method. The driving force for diffusion has been derived from a macroscopic free energy function, which describes phase segregation in microscopic model systems with long-range interactions evolving according to stochastic Kawasaki dynamics with nearest neighbour exchanges. Simulations of the static ageing process of eutectic tin–lead solder have been performed. The results predict break-up, coalescence, growth, and dissolution of phases, similar to experimental observations. It was also shown that external mechanical loading leads to faster coarsening rates. Quantitative comparison with experiments has been performed using the total interface length as the quantifying parameter and a good agreement was found.

To capture the time dependent mechanical behaviour an elasto-viscoplastic material model law been used for the material model. The extensive information on the microstructure found with the phase field model was used to assign different parameter values to the individual phases and interfaces. Results from simulations of mechanical loading of eutectic tin–lead solder showed a strong dependency on the underlying microstructure.
Aged microstructures exhibit more pronounced localisation of stresses and strains.

To investigate the reliability of the solder, the model has been extended to include damage. The modelling of softening behaviour often leads to bad solutions using the finite element method. Although the viscous nature of the material model is known to regularise the solution, for practical purposes this effect is usually only sufficient for highly rate-sensitive materials or high loading rates and the numerical results still can show a mesh dependency. Therefore, a gradient enhanced nonlocal damage formulation has been implemented. The results of the phase field model are used to assign different damage parameter values to the phases and interfaces. For the tin–lead system the phase boundaries are known to be the crack initiation sites. The cracks next propagate preferably along tin–lead or tin–tin grain boundaries. The approach yields results that are qualitatively comparable with these experimental findings.

Because, in the electronics industry, the tin–lead alloy needs to be replaced with a lead-free alternative in the near future, the final chapter deals with one of the most likely candidates, the near eutectic tin–silver–copper. Experiments performed on this ternary alloy revealed a disconcerting feature. Cyclic thermal ageing without any additional mechanical loading was already enough to lead to fracture along grain boundaries. In order to understand and explain this behaviour the experiments have been modelled using a three-dimensional finite element approach. The viscoplastic damage part of the model is extended to account for anisotropy of the material, both in the elastic as well as the thermal properties. Data obtained from Orientation Image Microscopy is used to take into account the microstructure at the grain level, which was found not to evolve over time. The results show a good qualitative agreement with the experiments, exhibiting stress concentrations leading to damage along the grain boundaries.

The presented modelling approaches have been applied to simulate the complex behaviour of solder alloys. A multiphase alloy who’s mechanical behaviour is determined by its evolving microstructure is modelled and the results are compared with experimental data, showing satisfactory agreement. Furthermore, an industrially interesting material, near eutectic tin–silver–copper, has been successfully investigated, predicting damage in the same areas as seen experimentally, indicating that the elastic and thermal anisotropic properties play an important part in the fatigue life of this alloy.
Samenvatting

Soldeerverbindingen worden in de micro-elektronica industrie toegepast om componenten te voorzien van zowel een elektrische als mechanische verbinding. De betrouwbaarheid van soldeerverbindingen is daarom van groot belang. Dit project is erop gericht door middel van modelvorming en numerieke simulaties, een beter inzicht te verkrijgen in de verschillende factoren die van invloed zijn op de levensduur van de verbinding.

Door hun lage smeltpunt en relatief hoge gebruikstemperatuur zijn soldeerverbindingen onderhevig aan hoge temperatuur deformatiemechanismen, zoals kruip. Hierdoor zijn ze vatbaar voor falen ten gevolge van vermoeiing na kort durende cyclische belasting. Verder speelt de voortdurende miniaturisatie een rol, doordat de microstructurele afmetingen de materiaal eigenschappen beïnvloeden.

Het project was gestart met in gedachte het tot dan toe veelgebruikte commercieel verkrijgbare eutectische tin–lood soldeer. De microstructuur van deze legering is continu onderhevig aan aanzienlijke veranderingen. Door tijdgebrek worden experimentele tests naar het faalgedrag van soldeerverbindingen vaak versneld. Resultaten verkregen met deze testmethoden moeten echter kritisch worden geëvalueerd, daar –door de verschillende tijdschalen die een rol spelen– zij vaak niet alle fysische processen op de juiste manier omvatten. Op dit punt kunnen numerieke simulaties meer inzicht verschaffen en uiteindelijk kunnen leiden tot vermindering van de tijd en kosten verbonden met het ontwerpen van betrouwbare soldeerverbindingen in elektronische componenten.


De veranderende microstructuur wordt beschreven aan de hand van een ‘phase-field’ model, dat is geïmplementeerd in een eindige elementen programma. De drijvende kracht achter de diffusie is afgeleid van een macroscopische vrije energie functie, die fasesegregatie in microscopische modelsystemen met langeafstandsinteracties beschrijft. Met het model zijn simulaties van statische verouderingsexperimenten van tin–lood soldeer verricht. De resultaten voorspellen fasescheiding, -vereniging, -groei en -oplossing, soortgelijk aan wat er in de experimenten geobserveerd was. Het model voorspelt verder dat externe belasting leidt tot snellere microstructuurveranderingen. Een kwantitatieve vergelijking
met de experimenten, aan de hand van de totale lengte van de scheidingsvlakken tussen
de fases, laat een goede overeenstemming zien.

Om het tijdsafhankelijke mechanische gedrag te beschrijven, wordt het elasto-viscoplasti-
tisch Perzyna materiaalmodel gebruikt. De uitgebreide informatie over de microstructuur
verkregen met het ‘phase-field’ model wordt gebruikt om verschillende parameters toe te
wijzen aan de individuele fasen en de scheidingsvlakken ertussen. De resultaten van simu-
laties van mechanische belasting tonen een sterke afhankelijkheid van de onderliggende
microstructuur. Een microstructuur waarin de fasescheiding verder gevorderd is vertoond
een meer uitgesproken lokalisatie van spanningen en rekken.

Om de betrouwbaarheid van het solderen te onderzoeken is het model verder uitgebreid
met een schadeformulering. De modellering van materiaalverzwakking leidt vaak tot
irreële oplossingen in een eindige elementen methode. Hoewel het viskeuze gedrag
van het materiaal de oplossing in enige mate regulariseert, is dit effect in de praktijk
alleen voldoende voor zeer snelheidsafhankelijke materialen, of in geval van zeer snelle
belastingen. Daarom is gekozen voor een niet-lokale schadeformulering, welke deze
problemen oplost door de introductie van een lengteschaal. Ook hier worden de resultaten
van het ‘phase-field’ model gebruikt om de verschillende fasen en de grensvlakken van
hun eigen parameterwaarden te voorzien. Van het tin–lood solderen is bekend dat de
grensvlakken tussen de fasen zwakke plekken zijn waar schade vaak begint. De scheuren
planten zich vervolgens bij voorkeur voort langs de tin-lood en tin-tin korrel grenzen.
De gevolgde aanpak levert resultaten die kwalitatief overeenstemmen met experimentele
bevindingen.

Omdat de tin–lood legering in de electronica industrie vervangen moet worden door loodv-
rije alternatieven in de nabije toekomst, wordt in het laatste deel aandacht geschonken aan
de tin–zilver–koper soldereigenschappen. Experimenten toonden aan dat voor dit materiaal cyca-
lische thermische belasting alleen al tot schade leidt op en in de buurt van de grensvlakken
tussen de korrels. Om dit gedrag te begrijpen zijn de experimenten gemodelleerd in een
drie-dimensionale eindige elementen omgeving. De viscoplastische schadeformulering is
aangepast om ook de richtingsafhankelijkheid van het materiaalgedrag omvatten, zowel
in de elastische als in de thermische eigenschappen. Gegevens verkregen via Orientation
Image Microscopy is gebruikt om de microstructuur op korrelniveau mee te nemen in
de berekeningen. De resultaten laten een goede kwalitatieve overeenkomst zien met de
experimenten. Spanningsconcentraties die uiteindelijk tot schade leiden treden op rond
de korrelgrenzen.

De voorgestelde modelleermethoden zijn toegepast om het ingewikkelde gedrag van soldeer-
legeringen te beschrijven. Een legering, wiens mechanische gedrag wordt bepaald door
de verschillende fasen waaruit de microstructuur is opgebouwd, is gemodelleerd en de
resultaten zijn vergeleken met experimentele observaties. Een toereikende overeenkomst is
gevonden. Verder is er ook nog gekeken naar een voor de industrie interessant materiaal, de
tin–zilver–koper legering. De resultaten tonen aan dat de richtingsafhankelijke elastische
een thermische eigenschappen een belangrijke rol spelen in het vermoeingsgedrag van deze
legering.
Notations and definitions as they are used in this thesis are summarised below. In the definitions, a Cartesian basis \(\vec{e}_1, \vec{e}_2, \vec{e}_3\) is used, and the Einstein summation convention is adopted.

### Quantities

<table>
<thead>
<tr>
<th>Type</th>
<th>Notation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>scalar</td>
<td>( \alpha; a; A )</td>
<td>scalar</td>
</tr>
<tr>
<td>vector</td>
<td>( \vec{a} = a_i \vec{e}_i )</td>
<td>vector</td>
</tr>
<tr>
<td>non-local variable</td>
<td>( \bar{a} = a_{i\bar{i}} \vec{e}_i )</td>
<td>time derivative</td>
</tr>
<tr>
<td>history value</td>
<td>( \bar{a} )</td>
<td></td>
</tr>
<tr>
<td>time derivative</td>
<td>( \dot{a} )</td>
<td></td>
</tr>
<tr>
<td>Truesdell rate</td>
<td>( \bar{A} = \dot{A} - L \cdot A - A \cdot L^c )</td>
<td></td>
</tr>
<tr>
<td>second-order tensor</td>
<td>( A = A_{ij} \vec{e}_i \vec{e}_j )</td>
<td></td>
</tr>
<tr>
<td>identity tensor</td>
<td>( I = \delta_{ij} \vec{e}_i \vec{e}_j )</td>
<td></td>
</tr>
<tr>
<td>fourth-order identity tensor</td>
<td>( I = \delta_{il} \delta_{jk} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td>fourth-order tensor</td>
</tr>
<tr>
<td>column</td>
<td>( \bar{a}_i )</td>
<td></td>
</tr>
<tr>
<td>matrix</td>
<td>( \bar{A} )</td>
<td></td>
</tr>
</tbody>
</table>

### Operations

<table>
<thead>
<tr>
<th>Operation</th>
<th>Notation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>scalar multiplication</td>
<td>( c = ab ); ( \bar{c} = a \bar{b} ); ( \bar{c} = a \bar{b} )</td>
<td></td>
</tr>
<tr>
<td>dyadic product</td>
<td>( \vec{a} \vec{b} = a_i b_j \vec{e}<em>i \vec{e}<em>j ); ( \bar{A} \vec{B} = A</em>{ij} \bar{B}</em>{kl} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td></td>
</tr>
<tr>
<td>inner product</td>
<td>( c = \bar{a} \vec{b} = a_i b_i ); ( C = A \cdot B = A_{ij} \bar{B}_{kl} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td></td>
</tr>
<tr>
<td>double inner product</td>
<td>( c = A : B = A_{ij} \bar{B}<em>{ji} ); ( \bar{C} = A : B = A</em>{ij} \bar{B}_{ji} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td></td>
</tr>
<tr>
<td>conjugate / transpose</td>
<td>( C^c = C_{ji} \vec{e}_i \vec{e}<em>j ); ( \bar{C}^c = \bar{C}</em>{ ji} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td></td>
</tr>
<tr>
<td>right conjugate</td>
<td>( C^{rc} = C_{ijlk} \vec{e}_i \vec{e}_j \vec{e}_k \vec{e}_l )</td>
<td></td>
</tr>
<tr>
<td>left conjugate</td>
<td>( C^{lc} = C_{ijlk} \vec{e}_j \vec{e}_k \vec{e}_l \vec{e}_i )</td>
<td></td>
</tr>
<tr>
<td>inverse</td>
<td>( \bar{A}^{-1} )</td>
<td></td>
</tr>
<tr>
<td>determinant</td>
<td>( \det(A) = (A \cdot \vec{e}_1) \cdot (A \cdot \vec{e}_2) \times (A \cdot \vec{e}_3) )</td>
<td></td>
</tr>
<tr>
<td>deviatoric part</td>
<td>( A^d = A - \frac{1}{3} A : I )</td>
<td></td>
</tr>
</tbody>
</table>
gradient operator
\[ \vec{\nabla} = \vec{e}_i \frac{\partial}{\partial x_i} \]

Laplacian
\[ \nabla^2 = \vec{\nabla} \cdot \vec{\nabla} = \vec{e}_i \frac{\partial}{\partial x_i} \left( \vec{e}_i \frac{\partial}{\partial x_i} \right) = \frac{\partial^2}{\partial x_i^2} \]

variation and derivative
\[ \delta A_{ij} = \frac{\delta A_{ij}}{\delta B_{kl}} \delta B_{lk} \vec{e}_i \vec{e}_j \]

Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
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<tbody>
<tr>
<td>c</td>
<td>mass fraction</td>
<td>[-]</td>
</tr>
<tr>
<td>b</td>
<td>damage evolution rate</td>
<td>[-]</td>
</tr>
<tr>
<td>B</td>
<td>left Cauchy–Green strain tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>C</td>
<td>right Cauchy–Green strain tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>D</td>
<td>deformation rate tensor</td>
<td>[s(^{-1})]</td>
</tr>
<tr>
<td>(e_\Delta)</td>
<td>incremental Almansi strain tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>E</td>
<td>Young’s modulus</td>
<td>[Pa]</td>
</tr>
<tr>
<td>E</td>
<td>Green–Lagrange strain tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>F</td>
<td>deformation gradient tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>g</td>
<td>weighting function</td>
<td>[-]</td>
</tr>
<tr>
<td>(g_i)</td>
<td>free energy coefficients</td>
<td>[J/m(^3)]</td>
</tr>
<tr>
<td>G</td>
<td>Lamé’s constant, shear modulus</td>
<td>[Pa]</td>
</tr>
<tr>
<td>(h_i)</td>
<td>hardening parameters</td>
<td>[Pa] or [-]</td>
</tr>
<tr>
<td>(H)</td>
<td>fourth-order material tensor</td>
<td>[Pa]</td>
</tr>
<tr>
<td>(j)</td>
<td>mass flux</td>
<td>[kg/(m(^2) s)]</td>
</tr>
<tr>
<td>J</td>
<td>determinant of (F)</td>
<td>[-]</td>
</tr>
<tr>
<td>K</td>
<td>Lamé’s constant</td>
<td>[Pa]</td>
</tr>
<tr>
<td>(\ell_c)</td>
<td>diffusion length parameter</td>
<td>[m]</td>
</tr>
<tr>
<td>(\ell_{\omega})</td>
<td>damage length parameter</td>
<td>[m]</td>
</tr>
<tr>
<td>M</td>
<td>molar weight</td>
<td>[kg/mole]</td>
</tr>
<tr>
<td>(M)</td>
<td>mobility tensor</td>
<td>[m(^5)/(J s)]</td>
</tr>
<tr>
<td>L</td>
<td>velocity gradient tensor</td>
<td>[-]</td>
</tr>
<tr>
<td>(\hat{n})</td>
<td>normal unit vector</td>
<td>[-]</td>
</tr>
<tr>
<td>N</td>
<td>rate sensitivity parameter</td>
<td>[-]</td>
</tr>
<tr>
<td>(N)</td>
<td>plastic flow direction</td>
<td>[-]</td>
</tr>
<tr>
<td>(\ddot{p})</td>
<td>force vector</td>
<td>[N/m(^2)]</td>
</tr>
<tr>
<td>t</td>
<td>time</td>
<td>[s]</td>
</tr>
<tr>
<td>T</td>
<td>temperature</td>
<td>[K]</td>
</tr>
<tr>
<td>(\vec{v})</td>
<td>velocity</td>
<td>[m/s]</td>
</tr>
<tr>
<td>(\vec{x})</td>
<td>coordinate vector</td>
<td>[m]</td>
</tr>
<tr>
<td>(\vec{y})</td>
<td>coordinate vector</td>
<td>[m]</td>
</tr>
<tr>
<td>z</td>
<td>damage driving parameter</td>
<td>[J/m(^3)]</td>
</tr>
<tr>
<td>(\alpha)</td>
<td>coefficient of thermal expansion tensor</td>
<td>[K(^{-1})]</td>
</tr>
<tr>
<td>(\gamma)</td>
<td>gradient energy coefficient</td>
<td>[J/m]</td>
</tr>
<tr>
<td>(\Gamma)</td>
<td>surface, boundary</td>
<td>[m(^2)], [m]</td>
</tr>
<tr>
<td>(\epsilon)</td>
<td>strain tensor</td>
<td>[-]</td>
</tr>
</tbody>
</table>
Notations and conventions

$\varepsilon_{vp}$ effective viscoplastic strain [-]

$\eta$ fluidity [s$^{-1}$]

$\kappa$ interface tension coefficient [J/m$^3$]

$\lambda$ viscoplastic multiplier [-]

$\mu$ chemical potential [J/m$^3$]

$\nu$ Poisson's ratio [-]

$\phi$ overstress function [-]

$\varphi$ yield function [Pa]

$\rho$ density [kg/m$^3$]

$\sigma$ Cauchy stress tensor [Pa]

$\tau$ Kirchhoff stress tensor [Pa]

$\tau_{eq}$ equivalent Kirchhoff stress [Pa]

$\tau_y$ yield stress [Pa]

$\Phi$ free energy [J/m$^3$]

$\chi_{ij}$ free energy coefficient [J/m$^3$]

$\psi$ $\partial \Phi_i / \partial c$ [J/m$^3$]

$\omega$ damage parameter [-]

$\Omega$ volume, area [m$^3$], [m$^2$]

$\Omega$ spin tensor
Chapter 1

Introduction

1.1 Solder and reliability

Soldering in its most general form is a technique for joining metal parts using a filler material of a low melting or liquidus point. The solder is placed at the joining position and heated, directly or indirectly through the parts that will be joined, to above its liquidus temperature, causing it to melt and wet the parts. After the heat source is removed the solder solidifies and the parts are joined.

As opposed to pure metals solder alloys usually do not have a single melting point, but a melting range, bounded by the solidus and the liquidus temperature, i.e. the temperatures below which the alloy is completely solid and above which it is fully fluid. A few special alloy compositions do have a specific melting point, their liquidus and solidus temperature coincide. These are the eutectic compositions, and their melting temperatures are known as eutectic points. Wetting refers to the interaction that must take place between the liquid solder and the solid surfaces to ensure satisfactory bonding. By definition, wetting is a measure of the material, generally a liquid, to spread over a solid.

For thousands of years, soldering has been used to create a metallurgical bond. The Egyptians were using hard soldering, where the joining material has a liquidus temperature above 300°C, as far back as 5500 years. Other civilisations are also known to have used soldering, the ancient Greeks, Celts, and Gauls, and of course the Romans who introduced tin–lead (Sn–Pb) solder, the most widely used solder alloy till recently. In these early applications the function of the soldered connection was only to provide a mechanical bond, but in the late 19th century the joints also started to be exploited for their electrical conductivity. In the 20th century microelectronics appeared on the scene and a third function was added, namely, thermal connectivity. The joint serves as a path through which heat from an electronic package can be evacuated. The ever decreasing size of packages and increasing power consumption presented new challenges for manufacturing reliable joints. The work presented in this thesis focuses on the use of solder in microelectronics.

Today, soldered connections play a crucial role in the microelectronics industry. Level 1 packaging, the bonding of the silicon chip, used to be primarily accomplished using the wire bonding technique. A metal wire is welded with one end to the die and with the other
to the substrate, thus creating an electrical connection, Fig. 1.1a. However, the use of solder balls in level 1 packaging is becoming technically more attractive, since a higher number of connections can be made in a given area. Fig. 1.1b shows a flip-chip configuration, so called because the silicon die is turned upside down with respect to the substrate. The die is then attached to the substrate by a regular grid of solder bumps, a ball grid array (BGA). For these purposes, joint dimensions can be as small as 50-100 µm. Solder joints are the predominate connection type for level 2 packaging, the connecting of a component to a printed circuit board (PCB), either using a pin-through-hole configuration or surface mount technology (SMT), see Figs. 1.1, 1.2, and 1.3.

Because soldered connections play such an important role in microelectronics, their reliability is a major issue. The lifespan of a solder connection often determines the lifetime of an apparatus. Several factors and developments have a major influence on the reliability of the connections.

First, temperature: High operational temperatures are reached in electronic packages. The ever increasing functionality and chip density on printed circuit boards, leads to higher energy fluxes and thus higher temperatures. Soldered connections are also being used in harsher environments, e.g. in the automotive industry, where joints close to the engine can be subjected to temperatures up to 150°C. This means that the joints exhibit high temperature deformation mechanisms associated with creep and relaxation and are susceptible to low cycle fatigue.

Figure 1.1 — Schematics of level 1 and level 2 packaging.
Second, **thermomechanical cycling**: The on and off switching of a system causes components to alternately heat up and cool down, leading to cyclic thermomechanical loading of the solder connections. Mechanical stresses and strains arise in the joint because of the mismatch between the coefficients of thermal expansion (CTE) of the electronic component, the solder interconnect, and the board or substrate. Additional mechanical loading may also come from other sources, for instance bending of the printed circuit board, vibrations of an engine, etc.

Third, **size**: Due to ongoing miniaturisation in microelectronics the solder configurations are being pushed to their limit. For example, Sn–Pb solder bumps are already being produced at sizes smaller than their natural radius of curvature of the liquid solder [Wassink and Verguld, 1985], as determined by the surface tension. This might lead to bad and/or vulnerable connections because of the high internal liquid pressure.

Fourth, **microstructure**: Many different soldering techniques are being applied in the microelectronics industry: wave soldering, re-flow soldering, infrared soldering, ultrasonic soldering, etc. A characteristic of all these technologies is that the rapid cooling rate, differences in thermal properties of the components being connected, together with inhomogeneities during cooling have a large influence on the microstructure of the alloy. Because of this, residual stresses can arise within a joint and the connected components that can have an adverse effect on the reliability. Combined with the fact that typical sizes of the connections keep decreasing, the microstructural length scales are now nearly of the same order as the joint dimensions, see Fig. 1.4.

A single, but usually a combination of the above factors can eventually cause cracking in the solder, delamination between solder and components, or even cracking of components, often triggered by low cycle fatigue. This has initiated the existing technological and scientific interest in the thermomechanical, fracture and fatigue properties of solders.

The reliability of the connections is often investigated experimentally, using accelerated test methods because of time limitations. In these experiments the fatigue process is accelerated by applying either a higher mechanical load, temperature, or frequency than the solder joint will experience during its normal lifetime. The results of these tests then have to be extrapolated to normal service conditions. The problem which arises, is that these deviating conditions can either trigger or bypass deformation and damage mechanisms different than those that would be observed during normal loading conditions. This is
a result of the time dependent behaviour exhibited by the materials involved, operating at temperatures close to their solidus point. Another mechanism which is not properly accounted for by these methods is the possible evolution of microstructure, e.g. grain growth, recrystallisation, and diffusion, which is also facilitated by the high temperatures. Predictions based on accelerated cycling tests should therefore be scrutinised with great care.

1.2 Lead–free solder

Until to date, the most commonly used solder alloy was the eutectic Sn–Pb system. The Pb in the alloy lowers the surface tension of the Sn, giving the solder very good wetting characteristics. It also prevents the $\beta$-Sn, which has a closely packed tetragonal structure, to transform into $\alpha$-Sn when cooled below 13.2°C. The $\alpha$-Sn has a cubic structure, and the transformation causes a volume increase which can lead to damage and fractures. The joints created with the alloy have adequate fatigue properties and because it has been used in microelectronics for decades its thermal fatigue behaviour is reasonably well understood and both accelerated test data and extensive field data are available.

Recently, a drive to develop Pb–free solder alloys has emerged, due to the adverse effect of Pb on health and environment. Because of the small quantities of solder used in microelectronics it is very difficult to recycle it, as opposed to when Pb is used in car batteries for example. Consequently, it can more easily leach into the surface water. In Europe and Japan, legislation is made to ban Pb from electric and electronic equipment. As of July 2006 no products containing Pb may be put on the market in Europe. However, exemptions are made in cases where substitution is not possible from a scientific and technical point of view or if the negative environmental or health impacts caused by substitution are likely to outweigh the human and environmental benefits of the substitution. For example, in applications that are critical to the health and safety of the users, in certain flip-chip configurations, where the as yet unsurpassed thermal fatigue properties of Pb containing solders are vital, and in applications where high melting point, ductile solders are required [EU, 2003; Goodman et al., 2004].

The second reason to remove Pb from solder alloys results from the ongoing miniaturisation, which calls for new solder alloys that can be used more easily and accurately to create the required small joints. Also, new techniques, such as cascade soldering, where several components are soldered on top of each other sequentially, ask for solders with different melting temperatures and mechanical properties. Finally, also the consumers play an important role. They are becoming more environmentally aware giving Pb–free products an economical advantage over their counterparts, by preferring ‘green’ products.

A suitable replacement for the Sn–Pb solder must meet various requirements. It should have good wetting characteristics, provide a strong mechanical bond, have good thermal fatigue properties, be non-toxic, have an acceptable liquidus point, be reasonably inexpensive, etc. Many tin based lead–free solder alloys have been proposed as a replacement for the eutectic Sn–Pb, e.g. Sn–Ag, Sn–Ag–Cu, Sn–Bi, Sn–Zn, Sn–Zn–Bi, Sn–Ag–Bi. However, much research is at present ongoing and further needed into the behaviour of these materials. Although lead–free solder is already in use for some applications, long term field data is still missing.
1.3 Microstructure and material properties

Because of their small size, solder joints behave different from bulk solder material. When a joint is made it cools down rapidly, and together with the high ratio of substrate surface to solder volume, which results in a large number of heterogeneous nucleation sites for the solidification process [Bonda and Noyan, 1992], this drastically influences the resulting microstructure. The typical microstructural sizes are relatively large compared to the joint’s dimensions, see Fig. 1.4, and can not be neglected when investigating the mechanical behaviour. An alloy usually does not consist of a homogeneous mixture of the different atoms. Upon cooling the material develops a non-homogeneous microstructure in which regions with specific compositions exists: the phases. For instance, the eutectic Sn–Pb solder contains on average 63% Sn, but will separate into an $\alpha$ phase containing ca. 10% Sn, and a $\beta$ phase containing ca. 97% Sn, see Figs. 1.4 and 1.5. The different thermal and mechanical characteristics of these individual phases will affect the resulting overall joint properties [Basaran and Jiang, 2002].

Additionally, the microstructure of most solder alloys is not stable. Under the influence of temperature and mechanical loading various processes like phase separation, phase coalescence, grain growth, and recrystallisation take place, altering the microstructure continuously over time [Hacke et al., 1998; Conrad et al., 1999; Jung and Conrad, 2001a,b; Matin et al., 2004]. Fig. 1.5 shows the coarsening of the microstructure of eutectic Sn–Pb solder that is statically aged at 150°C. After only 15 hours the average phase size has already drastically increased. Experiments performed by Frear et al. [1988] on eutectic Sn–Pb showed that regions of high stress within a soldered connection tend to coarsen more than regions subjected to lower loads. These regions of pronounced coarsening were also identified as being the crack nucleation sites prior to failure of the joint [Frear, 1989; Frear et al., 1997], stressing the importance of taking the microstructure into account when evaluating the reliability of a solder interconnect.

Further complications arise through the presence of the solder pad, and the under bump metallisation (UBM). This is a metallic layer placed on the copper pad onto which the solder has to adhere. It is typically composed of several layers of different metals stacked on

![Figure 1.5](image_url) — Optical micrographs illustrating the coarsening of the Pb-rich phase particles in 63Sn-37Pb samples under static ageing conditions. The dark regions are the Sn-rich matrix and the light regions Pb-rich particles (courtesy of M. Matin).
top of each other. Each layer has a specific function, either to act as a diffusion barrier, a passivation layer, or as a bonding layer, joining two materials which would not create a strong bond between themselves. Unfortunately, diffusion from one layer to the next and into the solder always takes place anyway, changing the composition of the solder and possibly creating intermetallics [Ghosh, 2000; Amagai et al., 2002], see Fig. 1.6. These intermetallics can create major problems for a connection, as they are very brittle and can grow large enough to completely traverse the joint. They cause stress concentrations to arise within the joint and often fracture is initiated at or near their interface.

Coarsening in the Sn–Pb solder system occurs readily and can easily be seen. In Pb–free solder coarsening is not so pronounced, especially in the alloys with a high Sn content like Sn–Ag–Cu and Sn–Ag. However the microstructure does evolve, adapting to its environmental conditions [Zhao et al., 2001; Allen et al., 2004a,b; Pang et al., 2004; Xiao et al., 2004] and consequently changing its material properties. In these solders the microstructure and subsequently the mechanical behaviour is often heavily influenced by the reaction of material from the solder pad with the solder. Other Pb–free alternatives, e.g. Sn–Bi [Miao and Duh, 2001] more closely resemble Sn–Pb and do show significant coarsening of phases.

The presence of the microstructure not only affects the material behaviour, but can also be the cause of internal stresses and strains inside the material. For example, the tetragonal structure of Sn causes this material to have different elastic constants and coefficients of thermal expansion for each of its crystal axes. The response of the material therefore dependents on the orientation of the crystals and the anisotropic properties lead to internal strains and stresses through inhomogeneous expansion when the joint’s temperature increases. The rapid cooling of a newly soldered connection can also be an origin of internal stresses. These residual stresses originating from the solidification process can have a negative influence on the fatigue life.

1.4 Objective

Fast technological developments in microelectronics, such as the ongoing miniaturisation and high power applications, ask for new improved solder techniques and solder concepts.
Performing reliability tests on interconnects and subsequently analysing the results is not a straightforward task. Performing the experiments in real service conditions will lead to unacceptable long testing times, and extrapolating results from accelerated tests might trigger problems with the intrinsic timescales involved. For these reasons, computer simulations are being exploited to predict the thermomechanical fatigue life of the joints, as they can provide more insight and permit a significant reduction in cost and time of the design of highly reliable soldered connections in electronic packaging and surface mount technology.

When performing these calculations the material is usually assumed to be homogeneous, i.e. the microstructure is left out of consideration. However, as discussed above, since the dimensions of soldered connections are approaching typical microstructural length scales, this does not provide an adequate description of the material's mechanical behaviour. Attempts have been made to incorporate microstructural variables, like the average phase or grain size, into the constitutive model of a solder alloy [Pan, 1991; Frear et al., 1997; Basaran and Chanderoy, 1998; Vianco et al., 1999; Basaran and Chanderoy, 2000]. Yet, this does not provide all the microstructural information needed to accurately describe the joint's behaviour. Recently, the degeneration of the joint's structural integrity has been analysed using cohesive zones [Abdul-Baqi et al., 2005]. Although such an approach includes more microstructural information into the simulation, the failure paths are predetermined and bulk damage can not be described properly.

The main objective of this work is to develop a finite element approach which can be used to understand the intricate thermomechanical fatigue process of multiphase alloys like solder. This is accomplished by not only taking into account the underlying microstructure of the material, but also its evolution over time under the influence of temperature and mechanical loading. Information on this microstructure should be as complete as possible, it can not be captured by a single parameter. The shape and distribution of grains or phases, the orientation of the crystals, and the composition of individual phases, all influence the material's response. Additionally, a constitutive model will be used which can describe deformation processes in the solder at a high homologous temperature, as typically the case in microelectronics applications. This can then be coupled to the microstructural information to account for the mechanical inhomogeneity. Finally, a continuum damage formulation will complete the model in order to account for the gradual reduction of material properties.

### 1.5 Outline of the thesis

At the time this project was initiated commercial eutectic Sn–Pb was still the most predominate solder alloy used in electronic devices. It is therefore used as a reference material in most of this thesis. Another important motivation for this choice is the vast experience that has been build up with this material in the electronics industry over several decades, leading to extensive information and a good understanding of its characteristics at an engineering scale, although a thorough understanding of its microstructural behaviour is still lacking. Eutectic Sn–Pb makes a better choice as a reference material than the newly emerged Pb–free replacement alloys, where much research is still needed to clarify the observed thermomechanical behaviour.
In chapter 2, a phase field model is proposed for describing the microstructure evolution of a multiphase alloy. The driving force for the evolution is found in the free energy, leading to a dependency on both temperature and mechanical loading. A demonstration of the approach is provided through the simulation of a static ageing experiment of eutectic Sn–Pb, and a quantitative comparison is made between the numerical results and the experimental data.

To account for the time dependent mechanical response, typically demonstrated by materials operating at temperatures near their melting point, in chapter 3, the microstructure evolution model is extended with a viscoplastic constitutive model. Information on the microstructure, obtained in chapter 2, is included in the formulation through the material parameters. Simulations of mechanical loading of Sn–Pb solder are presented to provide insight in the interaction of the microstructural features and the material response.

Next, in chapter 4, a continuum non-local damage approach is added, completing the model. The non-local approach ensures mesh insensitivity and the continuum approach does not require any prior knowledge on the location of damage onset or failure initiation. The phase field model is here used to assign a different sensitivity to damage to the different constituents of the microstructure.

In chapter 5 attention is focused on the Sn–Ag–Cu system, which is the most likely replacement candidate of Sn–Pb in the strive towards lead-free electronics. Experimental thermal cycling of this material, without any external loading, showed damage occurring near grain boundaries, presumably due to differences in the thermal expansion coefficients of the crystal axes of Sn. To investigate this hypothesis, anisotropic behaviour is included in the material model and three-dimensional simulations are performed, the results of which are confronted with the experimental data.

The thesis is concluded with a brief summary of the conclusions and a discussion.
Chapter 2

A nonlocal phase field model for microstructure evolution of tin–lead solder

Microstructural length scales are relatively large in typical soldered connections. A microstructure which is continuously evolving is known to have a strong influence on damage initiation and propagation in solder materials. In order to make accurate lifetime predictions by numerical simulations, it is therefore necessary to take the microstructural evolution into account. In this work this is accomplished by using a diffuse interface model incorporating a strongly nonlocal variable. It is presented as an extension of the Cahn–Hilliard model, which is weakly nonlocal since it depends on higher order gradients which are by definition confined to the infinitesimal neighbourhood of the considered material point. Next to introducing a truly nonlocal measure in the free energy, this nonlocal formulation has the advantage that it is numerically more efficient. Additionally, the model is extended to include the elastically stored energy as a driving force for diffusion after which the entire system is solved using the finite element approach. The model results in a computational efficient algorithm which is capable of simulating the phase separation and coarsening of a solder material caused by combined thermal and mechanical loading.

2.1 Introduction

Solder joints serve two important purposes. Firstly, they form the electrical connection between the component, e.g. a chip or resistor, and a substrate, e.g. a printed circuit board, and secondly, they constitute the mechanical bond that holds the component to the substrate. The most commonly used solder alloy up till now is the Sn–Pb system. Due to ongoing miniaturisation in microelectronics the Sn–Pb solder is performing at its limit. Sn–Pb solder bumps –the generally used joint configuration– are already being produced smaller than their minimum size as determined by the surface tension. This leads to a vulnerable connection, which can easily fail when subjected to loads. Furthermore, the increasing functionality and chip density on printed circuit boards, leads to higher energy fluxes and thus higher temperatures. Although legislation in Europe is moving toward Pb-free electrical and electronic equipment in 2008, exemptions will be made for applications where substitution is not possible from a scientific and technical point of view or if the negative environmental or health impacts caused by substitution are likely to outweigh the human and environmental benefits of the substitution. Examples are: Pb in high melting temperature type solders (i.e. Sn–Pb solder alloys containing more than 85% Pb,), Pb in solders for servers, storage and storage array systems (exemption granted until 2010), and Pb in solders for network infrastructure equipment for switching, signalling, transmission as well as network management for telecommunication.

Solder joints are known to behave differently than the bulk solder material [Bonda and Noyan, 1992]. Firstly, this is presumably due to the presence of a high ratio of substrate surface to solder volume, which results in a large number of heterogeneous nucleation sites during solidification. Secondly, a concentration gradient of elemental or metallurgical composition is formed in the joint, e.g. intermetallics. Thirdly, processed microstructural sizes are not negligible compared to typical joint dimensions. Each of these conditions contributes to a structure that is not homogeneous, so that the joint can no longer be regarded as a homogeneous continuum. Moreover, its microstructure and properties will change over time: the high homologous temperature at which solder materials operate provokes microstructural coarsening, resulting in a markedly different microstructure after only a short time following solidification, as can be seen in Fig. 1.5 [Conrad et al., 1999; Jung and Conrad, 2001a,b; Matin et al., 2003]. Phases separate out (phase precipitation) and the different thermal expansion coefficients, moduli, and creep properties of the two materials will affect the resulting overall joint properties [Basaran and Jiang, 2002].

During thermomechanical cycling of a soldered connection the coarsening process will not be homogeneous. Frear et al. [1988] found that regions of high shear strain within an eutectic Sn–Pb connection coarsened more. These regions of inhomogeneous coarsening are known to be crack nucleation sites during thermal cycling [Frear et al., 1988; Frear, 1989; Frear et al., 1997]. It is also well-known that thermal cycling leads to faster coarsening rates than static thermal ageing [Hacke et al., 1998], possibly due to the mechanical stresses which arise within the material due to differences in thermal expansion coefficients. Another factor which influences the diffusion process is the surface tension between different phases. It controls the number, steepness, orientation, and final shape of the interfaces within the coarsened structure. The magnitude of the surface tension can vary in different directions of the crystal lattice. Because the microstructure has a big influence on the material properties and coarsening readily occurs during the lifetime of
a soldered connection, it is important to take the microstructural evolution into account during numerical simulation of fatigue damage propagation.

Various attempts have been made to include a microstructural variable like the average grain or phase size in the model for a solder material [Pan, 1991; Basaran and Chanderoy, 1998, 2000; Vianco et al., 1999; Frear et al., 1997]. In the literature several equations can be found to describe grain size coarsening [Lifshitz and Slyozov, 1961; Clark and Aldan, 1973; Senkov and Myshlyaev, 1986], however no information about the distribution or shape of the grains is given. More information about the microstructure can be obtained by using a diffuse interface model [Cahn and Hilliard, 1958]. These models are based on the notion that the free energy is not only a function of a local order parameter, but also depends on the value of this parameter in its immediate neighbourhood. In this way the energy contribution of an interface to the free energy is included. Minimisation of the free energy then leads to coarsening of the system through the reduction of interfaces, either grain or phase boundaries. [Chan and Rey, 1995; Gurtin, 1996; Tikare et al., 1999; Zhu et al., 1999; Wu et al., 2001; Fan et al., 2002]. An extension to include elastic effects is straightforward through the addition of a strain energy in the free energy [Larché and Cahn, 1992; Leo et al., 1998; Dreyer and Müller, 2000, 2001; Hu and Chen, 2001; Li and Müller, 2001; Zhu et al., 2001b; Krill III and Chen, 2002; Ni et al., 2002; Vaithyanathan and Chen, 2002; Nestler et al., 2003]. Not only can a diffuse interface model be used to describe coarsening of phases, other applications include void diffusion [Bhate et al., 2002], phase transformation [Artemev et al., 2000; Artemev and Khachaturyan, 2001], and solidification [Nestler et al., 2000; Nestler and Wheeler, 2002].

Although the Cahn–Hilliard equation is well capable of describing the phase segregation process, it does not arise as an exact macroscopic (homogenised) description of microscopic models of interacting particles. Giacomin and Lebowitz [1997]: “Such derivations are both of intrinsic interest and also indicate something about the range of applicability of the macroscopic equations. The latter might be particularly relevant for the Cahn–Hilliard equation where all that is known mathematically about the behaviour of the solutions is restricted to the late stages of the coarsening process when the evolution is assumed to be dominated by the motion of sharp interfaces between well-formed domains of the pure phases.” In their work, Giacomin and Lebowitz [1997] derive a macroscopic equation describing phase segregation in microscopic model systems interacting via long-range Kac potentials, also known as local mean field interactions. This equation will be used here as the point of departure.

In the following a diffuse interface model that is dependent on a strongly nonlocal field variable will be introduced in section 2.2. Whereas a strongly nonlocal formulation for the free energy density is conventionally written in an integral format, in this contribution the specific free energy as proposed by Giacomin and Lebowitz [1997] is reformulated towards a differential form. The particular format adopted in this paper allows for a rather straightforward numerical approach, whereas it preserves all strongly nonlocal characteristics. Next, in section 2.3, an outline is given of the finite element implementation of the resulting model. Next, in section 2.4, the choice of material parameters used for the simulations is discussed, after which, in section 2.5, one-dimensional calculations are used to investigate the influence of the parameters on the model. Two-dimensional calculations are presented which are compared with experimental results.
2.2 A strongly nonlocal diffuse interface theory

In this section the underlying diffusion equation will be introduced, after which a diffuse interface theory will be presented based on a strongly nonlocal interaction at the interface.

2.2.1 Diffusion equation

The local form of mass balance per component reads

\[
\frac{\partial \hat{\rho}_i}{\partial t} = -\vec{\nabla} \cdot (\hat{\rho}_i \vec{v}_i) \tag{2.1}
\]

where \( \vec{v}_i \) is the velocity of component \( i \) and \( \hat{\rho}_i \) its mass concentration, defined as the mass per unit of volume,

\[
\hat{\rho}_i \equiv \frac{m_i}{\Omega} \tag{2.2}
\]

with \( \Omega \) denoting volume. Summation over all components of equation (2.1) yields the total mass balance

\[
\frac{\partial \rho}{\partial t} = -\vec{\nabla} \cdot (\rho \vec{v}) \tag{2.3}
\]

where \( \rho \) is the mass density of the mixture

\[
\rho = \sum_{i=1}^{N} \hat{\rho}_i \tag{2.4}
\]

and \( \vec{v} \) is the barycentric velocity

\[
\vec{v} = \sum_{i=1}^{N} c_i \vec{v}_i \quad \text{with mass fraction } c_i = \frac{\hat{\rho}_i}{\rho} \tag{2.5}
\]

From mass balance per component and total mass balance, equations (2.1) and (2.3), the diffusion equation can be derived. First the equations for local and total mass balance are rewritten,

\[
\frac{\partial \hat{\rho}_i}{\partial t} = -\vec{\nabla} \cdot (\hat{\rho}_i \vec{v}_i) \quad \Rightarrow \quad \frac{\partial c_i \rho}{\partial t} = c_i \frac{\partial \rho}{\partial t} + \rho \frac{\partial c_i}{\partial t} = -\vec{\nabla} \cdot (\hat{\rho}_i \vec{v}_i) \tag{2.6a}
\]

\[
\frac{\partial \rho}{\partial t} = -\vec{\nabla} \cdot (\rho \vec{v}) \quad \Rightarrow \quad c_i \frac{\partial \rho}{\partial t} = -c_i \vec{\nabla} \cdot (\rho \vec{v}) = -\vec{\nabla} \cdot (\hat{\rho}_i \vec{v}_i) + \rho \vec{v} \cdot \vec{\nabla} c_i \tag{2.6b}
\]

Substitution of (2.6b) in (2.6a) results in:

\[
\rho \frac{dc_i}{dt} = -\vec{\nabla} \cdot [\hat{\rho}_i(\vec{v}_i - \vec{v})] = -\vec{\nabla} \cdot \vec{j}_i \tag{2.7}
\]

where \( \vec{j}_i \) is the diffusion mass flow per unit area per unit time. and \( \frac{dc_i}{dt} \) denotes the material time derivative: \( \frac{\partial c_i}{\partial t} + \vec{v} \cdot \vec{\nabla} c_i \).
Using the phenomenological approach of classical irreversible thermodynamics [De Groot and Mazur, 1984], the following expression for the diffusion mass flow is found:

\[ \vec{j}_i = -\rho \mathbf{M} \cdot \vec{\nabla} \left( \delta \Phi \delta c_i \right) \quad (2.8) \]

where \( \Phi \) is the specific Helmholtz free energy and \( \mathbf{M} \) the mobility tensor. This means that when the derivative of the Helmholtz free energy with respect to the composition, also called the chemical potential, is homogeneous throughout the system, no net mass transport will take place, i.e. \( \vec{j}_i = 0 \). Note that this does not necessarily mean that the mass fraction has to be homogeneous throughout the system, as in the case of Fick’s law.

### 2.2.2 Free energy

To complete the diffusion equation an explicit expression for the free energy function is required. At any time instant there will be different types of contributions to the energy available in a system. Firstly, each phase will have different values for the free energy for different compositions, which is represented by the so-called configurational energy \( \Phi_0 \). Secondly, interfaces will contribute to the surface energy, denoted \( \Phi_\gamma \), and thirdly, stresses and strains will cause elastic energy, indicated as \( \Phi_e \), to be stored in the system. So the complete free energy is then given by

\[ \Phi = \Phi_0 + \Phi_\gamma + \Phi_e \quad (2.9) \]

**Configurational free energy**

For a system where only binary interactions take place the configurational free energy takes the following form [Prigogine, 1961]:

\[ \Phi_0 = \sum c_i g_i + \sum_i R T c_i \ln(c_i) + \sum_i \sum_{j>i} c_i c_j \chi_{ij} \quad \text{J/m}^3 \quad (2.10) \]

where \( g_i \) and \( \chi_{ij} \) are coefficients dependent on temperature, \( R \) is the gas constant, and \( T \) is temperature. The first term on the right hand side represents the free energy of the pure components. The last two terms are known as the Flory–Huggins free energy density of mixing. The second term arises because of the entropy of mixing, while the last term is a consequence of the mixing being non-ideal. If higher-order interactions become more important they can be taken into account by adding higher-order interaction terms.

For a two-phase material (phases \( \alpha \) and \( \beta \)), the configurational energy is represented by a double-well potential, see Fig. 2.1. It can be constructed from the configurational energy curves of the individual phases by assuming that at a certain composition only the phase with the lowest energy will exist. Equilibrium is then found when the chemical potential is homogeneous throughout the system. Note that this does not mean that the mass fraction has to be homogeneous. The configuration with the lowest possible free energy is found when, due to phase separation, all of the material has either one of the two compositions defined by the so-called binodal points. These binodal points can be found by constructing a double tangent to the free energy curve. If the binodal points are plotted at different temperatures the phase diagram will be obtained.
Two other points which are of interest are the so-called spinodal points. These can be found by setting the curvature of the configurational free energy to zero, i.e.

$$\frac{\partial^2 \Phi_0}{\partial c^2} = 0 \quad (2.11)$$

Note that from here on a binary system is assumed, and therefore the subscript $i$ can be omitted from $c$. The mass fraction of the other component is simply $1 - c$. In the region where the curvature is positive the system is stable with respect to small fluctuations in composition. However in the regions where the curvature is negative the system is locally unstable with respect to small fluctuations [Jones and Richards, 1999]. Note that not all phases have spinodal points.

**Surface free energy**

The *surface free energy* accounts for energy due to interfaces between distinct phase regions. Typically, this contribution is not only a function of the local mass fraction $c$. The composition of the surroundings (i.e. the interface transition) of a material point has to be taken into account as well. In diffuse interface theories this is done by taking the free energy to be dependent on both the local composition and its derivatives. Using the approach of Cahn and Hilliard [1958] a Taylor expansion of the Helmholtz free energy is used to obtain a diffusion equation of which the driving forces are a function of the composition and its derivatives. They showed that the total surface energy $F_\gamma$ could be approximated by

$$F_\gamma = \int_\Omega \Phi_\gamma \, d\Omega = \int_\Omega (\kappa \nabla \cdot \kappa \nabla) \, d\Omega \quad (2.12)$$

Under the assumption that $\vec{\nabla} \cdot \vec{n} = 0$ on the boundary this can be rewritten as

$$F_\gamma = \int_\Omega \Phi_\gamma \, d\Omega = \int_\Omega \gamma \vec{\nabla} \cdot \vec{\nabla} \, d\Omega \quad (2.13)$$

where $\gamma_{ch}$ is here called the gradient energy coefficient to avoid confusion with the interface tension coefficient. This leads to the following expression for the driving force or chemical potential $\mu$ originating from surface tension

$$\mu = \frac{\delta \Phi_\gamma}{\delta c} = -\gamma_{ch} \nabla^2 c \quad (2.14)$$
Gurtin [1996] followed a different approach based on a microforce balance to arrive at the same expression. Through the truncation in the Taylor expansion, the model becomes weakly nonlocal, since the considered derivatives of the composition are confined to an infinitesimal neighbourhood of the considered material point only.

Although the Cahn–Hilliard equation is well capable of describing the phase segregation process, it does not arise as an exact macroscopic description (homogenised) of microscopic models of interacting particles. Such derivations are both of intrinsic interest and also indicate something about the range of applicability of the macroscopic equations [Giacomin and Lebowitz, 1997]. In their work, Giacomini and Lebowitz [1997] derive a macroscopic equation describing phase segregation in microscopic model systems with long-range interactions evolving according to stochastic Kawasaki dynamics with nearest neighbour exchanges (see also Brandon [1994]). They derive the following expression for the specific free energy:

\[
\Phi = \Phi_0(c(\vec{x})) + \frac{1}{4} \int_\Omega g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right]^2 d\vec{y}
\]  

(2.15)

where \( \Omega \) is the region around the material point with position vector \( \vec{x} \), \( \vec{y} \) a local coordinate vector with respect to \( \vec{x} \) pointing to points in \( \Omega \), \( g \) is a symmetric smooth function \((C^\infty)\), and \( \Phi_0 \) is the equilibrium free energy density of a homogeneous system. Similar nonlocal extensions of the free energy have been used by Gajewski and Zacharias [2003] and Fosdick and Mason [1998].

Using the specific free energy (2.15) and taking the variation with respect to \( c(\vec{x}) \) in order to obtain the chemical potential then gives:

\[
\mu = \frac{\delta \Phi}{\delta c(\vec{x})} = \frac{\partial \Phi_0}{\partial c} + \frac{1}{2} \int_\Omega g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right] d\vec{y}
\]  

(2.16)

Which can be rewritten to yield:

\[
\mu = \frac{\partial \Phi_0}{\partial c} + \kappa \left[ c(\vec{x}) - \bar{c}(\vec{x}) \right]
\]  

(2.17)

where

\[
\bar{c}(\vec{x}) = \int_\Omega g'(\vec{y}) c(\vec{x} + \vec{y}) d\vec{y}, \quad \kappa = \frac{1}{2} \int_\Omega g(\vec{y}) d\vec{y}, \quad \text{and} \quad g' = g/\kappa
\]  

(2.18)

Thus the free energy is now a function of the local mass fraction \( c \) and a nonlocal mass fraction \( \bar{c} \) that is a weighted averaged value of \( c \) over a finite zone in the vicinity of the interface and \( \kappa \) is an interface tension coefficient with units \( J/m^3 \).

In analogy with Peerlings et al. [1996] equation (2.18) can be reformulated towards an implicit Helmholtz equation (see appendix A.):

\[
\bar{c} - \ell_c^2 \nabla^2 \bar{c} = \bar{c}
\]  

(2.19)

where \( \ell_c \) is an internal length parameter defined as

\[
\ell_c^2 = \frac{1}{2\Omega} \int_\Omega \vec{y}^2 g(\vec{y}) d\Omega
\]  

(2.20)
Note that the solution of this partial differential equation indeed leads to a strongly nonlocal solution as assumed in (2.18). Details on this particular topic have been elaborated within the context of strongly nonlocal damage [Peerlings et al., 2001]. Details on the explicit format of the nonlocal kernel (which is the result of the adopted mathematical approximation) can be found in this reference as well. Ren and Truskinovsky [2000] use a similar Helmholtz equations to introduce long-range elastic interactions in a system. Minimisation then leads to a finite scale microstructure with many interfaces, instead of only a single interface.

The reformulation of the integral expression (2.18) into differential equation (2.19) requires the introduction of boundary conditions. Either the value of $\bar{c}$, its normal derivative, or a linear combination of these quantities must be specified on the boundary. Here the homogeneous Neumann boundary condition has been used:

$$\nabla \bar{c} \cdot \vec{n} = 0 \quad (2.21)$$

This condition preserves the global mass fraction of the entire system in the nonlocal averaging:

$$c_{total} = \frac{1}{\Omega} \int_\Omega c \, d\Omega = \frac{1}{\Omega} \int_\Omega \bar{c} \, d\Omega \quad (2.22)$$

More comments on this boundary condition can be found in Peerlings et al. [2001].

Equation (2.19) can be substituted into (2.17) which leads to an expression similar to the Cahn–Hilliard equation except that in this case the nonlocal mass fraction appears in the gradient energy term:

$$\frac{\delta \Phi}{\delta \bar{c}} = -\gamma \nabla^2 \bar{c} \quad (2.23)$$

where the gradient energy coefficient $\gamma$ equals $\kappa \ell_c^2$. Note that if the weakly nonlocal explicit approximation of the nonlocal averaging equation is used ($\bar{c} = c + \ell_c^2 \nabla^2 c$) the model would yield the classical Cahn–Hilliard theory with $\gamma_{ch} = \kappa \ell_c^2$. In the following the strongly nonlocal implicit Helmholtz equation will be used to calculate $\bar{c}$. It is again emphasised that the main difference resides in the finite character of the spatial interactions throughout the interface.

### Elastically stored free energy

The elastically stored energy is due to stresses and strains which occur in the material. Thus in order to calculate it, a constitutive model needs to be chosen and the momentum equation, has to be solved. As a first approximation linear elasticity is assumed and inertia effects and body forces are left out of consideration. The momentum equation than reads

$$\mathbf{\nabla} \cdot \mathbf{\sigma} = \mathbf{\nabla} \cdot [\mathbf{H} \cdot (\mathbf{\varepsilon} - \mathbf{\varepsilon}_T)] = \mathbf{\bar{0}} \quad (2.24)$$

where $\mathbf{\sigma}$ denotes the stress tensor and $\mathbf{H}$ the fourth-order material tensor. It is assumed that deformations are small, i.e. the infinitesimal mechanical strain tensor $\mathbf{\varepsilon} = \frac{1}{2} (\mathbf{\nabla} \mathbf{\bar{u}} + (\mathbf{\nabla} \mathbf{\bar{u}})^\mathsf{T})$ is used, where $\mathbf{\bar{u}}$ is the macroscopic displacement of the material. The thermal strains $\mathbf{\varepsilon}_T$ are defined as $\mathbf{\varepsilon}_T = \mathbf{\alpha}(c) (T - T_c)$ with $\mathbf{\alpha}(c)$ the composition dependent coefficients of thermal
expansion and $T_c$ the critical temperature of the solder. The specific elastic energy is then defined as

$$\Phi_e = \frac{1}{2} \sigma : \varepsilon = \frac{1}{2} \varepsilon : \mathbf{H} : \varepsilon$$

(2.25)

Note that the elastically stored energy term introduces a third-order gradient of the displacements into the diffusion equation (2.7). Because the system will be solved by using a finite element method it is convenient to eliminate these third-order gradient terms. This is accomplished by introducing a new independent variable $\psi$, defined as the variation of the elastic energy with respect to the local mass fraction

$$\psi = \frac{\delta \Phi_e}{\delta c} = \frac{1}{2} \varepsilon : \frac{\partial \mathbf{H}}{\partial c} : \varepsilon$$

(2.26)

### 2.2.3 System of equations

The system of equations which has to be solved, consists of the diffusion equation for the local mass fraction, an implicit Helmholtz equation for the nonlocal mass fraction, the momentum equation for stresses and strains, and the equation for the variation of the elastically stored energy with respect to the local mass fraction:

$$\rho \frac{dc}{dt} = \vec{V} \cdot \left[ \rho \mathbf{M}(c) \cdot \nabla \left( \frac{\partial \Phi_0}{\partial c} - \kappa (\bar{c} - c) + \psi \right) \right]$$

(2.27a)

$$\bar{c} - \ell_c^2 \nabla^2 \bar{c} = c$$

(2.27b)

$$\vec{V} \cdot \sigma = \vec{0}$$

(2.27c)

$$\psi = \frac{1}{2} \varepsilon : \frac{\partial \mathbf{H}}{\partial c} : \varepsilon$$

(2.27d)

The boundary condition for (2.27a) is the zero mass flux, i.e. no mass is exchanged with surroundings:

$$\vec{j} \cdot \vec{n} = 0$$

(2.28)

For the nonlocal equation (2.27b) the homogeneous Neumann boundary condition (2.21) is chosen, which is the nonlocal equivalent of the boundary condition used by Cahn and Hilliard. For the momentum equation the usual boundary conditions are applied:

$$\sigma \cdot \vec{n} = \vec{p}$$

(2.29)

where $\vec{p}$ denotes external applied forces. In the next section the solution method for this system will be discussed.
2.3 Finite element implementation

The system of equations is solved using the finite element method. The first step is the time discretisation of the diffusion equation, according to an implicit or backward Euler scheme, which is unconditionally stable and first-order accurate:

$$\rho^{t+\Delta t} \frac{dc}{dt}\bigg|^{t+\Delta t}_{t} = f(c^{t+\Delta t}) \quad \Rightarrow \quad c^{t+\Delta t} = \frac{1}{\rho^{t+\Delta t}} \Delta t f(c^{t+\Delta t}) + c^t$$  \hspace{1cm} (2.30)

A standard Newton–Raphson iterative procedure will be used to solve the set of equations (2.27). Therefore each field variable is decomposed in the following iterative fashion:

$$c^{n+1} = c^n + \delta c, \text{ etc.}$$

Here the subscript $n$ denotes the solution from the previous iteration and $\delta$ the current iterative correction. In the following all variables are assumed to be evaluated at $t + \Delta t$ at iteration $n$ unless denoted otherwise.

2.3.1 Weighted residual formulation

Diffusion equation

After time discretisation, the diffusion equation is multiplied with a weighting function $w_c$. Integration of the product over the domain $\Omega$ leads to the weighted residual form of the diffusion equation. Applying partial integration results in the weak form:

$$\int_{\Omega} w_c \rho c \, d\Omega = \Delta t \int_{\Omega} \nabla \cdot \left[ w_c \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \right] \, d\Omega$$

$$\quad - \Delta t \int_{\Omega} \nabla w_c \cdot \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \, d\Omega + \int_{\Omega} w_c \rho \, d\Omega \quad \forall w_c$$  \hspace{1cm} (2.31)

Since no mass is exchanged through the external surface, the zero mass flux boundary condition is used for the diffusion equation:

$$\int_{\Gamma} w_c \vec{j} \cdot \vec{n} \, d\Gamma = - \int_{\Gamma} \left[ w_c \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \right] \cdot \vec{n} \, d\Gamma =$$

$$\quad = - \int_{\Omega} \nabla \cdot \left[ w_c \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \right] \, d\Omega = 0$$  \hspace{1cm} (2.33)

where use has been made of the Gauss divergence theorem and where $\vec{n}$ is the unit normal vector on the surface directed away from the domain. Subsequent linearisation yields

$$\int_{\Omega} w_c \left[ \delta \rho (c - c^t) + \rho \delta c \right] \, d\Omega + \Delta t \int_{\Omega} \nabla w_c \cdot \left( \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \right) \, d\Omega = - f^c_{int}$$  \hspace{1cm} (2.34)

with

$$f^c_{int} = \int_{\Omega} w_c \rho (c - c^t) \, d\Omega + \Delta t \int_{\Omega} \nabla w_c \cdot \rho M \cdot \nabla \left( \frac{\delta \Phi}{\delta c} \right) \, d\Omega$$  \hspace{1cm} (2.35)
Assuming $M$ and $\rho$ to be functions of $c$ only, and substituting the free energy density as defined in section 2.2.2 results in

$$
\int_\Omega \left[ w_c \left( \rho + \frac{\partial \rho}{\partial c} (c - c^*) \right) + \Delta \bar{v} w_c \cdot \left\{ \rho M \frac{\partial^3 \Phi_0}{\partial c^3} \bar{v} c^c \right. \\
+ \left. \left( \frac{\partial \rho}{\partial c} M + \rho \frac{\partial M}{\partial c} \right) \left\{ \left( \frac{\partial^2 \Phi_0}{\partial c^2} + \kappa \right) \bar{v} c - \kappa \bar{v} \bar{c} + \bar{v} \psi \right\} \right] \delta c \, d\Omega \\
+ \int_\Omega \left[ \Delta \bar{v} w_c \cdot \rho M \left( \frac{\partial^2 \Phi_0}{\partial c^2} + \kappa \right) \cdot \bar{v} \delta c \right] \, d\Omega \\
+ \int_\Omega \left[ \Delta \bar{v} w_c \cdot \rho M \cdot \left( \bar{v} \delta \psi - \kappa \bar{v} \delta \bar{c} \right) \right] \, d\Omega = -f_{int}^c
$$

(2.36)

**Nonlocal equation**

Multiplication of the nonlocality equation with a weighting function $w_\tilde{c}$ and integration of this product over the domain yields:

$$
\int_\Omega w_\tilde{c} (\tilde{c} - \ell^2 \nabla^2 \tilde{c}) \, d\Omega = \int_\Omega w_\tilde{c} c \, d\Omega \quad \forall \ w_\tilde{c}
$$

(2.37)

Applying partial differentiation and the Gauss divergence theorem results in

$$
\int_\Omega w_\tilde{c} \tilde{c} \, d\Omega + \int_\Omega \bar{v} w_\tilde{c} \cdot \ell^2 \nabla \tilde{c} \, d\Omega = \int_\Omega w_\tilde{c} c \, d\Omega + \int_\Gamma w_\tilde{c} \ell^2 \bar{v} \cdot \bar{n} \, d\Gamma
$$

(2.38)

The last term on the right hand side disappears after applying the usual homogeneous Neumann boundary condition $\bar{v} \tilde{c} \cdot \bar{n} = 0$. Subsequent linearisation gives:

$$
\int_\Omega w_\tilde{c} (\delta \tilde{c} - \delta c) \, d\Omega + \int_\Omega \bar{v} w_\tilde{c} \cdot \ell^2 \bar{v} \delta \tilde{c} \, d\Omega = -f_{int}^\tilde{c}
$$

(2.39)

with

$$
f_{int}^\tilde{c} = \int_\Omega w_\tilde{c} (\tilde{c} - c) \, d\Omega + \int_\Omega \bar{v} w_\tilde{c} \cdot \ell^2 \bar{v} \tilde{c} \, d\Omega
$$

(2.40)

**Momentum equation**

Multiplication of the momentum equation with a weighting function $\bar{w}_u$ and integration over the domain yields the weighted residual integral:

$$
\int_\Omega \bar{w}_u \cdot (\bar{v} \cdot \sigma) \, d\Omega = 0 \quad \forall \ \bar{w}_u
$$

(2.41)

Applying partial integration and the Gauss divergence theorem results in

$$
\int_\Omega (\bar{v} \bar{w}_u)^c : \sigma \, d\Omega = \int_\Gamma \bar{w}_u \cdot \bar{p} \, d\Gamma \quad \text{with} \ \bar{p} = \sigma \cdot \bar{n}
$$

(2.42)

where $\bar{p}$ is the external surface load. Now the constitutive equation $\sigma = \mathbb{H} : (\varepsilon - \varepsilon_T)$ is substituted, where it is assumed that $\mathbb{H}$ and $\alpha$ are solely dependent on the local composition. Linearisation constant temperature gives

$$
\int_\Omega (\bar{v} \bar{w}_u)^c : \left( \frac{\partial \mathbb{H}}{\partial c} : (\varepsilon - \varepsilon_T) - \mathbb{H} : \frac{\partial \alpha}{\partial c} (T - T_c) \right) \delta c + \mathbb{H} : \delta \varepsilon \right) \, d\Omega = \tilde{f}_{ext}^u - \tilde{f}_{int}^u
$$

(2.43)
with

$$f_{\text{ext}}^u - f_{\text{int}}^u = \int_{\Gamma} \tilde{\mathbf{u}} \cdot \tilde{\mathbf{p}} \, d\Gamma - \int_{\Omega} (\tilde{\nabla} \cdot \tilde{\mathbf{u}})^c : \mathbf{H} : (\mathbf{\varepsilon} - \mathbf{\varepsilon}^T) \, d\Omega$$

(2.44)

Variation of the elastic energy

Multiplication of equation (2.27d) with a weighting function \( w_\psi \) and subsequently integrating over the domain gives

$$\int_{\Omega} w_\psi \, d\Omega = \int_{\Omega} w_\psi \left( \frac{1}{2} \mathbf{\varepsilon} : \frac{\partial \mathbf{H}}{\partial \mathbf{c}} : \mathbf{\varepsilon} \right) \, d\Omega \quad \forall w_\psi$$

(2.45)

Linearisation yields

$$\int_{\Omega} w_\psi \delta \psi \, d\Omega - \int_{\Omega} w_\psi \mathbf{\varepsilon} : \frac{\partial \mathbf{H}}{\partial \mathbf{c}} : \delta \mathbf{\varepsilon} \, d\Omega + \int_{\Omega} w_\psi \mathbf{\varepsilon} : \frac{\partial \mathbf{H}}{\partial \mathbf{c}} : (T - T_c) \delta \mathbf{c} \, d\Omega - \int_{\Omega} w_\psi \frac{1}{2} \mathbf{\varepsilon} : \frac{\partial^2 \mathbf{H}}{\partial \mathbf{c}^2} : \mathbf{\varepsilon} \delta \mathbf{c} \, d\Omega = -f_{\text{int}}^\psi$$

(2.46)

with

$$f_{\text{int}}^\psi = \int_{\Omega} w_\psi \psi \, d\Omega - \int_{\Omega} w_\psi \frac{1}{2} \mathbf{\varepsilon} : \frac{\partial \mathbf{H}}{\partial \mathbf{c}} : \mathbf{\varepsilon} \, d\Omega$$

(2.47)

In the next section the discretisation of the above derived weak residual forms is discussed.

### 2.3.2 Discretisation

In order to obtain a finite element formulation the equations are now discretised. The domain is subdivided into \( n \) elements. The unknown fields, \( c, \bar{c}, \tilde{\mathbf{u}}, \) and \( \psi \), are approximated by \( C^0 \) continuous interpolation functions, \( N^c, N_{\bar{c}}, N_{\tilde{\mathbf{u}}}, \) and \( N^\psi \), within each element. The finite element approximations for the unknown fields and their derivatives are written as

\[
\begin{align*}
    c &= \sum_{A=1}^{n_c} N^c_A c_A \\
    \tilde{c} &= \sum_{A=1}^{n_{\bar{c}}} N_{\bar{c}}^A \tilde{c}_A \\
    \bar{u} &= \sum_{A=1}^{n_{\tilde{u}}} N_{\tilde{u}}^A \bar{u}_A \\
    \psi &= \sum_{A=1}^{n_{\psi}} N^\psi_A \psi_A
\end{align*}
\]

(2.48a)

\[
\begin{align*}
    \nabla c &= \sum_{A=1}^{n_c} \nabla N^c_A c_A \text{ with } \nabla N^c_A &= \nabla N^c_A \\
    \nabla \tilde{c} &= \sum_{A=1}^{n_{\bar{c}}} \nabla N_{\bar{c}}^A \tilde{c}_A \text{ with } \nabla N_{\bar{c}}^A &= \nabla N_{\bar{c}}^A \\
    \nabla \bar{u} &= \sum_{A=1}^{n_{\tilde{u}}} \nabla N_{\tilde{u}}^A \bar{u}_A \text{ with } \nabla N_{\tilde{u}}^A &= \nabla N_{\tilde{u}}^A \\
    \nabla \psi &= \sum_{A=1}^{n_{\psi}} \nabla N^\psi_A \psi_A \text{ with } \nabla N^\psi_A &= \nabla N^\psi_A
\end{align*}
\]

(2.48b)

where \( n_c, n_{\bar{c}}, n_{\tilde{u}}, \) and \( n_{\psi} \) are the total number of nodes for the corresponding field variables. The capital Latins subscripts indicate a nodal quantity. Following the standard Galerkin approach, the interpolation functions for the field variables are also used for the corresponding weighting functions.
Now a Cartesian basis in $\mathbb{R}^3$ is chosen, with respect to which the equations are written in column and matrix format. The total system is next divided into two systems of two coupled equations which are solved iteratively, using a conjugate gradient solver. The first set contains the diffusion equation and the nonlocality equation. The second set incorporates the momentum balance and the variation of the elastically stored energy. After substituting the discretised field variables into the weak forms, equations (2.36), (2.39), (2.43), and (2.46), the two systems can be written in matrix form:

$$
\begin{bmatrix}
K_{cc} & K_{cc} \\
K_{\bar{c}c} & K_{\bar{c}c}
\end{bmatrix}
\begin{bmatrix}
\delta c \\
\delta \bar{c}
\end{bmatrix}
= -
\begin{bmatrix}
f^c \\
f^\bar{c}
\end{bmatrix}
(2.49)
$$

$$
\begin{bmatrix}
K_{uu} & 0 \\
K_{\psi u} & K_{\psi\psi}
\end{bmatrix}
\begin{bmatrix}
\delta u \\
\delta \psi
\end{bmatrix}
= \begin{bmatrix}
f^{ext} \\
f^{\psi}\int
\end{bmatrix}
- \begin{bmatrix}
f^u \\
f^{\psi}\int
\end{bmatrix}
(2.50)
$$

with the submatrices defined as follows:

$$
K_{cc} = \int_{\Omega} w_c^T \frac{N_c}{T_c} \left( \rho + \frac{\partial \rho}{\partial c_n} (c_n - c^t) \right) N_c \, d\Omega
+ \Delta t \int_{\Omega} w_c^T B_c^T \left\{ \rho \frac{\partial^3 \Phi_0}{\partial c_n^3} B_c \frac{\partial c_n}{\partial c^t} N_c + \left( \frac{\partial^2 \Phi_0}{\partial c_n^2} + \kappa \right) B_c \right\}
+ \frac{\partial \rho \frac{M}{\partial c_n}}{\partial c_n} \left\{ \frac{\partial^2 \Phi_0}{\partial c_n^2} + \kappa \right\} B_c \psi \right\} N_c \, d\Omega
(2.51)
$$

$$
K_{\bar{c}c} = -\Delta t \int_{\Omega} w_c^T B_c^T \rho M \kappa B_c \, d\Omega
(2.52)
$$

$$
K_{\bar{c}\bar{c}} = \int_{\Omega} \left\{ w_c^T \frac{N_c}{T_c} \frac{N_c}{T_c} + w_c^T \frac{B_c^T}{B_c} \frac{B_c^T}{B_c} \right\} \, d\Omega
(2.53)
$$

$$
K_{cc} = -\int_{\Omega} w_c^T \frac{N_c}{T_c} \frac{N_c}{T_c} d\Omega
(2.54)
$$

$$
K_{uu} = \int_{\Omega} w_c^T B_u^T \int_{\psi} B_u \, d\Omega
(2.55)
$$

$$
K_{\psi\psi} = \int_{\Omega} w_c^T \frac{N_c}{T_c} \frac{N_c}{T_c} d\Omega
(2.56)
$$

$$
K_{\psi u} = -\int_{\Omega} w_c^T \frac{N_c}{T_c} \frac{N_c}{T_c} \frac{1}{2} (B_u u - \alpha_n (T - T_c)) \frac{\partial (\frac{H}{H} + \frac{H^T}{H})}{\partial c_n} B_u \, d\Omega
(2.57)
$$

$$
K_{\psi u} = -\int_{\Omega} w_c^T \frac{N_c}{T_c} \frac{N_c}{T_c} \frac{1}{2} (B_u u - \alpha_n (T - T_c)) \frac{\partial (\frac{H}{H} + \frac{H^T}{H})}{\partial c_n} B_u \, d\Omega
(2.58)
$$

### 2.4 Material parameters

The eutectic Sn–Pb system has been chosen for the simulations, because it has been studied extensively in the experimental literature. Hence, a considerable amount of experimental material data is available. For this system the $\alpha$-phase is a Pb-rich BCC crystal structure, which for the eutectic forms islands in a Sn-rich matrix, the $\beta$-phase which has a tetragonal structure. From here on the field variable $c$ denotes the mass fraction of tin (Sn), i.e. a material volume with $c = 0$ consists of 100% Pb and a material volume with $c = 1$ of 100% Sn. All the simulations are performed assuming a constant temperature of 150°C.
The necessary material data were obtained from literature and are summarised in table 2.1. The data used for the configurational free energy will be discussed in the following section. The internal length parameter \( \ell_c \) is directly related to the interface length. For a metal it represents the distance over which the presence of an interface disturbs/influences the crystal lattice, it is taken to be in the order of magnitude of the real interface width, approximately 50 atomic distances, i.e. 25 nm [Dreyer and Müller, 2000]. Furthermore, for a given \( \Phi \) and a measured interface length the gradient energy parameter \( \kappa \) can be determined.

The interface tension coefficient \( \kappa \) determines the intensity of the energy contained in the interface. High values of \( \kappa \) lead to a homogeneous distribution, since the presence of interfaces in the system is energetically unfavourable. On the other hand, low values of \( \kappa \) provide little resistance to interface formation therefore an unlimited amount of interfaces with very small width will develop during spinodal decomposition and moreover coarsening will not occur. The influence of \( \kappa \) will be demonstrated using one-dimensional calculations. For two-dimensional calculations \( \kappa \) is taken such that the smallest phase region observed in the experimental micrographs of the as-cast microstructure can be adequately described.

The mobility of the \( \alpha \)-phase is known to be significantly lower than that of the \( \beta \)-phase [Decker et al., 1977; Gupta et al., 1999; Sen and Ghorai, 1989; Ghosh and Liu, 1998], hence it is assumed to be negligible. The mobility of the \( \beta \)-phase is fitted using the comparison between a two-dimensional simulation and an ageing experiment, which will be discussed later. All parameters are assumed to vary linearly with the tin mass fraction \( c \).

<table>
<thead>
<tr>
<th>Table 2.1 — Material parameters for the Sn–Pb solder system at 150°C.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parameter</td>
</tr>
<tr>
<td>mass density of Sn</td>
</tr>
<tr>
<td>mass density of Pb</td>
</tr>
<tr>
<td>Young’s modulus of Sn</td>
</tr>
<tr>
<td>Young’s modulus of Pb</td>
</tr>
<tr>
<td>Poisson’s ratio of Sn</td>
</tr>
<tr>
<td>Poisson’s ratio of Pb</td>
</tr>
<tr>
<td>thermal expansion coefficient of Sn</td>
</tr>
<tr>
<td>thermal expansion coefficient of Pb</td>
</tr>
<tr>
<td>interface tension coefficient</td>
</tr>
<tr>
<td>mobility of ( \alpha )-phase</td>
</tr>
<tr>
<td>mobility of ( \beta )-phase</td>
</tr>
<tr>
<td>internal length scale</td>
</tr>
</tbody>
</table>

**Configurational free energy**

Fig. 2.2 shows the configurational part of the Helmholtz free energy curve for the Sn–Pb system at 150°C. The data used to calculate this curve is taken from MTDATA® [MTData, 1998; Dreyer and Müller, 2000]. For a binary system two free energy curves can be found, one for each phase. However this data supplies the energy in Joules per mole as a function of particle concentration \( y \). Since the current model requires energy per unit of volume
(see the configurational free energy function, equation (2.10)), the data has to be rewritten accordingly. Particle concentration $y$ can be replaced by mass fraction $c$ using the following relationship:

$$y = \frac{M_{\text{Pb}} c}{M_{\text{Sn}} - c (M_{\text{Pb}} - M_{\text{Sn}})}$$

(2.59)

where $M_i$ is the molar weight of component $i$ in kilogrammes per mole. Next the equation has to be divided by the averaged molecular weight of the specific composition given by

$$\bar{M} = \frac{M_{\text{Pb}} M_{\text{Sn}}}{M_{\text{Sn}} - c (M_{\text{Sn}} - M_{\text{Pb}})}$$

(2.60)

and multiplied with the density of that composition given by

$$\rho = \frac{\rho_{\text{Pb}} \rho_{\text{Sn}}}{\rho_{\text{Sn}} - c (\rho_{\text{Sn}} - \rho_{\text{Pb}})}$$

(2.61)

Now two free energy curves per unit volume can be found, one for each phase, see Fig. 2.2.

![Figure 2.2](image)

**Figure 2.2** — Configurational free energy $\Phi_0$ for the $\alpha$ and $\beta$-phase at 150°C and the fitted single free energy curve as a function of the tin mass fraction $c$.

Next it is assumed that at a certain composition the material evolves to the phase which has the lowest free energy at that composition. Thus a single (double-well) free energy curve can be constructed. To facilitate the numerical solution procedure, this curve is next fitted to the following function, in which the singularity at the intersection of the two curves is eliminated.

$$\Phi_0 = g_1 c + g_2 (1 - c) + \frac{g_3 c \ln c + g_4 (1 - c) \ln(1 - c) + g_5 c (1 - c)}{}$$

(2.62)

The resulting parameter the free energy function at 150°C are (in GJ/m$^3$),

$$g_1 = -1.3634 \quad g_3 = 0.3630 \quad g_5 = 0.8750$$

$$g_2 = -1.5263 \quad g_4 = 0.1643$$

(2.63)
2.5 Numerical simulations

2.5.1 One-dimensional calculations

One-dimensional calculations have been performed to investigate the response of the model. Several initial configurations are used to investigate the influence of the internal length scale parameter $\ell_c$ and the interface tension coefficient $\kappa$ on the developing equilibrium interface. The effect of external loading is studied as well. Three initial configurations are assumed which are shown in Fig. 2.3. In the first configuration an initial condition is taken that is a stepwise change of the mass fraction in the middle of the domain. This configuration is used to investigate interface evolution. The second configuration consists of two tin regions next to each other to investigate coalescence. Here the initial interfaces are taken to be discontinuous as well. Finally, spinodal decomposition will be investigated starting from a homogeneous, eutectic, mass fraction distribution with random imperfections superposed.

Interface evolution

Calculations were performed to investigate the influence of the internal length scale parameter $\ell_c$ and the interface tension coefficient $\kappa$. When using the explicit formulation, see section 2.2.2, for calculating the nonlocal field, which is identical to the Cahn–Hilliard model, increasing $\kappa$ will have the same effect as increasing $\ell_c^2$. Both these parameters serve as a multiplication factor for the Laplacian of the local mass fraction. The implicit formulation (2.19) clearly distinguishes $\kappa$ from $\ell_c$. Substituting the nonlocality equation in the diffusion equation yields

$$\rho \frac{dc}{dt} = \vec{\nabla} \cdot \left( \rho \boldsymbol{M} \cdot \vec{\nabla} \left( \frac{\partial \Phi_0}{\partial c} + \gamma \nabla^2 \bar{c} + \psi \right) \right)$$

(2.64)

It seems here that the gradient energy coefficient $\gamma$, which equals $\kappa \ell_c^2$, is multiplication factor similar to that in the Cahn–Hilliard theory, however, $\bar{c}$ itself is influenced by $\ell_c$ through equation (2.27b). The length $\ell_c$ clearly controls the interface width over which the surface tension develops, whereas $\kappa$ sets the intensity of that interface tension.
Fig. 2.4 shows the local and nonlocal mass fraction for a reference calculation and for a calculation where $\ell_c$ is taken 4 times higher and $\kappa$ 16 times lower, i.e. the same value for the product $\gamma$ is retained. The latter case results in a steeper interface, Fig. 2.4a. This can be explained by the fact that the surface energy is directly dependent on $\kappa$, equation (2.23), and only indirectly on $\ell_c$, equation (2.27b). The interface is steeper but its interaction volume is larger (Fig. 2.4b) with a reduced surface tension. If $\gamma$ is kept constant and $\ell_c$ increases, then the energetic volume tends to increase. Since $\gamma$ has not changed, the increase of that volume is compensated by a narrowing of the interface. Fig. 2.4b shows that the nonlocal field is more diffuse for the larger value of $\ell_c$, indicating that the size of the region which contributes to the nonlocal effect is larger. Fig. 2.5a shows the interface width $\xi$ as a function of $\ell_c$ while keeping $\kappa \ell_c^2$ constant. The interface width is defined as the distance over which the mass fraction deviates 0.05 or more from the bulk mass fraction, either of the $\alpha$ or $\beta$-phase. If another value for the deviation is chosen the results are affected quantitatively but not qualitatively. Figs. 2.5b and 2.5c show the influence of $\ell_c$ and $\kappa$ on the interface width. It can be seen that $\xi$ is linearly dependent on both $\ell_c$ and $\sqrt{\kappa}$. It should be noted that the model is more sensitive to variations of $\sqrt{\kappa}$ than of $\ell_c$. This effect can also be seen in Fig. 2.5a.

The mass fraction of the bulk phases (binodal points, see Fig. 2.1) for a real system is not only determined by $\Phi_0$, but by the total energy $\Phi$ stored in the system. Surface energy

Figure 2.5 — Interface width as a function of (a) the internal length scale, $\kappa \ell_c^2 = \text{constant}$, (b) the internal length scale $\ell_c$, $\kappa = \text{constant}$, and (c) the square root of the interface tension coefficient $\kappa$, $\ell_c = \text{constant}$.
Φγ has no influence because it is by definition only present in interfaces and not in the bulk. However, the elastically stored energy Φe can change the binodal values of the mass fractions. To investigate this, external loading—both tension and compression—is applied to the system.

Fig. 2.6a shows the binodal value of the α-phase for the complete free energy function, i.e. including surface energy and elastically stored energy. The influence of the elastically stored energy causes the binodal value to decrease independent of the loading direction. The binodal value of the β-phase is influenced similarly, see Fig. 2.6b. Under the influence of straining the shape of the interface between the two phases also changes. The interface width decreases under the influence of either compression or tension and the maximum slope increases, as is shown in Fig. 2.7a. In this case the ratio Φγ/Φ is changed due to an increase of the total free energy, while Φγ remains the same.

**Coalescence**

To demonstrate the distinct influence of ℓc and κ on coalescence, simulations are performed starting from an initial condition of two tin regions in a Pb matrix with a sharp
interface, see Fig. 2.3b. Fig. 2.8a shows the development of the tin mass fraction distribution for two calculations with different values of $\ell_c$ and $\kappa$, but with $\gamma$ identical. It shows that for a higher value of $\ell_c$—or equivalently: a lower value of $\kappa$—the interface becomes more diffuse, as explained in section 2.5.1, and the speed at which the two regions coalesce increases. The reason for this increase can be found in the increase of the volume which contributes to the nonlocal free energy and because of which the two regions will sense each others presence more.

Fig. 2.8b shows the development of the tin mass fraction for higher values of $\gamma$. In this case it is noted that eliminating surface is more important than keeping the mass fraction of the bulk material at the binodal values $c^\alpha$ and $c^\beta$. For lower $\gamma$ (Fig. 2.8a) the two regions coalesce because they gradually move closer together, while in the case of high $\gamma$ ($\kappa = 640$ GJ/m$^3$ and $\ell_c = 6.25$ nm, i.e. $\gamma = 25 \cdot 10^{-6}$ J/m) the tin mass fraction in the existing regions is lowered in order to eliminate the Pb-rich region in between. This is caused by the fact that the increase of $\Phi_0$ is less than the decrease of $\Phi_\gamma$. After coalescence has occurred, the Sn-rich region also obtains its binodal value. Also coalescence occurs more readily in this case. If $\gamma$ is increased even more ($\kappa = 20$ GJ/m$^3$, $\ell_c = 100$ nm, $\gamma = 200 \cdot 10^{-6}$ Jm$^2$), surface tension becomes so high compared to $\Phi_0$ that it is energetically more favourable to have no surface. Therefore in this case a homogeneous solution is obtained.
Spinodal decomposition

One-dimensional simulations of phase separation and subsequent coarsening are next performed. The initial condition is a homogeneous distribution of the tin mass fraction at the eutectic composition of the Sn–Pb system (c = 0.63). To trigger spinodal decomposition random imperfections on the mass fraction are imposed (normal distribution with mean 0 and variance 0.01) as shown in Fig. 2.3c. The imperfections are the same for all calculations. First a reference calculation is performed with ℓ_c = 25 nm and κ = 20 GJ/m^3 which is shown in Fig. 2.9a and demonstrates the capability of the model to describe phase separation and coarsening.

The influence of the internal length parameter and the interface energy coefficient is also investigated for this case. Fig. 2.9 shows that a higher value for ℓ_c while keeping γ constant leads to faster decomposition of phases. Furthermore, initially more and smaller regions appear compared to the reference calculation. However, after 15 hours the structure does not appear to be less coarsened, indicating that more coalescence has already occurred compared to the reference case as was also expected after the investigation of the coalescence of the two tin regions, section 2.5.1. Increasing γ through either ℓ_c or κ, decreases the rate at which decomposition occurs, see Fig. 2.10a, and increases the size of the initial phase regions. Because of these large initial regions, the structure seems more coarsened after 15 hours compared to the reference case, Fig. 2.9a, but in fact almost no coarsening occurred after the decomposition stage.

A calculation with external loading of 10% imposed tensile strain is also performed to demonstrate the effect of the elastically stored energy in the system. The results are shown in Fig. 2.10b. In this case decomposition is faster, and the structure after 15 hours shows more coarsening. As predicted by the analysis of a single interface (Fig. 2.6a) the results also
show a shift of the binodal points, downward for the $\alpha$-phase and upward for the $\beta$-phase, and steeper interfaces compared to the reference calculation.

### 2.5.2 Two-dimensional coarsening

A two-dimensional simulation of a physical experiment has been performed next. For the experiment tin and lead were mixed at their eutectic composition, i.e. 0.63 mass fraction of tin. Next the specimen was quenched in liquid nitrogen, after which it was aged at 150°C for 288 hours during which time micrographs were taken of a single spot using a Scanning Electron Microscope (SEM). Typical micrographs obtained are shown in Fig. 1.5. For the simulation it is assumed that the specimen is cooled instantaneously from above the critical temperature to 150°C after which it is aged. A domain of $10 \times 10 \, \mu m$ has been modelled. The initial condition was obtained from the image analysis of the SEM micrograph of the as-solidified microstructure. For the finite element procedure 20,000 triangular elements have been used. All interpolation functions have been taken linear. Furthermore, use has been made of periodic boundary conditions for all variables.

The results of the simulation together with the SEM micrographs of the same area at the same time are shown in Fig. 2.11. The simulated microstructure exhibits growth and dissolution of the $\alpha$-phase along with break-up and coalescence events, similar to what can be observed experimentally.

For the determination of the unknown mobility and the subsequent comparison of the numerical simulations with the experimental data the results need to be quantified. Since minimisation of surface energy, and thus minimisation of surface area, is a driving force for...
the diffusion process, the total interface length per unit area, $\Gamma$, is taken as a quantifiable number to be compared. Another parameter which is often used to quantify the coarsening process is the average phase size. However since the domain which is modelled is rather small this parameter has not been used. After only two hours, most of the Pb-rich regions are already on the edge, and this prevents proper calculation of the average $\alpha$-phase size.

After quantifying both the experimental micrographs and the numerical simulation, the yet unknown mobility of lead in the tin matrix $M_\beta$ is first extracted from the results. This is done by fitting the development of $\Gamma$ for the simulation to that for the experiment using only the mobility, Fig. 2.13 shows the obtained surface length for both the experimental results and the simulation after fitting. The mobility is found to be $2.2 \cdot 10^{-25} \, m^2/(J \, s)$.

Next another simulation is performed to validate the model with the initial condition taken from another spot of the larger SEM micrograph. Fig. 2.12 also shows the computed surface length for this validation. The curve of the validation experiment does not start at the same value as the experiment because the phase distribution of the as-cast structure is not completely homogeneous. However it can be seen that during the ageing process the curve of the simulation approaches that of the experiment.
2.6 Conclusions

In this paper an extension of the Cahn–Hilliard model is presented to describe the microstructure evolution of Sn–Pb solder. The free energy density is taken to be a function of the local and a strongly nonlocal mass fraction which in this case is calculated by solving a Helmholtz partial differential equation. The difference between the nonlocal and the local mass fraction is associated with the surface energy. The variation with respect to local mass fraction of this energy acts as a driving force for the coarsening process. Incorporation of additional driving forces into the free energy is straightforward. In this way the effect of stresses on diffusion is also accounted for through the addition of the elastically stored energy. Next to introducing a strongly nonlocal measure in the free energy, this particular implementation has the advantage that higher level derivatives are absent in the formulation, facilitating the numerical solution.

The model includes an internal length scale $\ell_c$ and an interface tension coefficient $\kappa$ as parameters. Their product $\gamma = \kappa \ell_c^2$ now represents the gradient energy coefficient as used in the classical Cahn–Hilliard theory, even though it acts on the spatial variation of a nonlocal mass fraction (in the vicinity of the interface). One-dimensional simulations are used to emphasise the influence of these parameters on interface development and coarsening. It is shown that increasing either the internal length scale or the interface tension coefficient increases interface width and decreases the rate of phase separation. Increasing $\ell_c$ while changing $\kappa$ appropriately to keep $\gamma$ constant leads to steeper interfaces, whereas the area of influence of the interface is expanded and coalescence of two regions occurs more readily. Furthermore, it is shown that in this case phases separate out at a higher rate. These distinct features cannot be obtained with a classical Cahn–Hilliard theory, since there is only a single gradient energy coefficient present in that case.

The influence of external loading on the phase separation and coarsening process is also investigated. It is found that elastically stored energy, either due to tensile or compressive loading, causes a shift of the binodal points and decreases the interface width. Furthermore, it increases the decomposition rate and the coarsening rate.


Figure 2.13 — Experimental (left) and numerical (right) thermal ageing of Sn–Pb solder at 150°C, (size is $10 \times 10 \mu m$).
The numerical implementation of the nonlocal diffuse interface theory is outlined within the framework of the finite element method. The mobility is determined indirectly and the model is validated by comparing a two-dimensional simulation with experimental data. The model is able to predict break-up, coalescence, growth and dissolution of phase regions, similar to what can be observed experimentally. The total surface length per unit area is used to quantify the microstructure. The results are found to be in a reasonable agreement with the acquired experimental data.
A phase field model is presented to describe the mechanical behaviour of microstructure dependent materials, which is demonstrated by applying it to eutectic Sn–Pb solder. This solder material is known to be heavily influenced by its continuously evolving microstructure. For the constitutive behaviour of the different phases the elasto-viscoplastic Perzyna model has been used. It is coupled to the phase field model through the constitutive parameters which are taken dependent on the mass fraction field resulting from the solution of the phase field equations.

3.1 Introduction

Solder joints serve two important purposes. Firstly, they form the electrical connection between the component, e.g. a chip or resistor, and a substrate, e.g. a printed circuit board. Secondly, they constitute the mechanical bond that holds the component fixed to the substrate. During their life the joints are subjected to combined thermal and mechanical stresses which considerably impacts the fatigue lifetime. Therefore, their resistance to fatigue under thermal cycling is an important mechanical property of solder joints. Because of time limitations solder joints are often tested for their thermomechanical fatigue properties by subjecting them to accelerated test methods. However, during these accelerated experiments, damage mechanisms might be exhibited by the solder alloy which do not occur during normal usage. Reversely, some failure mechanisms are governed by time constants which are not reached in these test methods. Predictions based on accelerated cycling tests are therefore unreliable. Thermomechanical modelling could help to simulate the real life conditions of a solder joint and thus give a more accurate prediction of its lifetime. Furthermore these techniques permit a significant reduction in cost and time of the design of highly reliable soldered connections in electronic packaging and surface mount technology.

The service temperature $T$ of solder alloys is typically above half of their melting temperature $T_m$, i.e. their homologous temperature $T_H$, which is the ratio between $T$ and $T_m$ is greater than 0.5. Because of this the deformation behaviour is governed by high temperature mechanisms like creep, grain boundary sliding, and relaxation. Therefore, in literature when modelling the mechanical behaviour of solder alloys, use is generally made of viscoplastic material models [Amagai, 1999; Basaran and Chanderoy, 2000]. Numerous creep rate functions have been proposed, for which an extensive review can be found in Ju et al. [1994]. One of the commonly used viscoplastic models for solder alloys is the Anand model [Anand, 1985]. This model does not have an explicit yield condition, and thus there will be no discrete switching between elastic and viscoplastic behaviour. This should improve the numerical efficiency of the model. Furthermore, the model contains a single internal variable that represents the resistance against inelastic flow. A drawback of this model is that it contains many non-standard parameters which need to be determined. In this contribution the viscoplastic strain rate function defined by Perzyna [1966] is used to describe the time dependent mechanical behaviour [Van der Sluis et al., 1999]. In literature it is most commonly used to model secondary creep: [Ju et al., 1994; Basaran et al., 1998]. Its parameters are more transparent than those of the Anand model and adaptation of the hardening law is straightforward, leading to a highly flexible model. The standard Perzyna strain rate function does not account for grain size and temperature dependent creep activation energy and power law break down region of creep. It is well known that grain/phase size makes a significant difference in solder alloy behaviour, and that the overall joint properties are heavily affected by the microstructure [Basaran and Chanderoy, 1998; Basaran and Tang, 2002]. Vice versa, the evolution of the microstructure is affected by the stress state in the material. The Sn–Pb system is known to coarsen under the influence of thermal or thermomechanical loading [Hacke et al., 1998; Vianco et al., 1999; Matin et al., 2004]. Frear et al. [1988] found that regions of high shear strain within an eutectic Sn–Pb connection coarsened more. These regions of inhomogeneous coarsening are known to be crack nucleation sites during thermal cycling [Frear, 1989; Hacke et al., 1998].
Various attempts have been made to include a microstructural variable like the average grain or phase size in the mechanical model for a solder material [Frear et al., 1997; Basaran and Chanderoy, 1998; Vianco et al., 1999; Basaran and Chanderoy, 2000]. In the literature several equations can be found to describe grain size coarsening [Lifshitz and Slyozov, 1961; Clark and Aldan, 1973; Senkov and Myshlyaev, 1986], however no information about the distribution or shape of the grains is given. More information about the microstructure can be obtained by using a diffuse interface model [Cahn and Hilliard, 1958]. These models are based on the notion that the free energy is not only a function of a local order parameter, but also depends on the value of this parameter in its immediate neighbourhood. In this way the contribution of an interface to the free energy is included. Minimisation of the free energy then leads to coarsening of the system through the reduction of interfaces, either grain or phase boundaries [Leo et al., 1998; Zhu et al., 1999; Dreyer and Müller, 2000; Ni et al., 2002; Ubachs et al., 2004].

In various papers the influence of infinitesimal elastic strains on the evolution of the microstructure is investigated [Artemev et al., 2000; Dreyer and Müller, 2001; Zhu et al., 2001a; Ni et al., 2002]. In this paper, however, a phase field model is coupled to a finite strain elasto-viscoplastic model to investigate the impact of the underlying microstructure. First, the evolution of the microstructure is dealt with using a phase field model based on a strongly nonlocal mass fraction which accounts for the evolution under thermal loading [Ubachs et al., 2004]. The phase field provides extensive information on the microstructure: the mass fractions of the individual phases, their shape and orientation, and position and shape of interfaces between phases. This information is further used to determine local material parameters for the constitutive material model applied, i.e. a viscoplastic Perzyna model. In this way a microstructure dependent mechanical model for multiphase alloys is obtained. A demonstration of the model is given by its application to eutectic Sn–Pb solder.

First, in section 3.2, the phase field model will be introduced which is used to calculate the distribution of the phases in the system. Next, the viscoplastic material model used for each of the individual phases is treated in section 3.3, followed by some aspects of the numerical implementation in section 3.4. Section 3.5 deals with the choice of the material parameters. In section 3.6 numerical simulations of mechanical loading of a Sn–Pb solder alloy with different microstructure and strain rates are discussed, after which the conclusions will be presented in section 3.7.

### 3.2 Phase field model

In order to describe the microstructure and its evolution a phase field model is used. The mass fraction $c$ is taken as the field variable, and it is assumed that it varies continuously throughout the system. The evolution of the mass fraction field is obtained by solving the nonlinear diffusion equation:

$$
\frac{d c}{d t} = -\nabla \cdot \rho M \cdot \nabla \frac{\delta \Phi}{\delta c},
$$

(3.1)

where $\rho$ is the mass density, $t$ is time, $M$ the mobility tensor and $\Phi$ the free energy. The variation of the free energy with respect to the mass fraction is the chemical potential and is the driving force behind the diffusion process: it determines how the microstructure
will evolve. For the moment the free energy is assumed to consist out of two parts: the configurational energy, which determines the equilibrium mass fractions for the different phases, and the interface energy which controls the number of interfaces, their size, and orientation.

The free energy function which is the starting point for finding an expression for the mass flux is taken from Giacomin and Lebowitz [1997], who derived a macroscopic equation describing phase segregation in microscopic model systems with long-range interactions evolving according to stochastic Kawasaki dynamics with nearest neighbour exchanges (see also Brandon [1994]). They obtained the following expression for the specific free energy:

\[
\Phi = \Phi_0(c(\vec{x})) + \frac{1}{4} \int_\Omega g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right]^2 d\vec{y},
\] (3.2)

where \( \Omega \) is the region around the material point with position vector \( \vec{x} \), \( \vec{y} \) a local coordinate vector with respect to \( \vec{x} \) pointing to points in \( \Omega \), \( g \) is the nonlocal kernel (a symmetric smooth function \( (C^\infty) \)), and \( \Phi_0 \) is the equilibrium free energy density of a homogeneous system. Similar nonlocal extensions of the free energy have been used by Gajewski and Zacharias [2003] and Fosdick and Mason [1998].

In this work the introduced nonlocality which accounts for interface effects is assumed to be Lagrangian (material) in nature, i.e. the volume over which long range interactions occur is defined by the material configuration. Using the specific free energy (3.2) and taking the variation with respect to \( c(\vec{x}) \) in order to obtain the chemical potential \( \mu \) then gives:

\[
\mu = \frac{\delta \Phi}{\delta c(\vec{x})} = \frac{\partial \Phi_0}{\partial c} + \frac{1}{2} \int_{\Omega_0} g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right] d\vec{y},
\] (3.3)

which can be rewritten to yield:

\[
\mu = \frac{\partial \Phi_0}{\partial c} + \kappa [c(\vec{x}) - \bar{c}(\vec{x})],
\] (3.4)

where \( \kappa \) is the interface tension coefficient. In here, the nonlocal mass fraction \( \bar{c} \) is defined as

\[
\bar{c}(\vec{x}) = \int_{\Omega_0} g'(\vec{y}) c(\vec{x} + \vec{y}) d\vec{y}, \quad \kappa = \frac{1}{2} \int_{\Omega_0} g(\vec{y}) d\vec{y}, \quad \text{and} \quad g' = \frac{g}{\kappa}.
\] (3.5)

Eq.(3.5) can be reformulated towards an implicit Helmholtz equation [Ubachs et al., 2004]:

\[
\bar{c} - \ell^2 \nabla_0^2 \bar{c} = c,
\] (3.6)

where \( \nabla_0^2 \) represents Laplacian taken in the material (Lagrangian) configuration, and \( \ell \) is an internal length parameter, which, when using an isotropic weighting function, which is only dependent on a radial coordinate \( r \), is defined as

\[
\ell^2 = \frac{1}{2\Omega} \int_{\Omega_0} r^2 g(r) dr.
\] (3.7)

Note that the solution of this partial differential equation indeed leads to a strongly nonlocal solution as assumed in (3.5). Details on this particular topic have been elaborated within the context of strongly nonlocal damage [Peerlings et al., 2001].
explicit format of the nonlocal kernel (which is the result of the adopted mathematical approximation) can be found in this reference as well.

The reformulation of the integral expression (3.5) into differential equation (3.6) requires the introduction of boundary conditions. Either the value of \( \bar{c} \), its normal derivative, or a linear combination of these quantities must be specified on the boundary. Here the homogeneous Neumann boundary condition has been used:

\[
\vec{V}_0 \bar{c} \cdot \vec{n} = 0, \quad (3.8)
\]

where \( \vec{n} \) is the normal vector of the boundary. This condition preserves the global mass fraction of the entire system in the nonlocal averaging:

\[
c_{\text{total}} = \frac{1}{\Omega_0} \int_{\Omega_0} c \, d\vec{x} = \frac{1}{\Omega_0} \int_{\Omega_0} \bar{c} \, d\vec{x}. \quad (3.9)
\]

More comments on this boundary condition can be found in Peerlings et al. [2001]. The complete nonlocal phase field model now reads

\[
\rho \frac{dc}{dt} = \nabla \cdot (\rho \mathbf{M} \cdot \nabla (\frac{\partial \Phi_0}{\partial c} - \kappa (\bar{c} - c))), \quad (3.10)
\]

\[
\bar{c} - \ell^2 \nabla^2_o \bar{c} = c. \quad (3.11)
\]

More information on the diffusional phase field model can be found in Ubachs et al. [2003, 2004].

The solution of the system of equations yields detailed information on the distribution of phases in the material, the position and orientation of interfaces, and the mass fractions in the separate phases. The mechanical behaviour of the material is dependent on its microstructure. The various phases have different mechanical properties and therefore stresses and strains will not be homogeneous throughout the material. Knowledge of the mass fraction field allows to account for the heterogeneity of the material. The differences in constitutive behaviour of the various phases and interfaces can be dealt with in a continuum manner.

The eutectic Sn–Pb system is used to demonstrate the suggested approach. The same constitutive model is assumed for both the Pb-rich \( \alpha \)-phase and the Sn-rich \( \beta \)-phase. However, the parameters used are different for each phase.

### 3.3 Hyperelasto-viscoplastic single phase modelling

#### 3.3.1 Kinematics

Transformation from the undeformed configuration at time \( t_0 \) to the current configuration at time \( t \) is described by the standard deformation gradient tensor \( \mathbf{F} \), whose determinant \( J = \det(\mathbf{F}) \) represents the relative volume change. The conventional multiplicative decomposition is used to split the deformation into an elastic and inelastic (viscoplastic) part, denoted by subscripts \( e \) and \( \nu p \) respectively:

\[
\mathbf{F} = (\vec{V}_0 \vec{x})^e = \mathbf{F}_e \cdot \mathbf{F}_{\nu p}. \quad (3.12)
\]
Figure 3.1 — Schematic one-dimensional representation of the elasto-viscoplastic Perzyna model

The right and left Cauchy-Green strain tensors, $C$ and $B$, and the Green-Lagrange strain tensor $E$ are classically given by

$$
C = F^c \cdot F, \quad B = F \cdot F^c, \quad \text{and} \quad E = \frac{1}{2}(C - I). \quad (3.13)
$$

Material velocity is taken into account by the deformation rate and spin tensors $D$ and $\Omega$, the symmetric and skewsymmetric parts of the velocity gradient tensor $L$, respectively. Decomposition (3.12) implies the introduction of elastic and viscoplastic rate tensors:

$$
L = (\nabla \mathbf{v})^c = \dot{F} \cdot F^{-1} = \dot{F}_e \cdot F_e^{-1} + F_v \cdot \dot{F}_v \cdot F_v^{-1} = \dot{L}_e + L_{vp} = (D_e + \Omega_e) + (D_{vp} + \Omega_{vp}). \quad (3.14)
$$

To make the decomposition unique it is commonly assumed that the viscoplastic spin $\Omega_{vp}$ is zero.

### 3.3.2 The Perzyna model

The one-dimensional mechanical representation of the elasto-viscoplastic Perzyna model consists of a spring, a linear viscous dashpot, a hardening spring and a friction slider, see Fig. 3.1. The elastic spring represents the elastic part of the material response. The viscoplastic response, represented by the viscous dashpot becomes manifest as soon as the stress exceeds a characteristic value and the plastic slider gives way.

For the three-dimensional case, the material is assumed to behave viscoplastically after an equivalent stress measure exceeds the current yield stress $\tau_y$. Here the Von Mises equivalent stress is used which reads in terms of the Kirchhoff stress tensor $\tau$:

$$
\tau_{eq} = \sqrt{\frac{3}{2} \tau^d : \tau^d}, \quad (3.15)
$$

with $\tau^d$ the deviatoric part of the stress tensor. The evaluation of the occurrence of yield is commonly evaluated through a yield criterion $\varphi$. It can be represented as a yield surface in stress space. The classical Van Mises-Huber yield function $\varphi$ is used to evaluate the stress state and to check whether the deformation is purely elastic ($\varphi < 0$) or viscoplastic ($\varphi \geq 0$):

$$
\varphi(\tau, \varepsilon_{vp}) = \tau_{eq} - \tau_y(\tau_{y0}, \varepsilon_{vp}, \dot{\varepsilon}_{vp}). \quad (3.16)
$$

In here, $\tau_y$ accounts for linear or nonlinear hardening or softening laws. The initial yield stress is denoted by $\tau_{y0}$ while the hardening behaviour will be related to the effective
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Viscoplastic strain measure \( \varepsilon_{vp} \) and its rate \( \dot{\varepsilon}_{vp} \). The effective viscoplastic strain is obtained through integration over the loading history of its time derivative:

\[
\varepsilon_{vp} = \int_{0}^{t} \dot{\varepsilon}_{vp} \, dt. \tag{3.17}
\]

### 3.3.3 Hyperelastic model

A hyperelastic model is chosen to correctly describe the elastic part of the material behaviour at finite deformations. In that case it is assumed that an elastic strain energy function \( \Phi_e(E) \) exists, which can be used to calculate the stresses. Here we use it to relate the 2nd Piola-Kirchhoff stress tensor \( S \) to the right Cauchy–Green strain tensor \( C \):

\[
S = J F^{-1} \cdot \sigma \cdot F^{-c} = F^{-1} \cdot \tau \cdot F^{-c} = \frac{\partial \Phi_e}{\partial E} = 2 \frac{\partial \Phi_e}{\partial C}, \tag{3.18}
\]

where \( \sigma \) is the Cauchy stress tensor. Since the elastic part of the constitutive model is needed in rate form, differentiation with respect to time is performed, yielding

\[
\dot{S} = 2 \frac{\partial^2 \Phi_e}{\partial C^2} : \dot{C}, \tag{3.19}
\]

In order to formulate the model in the current configuration Eq.(3.19) is pushed forward which now can be written in terms of the Kirchhoff stress as

\[
\nabla \mathbf{\tau} = F \cdot \dot{S} \cdot F^{-c} = 4F \cdot \left[ \frac{\partial^2 \Phi_e}{\partial C^2} : (F^c \cdot D \cdot F) \right] \cdot F^c = \mathbb{H} : D, \tag{3.20}
\]

where \( \mathbb{H} \) is the fourth order constitutive tensor which is determined by the choice of the elastic energy function, and \( \nabla \mathbf{\tau} \) is the objective Truesdell derivative

\[
\nabla \mathbf{\tau} = \dot{\mathbf{\tau}} - \mathbf{L} \cdot \mathbf{\tau} - \mathbf{\tau} \cdot \mathbf{L}^c \quad \text{with} \quad \mathbf{L} = \dot{F} \cdot F^{-1}. \tag{3.21}
\]

An elastic energy function is adopted, which characterises isotropic, compressible material behaviour [Perić, 1992]:

\[
\Phi_e = \frac{1}{2} G [\mathbf{C} : \mathbf{I} - 3 - 2 \ln(J)] + \frac{1}{2} K \ln^2(J), \tag{3.22}
\]

where \( G \) and \( K \) are Lamé's constants:

\[
G = \frac{E}{2(1 + \nu)} \quad \text{and} \quad K = \frac{\nu E}{(1 + \nu)(1 - 2\nu)}, \tag{3.23}
\]

with \( E \) and \( \nu \) the Young's modulus and Poisson's ratio respectively. The first derivative of Eq.(3.22) with respect to \( C \) then becomes

\[
\frac{\partial \Phi_e}{\partial C} = \frac{1}{2} G (I - C^{-1}) + \frac{1}{2} K \ln(J) C^{-1}, \tag{3.24}
\]
and its second derivative\(^1\)

\[
\frac{\partial^2 \Phi_e}{\partial C^2} = \frac{1}{2} [G - K \ln(J)] C^{-1} : [\mathbf{I}^c - \frac{1}{4} K C^{-1} C^{-1}] .
\] (3.26)

This then leads to the following expression for \( \mathbf{H} \), Eq.(3.20),

\[
\mathbf{H} = 2 [G - K \ln(J)] \mathbf{I}^c + K \mathbf{I}.
\] (3.27)

### 3.3.4 Viscoplastic model

During viscoplastic deformation the yield stress will change as a function of the effective viscoplastic strain. Plastic flow does not induce any stresses in the material, making use of the additive decomposition of the deformation rate tensor, the constitutive model then becomes

\[
\mathbf{\nabla} \mathbf{\tau} = \mathbf{H} : \mathbf{D} = \mathbf{H} : (\mathbf{D} - \mathbf{D}_{vp}),\] (3.28)

With ongoing viscoplastic deformation the direction of the viscoplastic strain rate \( \mathbf{D}_{vp} \) is defined by the commonly used normality or associative flow rule which states that the strain rate is directed along the normal to the yield surface and denoted by \( \partial \phi / \partial \mathbf{\tau} \). Its length is characterised by the rate of the viscoplastic multiplier \( \dot{\lambda} \). Thus the expression for the viscoplastic strain rate can be formulated as [Perzyna, 1985]:

\[
\mathbf{D}_{vp} = \dot{\lambda} \mathbf{N}, \quad \text{where} \quad \mathbf{N} = \frac{\partial \phi}{\partial \mathbf{\tau}} = \frac{3}{2} \frac{\mathbf{\tau}_d}{\mathbf{\tau}_{eq}}, \quad \text{and} \quad \dot{\lambda} = \eta \phi(\phi),\] (3.29)

with \( \eta \) a fluidity parameter which determines the magnitude of the viscoplastic strain rate and \( \phi(\phi) \) the overstress function. Viscoplastic strains are nonzero only if \( \phi \geq 0 \), i.e. for stress states on or outside the yield surface. A power law is chosen for the overstress function [Perzyna, 1971]:

\[
\phi(\phi) = \left( \frac{\phi}{\phi_0} \right)^N = \left( \frac{\mathbf{\tau}_{eq} - \mathbf{\tau}_y}{\phi_0} \right)^N,
\] (3.30)

with \( N \) the rate sensitivity parameter and \( \phi_0 \) is a scaling parameter. A higher value of \( N \) reduces the strain rate sensitivity of the model [Perić, 1993].

The relation between the rate of the hardening variable \( \dot{\epsilon}_{vp} \) and of the viscoplastic multiplier \( \dot{\lambda} \) is set to the straightforward relation:

\[
\dot{\epsilon}_{vp} = \dot{\lambda} = \sqrt{\frac{2}{3} \mathbf{D}_{vp} : \mathbf{D}_{vp}}.
\] (3.31)

The hyperelastic material model returns the stress when the elastic deformation is known. However, as can be seen again from the mechanical model, the elastic deformation has

\[^1\] Here use has been made of the following tensor identity:

\[
\frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}} = -\mathbf{C}^{-1} : \mathbf{\tau}^c : \mathbf{C}^{-1}.
\] (3.25)
to be isolated from the total deformation, which is generally known during an analysis. This is only possible if the viscoplastic stress can be evaluated. As is described above, the viscoplastic strain rate is related to the stress by the flow rule. An integration procedure is needed to calculate the viscoplastic strain.

In case of viscoplastic behaviour, i.e. \( \varphi \geq 0 \) the model now consists of the following system of equations:

\[
\begin{align*}
\nabla \tau &= H : (D - \dot{\lambda} N), \\
\dot{\lambda} &= \eta \left( \frac{\tau_{eq} - \tau_y}{\varphi_0} \right)^N.
\end{align*}
\]

(3.32a)

(3.32b)

In case of fully elastic behaviour, i.e. \( \varphi < 0 \), \( \dot{\lambda} \) will equal zero and only Eq.(3.32a) needs to be solved.

### 3.3.5 Phase field coupling

As mentioned earlier each phase is assumed to obey the same constitutive law, only the parameters differ. This means that the elastically and viscoplastically stored energy \( \Phi_e \) and \( \Phi_{vp} \) are dependent on the mass fraction \( c \). In this way information on the microstructure obtained with the phase field model is included in the constitutive model. In their turn, the mechanically stored energies will influence the phase field and its evolution. The chemical potential now reads

\[
\mu = \frac{\partial \Phi_0}{\partial c} + \frac{\partial \Phi_e}{\partial c} + \frac{\partial \Phi_{vp}}{\partial c} + \kappa [c - \bar{c}].
\]

(3.33)

However, in this contribution the microstructure evolution is evaluated separately from the mechanical part of the model. Therefore, the influence of the mechanically stored energies on the diffusion is not taken into account.

### 3.4 Finite element implementation

#### 3.4.1 Momentum equation

In the absence of body/volumetric forces, the momentum equation is given by the zero divergence of the Cauchy stress tensor:

\[
\nabla \cdot \sigma = 0.
\]

(3.34)

The gradient operator in this equation is the spatial, or Eulerian gradient operator, i.e. it is taken in the current configuration. Multiplying the equation with a vectorial weighting function \( \tilde{w} \) and integrating over the domain \( \Omega \) with a boundary \( \Gamma \) whose unit outward normal is \( \tilde{n} \), yields

\[
\int_{\Omega} \tilde{w} \cdot (\nabla \cdot \sigma) \, d\Omega = 0 \quad \forall \tilde{w}.
\]

(3.35)
Applying the divergence theorem, using $\sigma = \sigma^c$ and applying the natural boundary condition $\sigma \cdot \hat{n} = \hat{\rho}$ on $\Gamma$ leads to

$$\int_{\Omega} (\nabla \tilde{w})^c : \sigma \, d\Omega = \int_{\Gamma} \tilde{w} \cdot \hat{\rho} \, d\Gamma. \quad (3.36)$$

Because of the nonlinear and time dependent nature of the stress this equation needs to be solved iteratively by using a standard Newton–Raphson iteration procedure. First the left hand side is transformed to the last known equilibrium configuration at time $t$ according to $\tilde{\mathbf{V}} = \mathbf{F}_\Delta \cdot \tilde{\mathbf{V}}_t$ (with $\mathbf{F}_\Delta = \mathbf{F} \cdot [\mathbf{F}^t]^{-1}$ the incremental deformation gradient tensor) and $d\Omega = (J/J^t) \, d\Omega^t$, and using $\mathbf{r} = f \, \sigma$ results in

$$\int_{\Omega^t} (\tilde{\mathbf{V}}_t \tilde{w})^c : \mathbf{F}_\Delta^{-1} \cdot \mathbf{r} \frac{1}{J^t} \, d\Omega^t = \int_{\Gamma} \tilde{w} \cdot \hat{\rho} \, d\Gamma. \quad (3.37)$$

Next, the unknown Kirchhoff stress tensor and incremental deformation gradient tensor are decomposed in an iterative manner,

$$\mathbf{r} = \mathbf{r}^{(i+1)} = \mathbf{r}^{(i)} + \delta \mathbf{r} \quad \land \quad \mathbf{F}_\Delta = \mathbf{F}_\Delta^{(i+1)} = \mathbf{F}_\Delta^{(i)} + \delta \mathbf{F}_\Delta, \quad (3.38)$$

leading to the following linearised weak formulation of the equilibrium condition

$$\int_{\Omega^t} \left[ \left( \tilde{\mathbf{V}}_t \tilde{w} \right)^c : \left( [\mathbf{F}_\Delta]^{-1} \cdot \mathbf{r}^{(i)} + [\mathbf{F}_\Delta^{(i)}]^{-1} \cdot \delta \mathbf{r} \right) \right] \frac{1}{J^t} \, d\Omega^t = \int_{\Gamma} \tilde{w} \cdot \hat{\rho}^{(i+1)} \, d\Gamma - \int_{\Omega^t} \left( \tilde{\mathbf{V}}_t \tilde{w} \right)^c : \left( [\mathbf{F}_\Delta^{(i)}]^{-1} \cdot \mathbf{r}^{(i)} \right) \frac{1}{J^t} \, d\Omega^t. \quad (3.39)$$

Introducing the variational deformation gradient tensor

$$\mathbf{L}_\delta^c = (\tilde{\mathbf{V}} \delta \vec{x})^c = \delta \mathbf{F} \cdot \mathbf{P}^{-1} = \delta \mathbf{F}_\Delta \cdot \mathbf{F}_\Delta^{-1}, \quad (3.40)$$

allows for rewriting $\delta [\mathbf{F}_\Delta^{-1}]$ as $-[\mathbf{F}_\Delta^{(i)}]^{-1} \cdot \delta \mathbf{F}_\Delta \cdot [\mathbf{F}_\Delta^{(i)}]^{-1} = -[\mathbf{F}_\Delta^{(i)}]^{-1} \cdot \mathbf{L}_\delta^c$. Transformation back to the last known configuration then results in

$$\int_{\Omega} (\nabla \tilde{w})^c : \left[ -\mathbf{I} \cdot \mathbf{r}^{(i)} : \mathbf{L}_\delta^c + \delta \mathbf{r} \right] \frac{1}{J^{(i)}} \, d\Omega = \int_{\Gamma} \tilde{w} \cdot \hat{\rho}^{(i+1)} \, d\Gamma - \int_{\Omega} (\nabla \tilde{w})^c : \mathbf{r}^{(i)} \frac{1}{J^{(i)}} \, d\Omega. \quad (3.41)$$

### 3.4.2 Incremental time discretisation

To solve Eq. (3.41), an expression for the Kirchhoff stress tensor is needed. It can be derived from the constitutive equations, system (3.32). Since this system can not be integrated analytically, an incremental time integration scheme will be used. The system is formulated in terms of invariant variables, which remain unaltered under superposed spatial rigid body motions, thereby preserving the principle of objectivity. This ensures that no spurious stresses are generated due to rigid body motions. An implicit (or backward) Euler scheme is used, resulting in

$$\Delta \mathbf{S} = \mathbf{S} - \mathbf{S}' = \frac{1}{2} \mathbf{H} \cdot \left( \mathbf{C} - \mathbf{C}' - 2 \mathbf{F}^c \cdot \Delta \lambda \mathbf{N} \cdot \mathbf{F} \right), \quad (3.42a)$$

$$\Delta \lambda = \lambda - \lambda' = \Delta t \eta \left( \frac{T_{eq} - T_y}{\varphi_0} \right)^N, \quad (3.42b)$$
which can be rewritten in terms of the Kirchhoff stress by applying a push forward towards the current configuration yielding

\[ \tau = F_\Delta \cdot \tau^t \cdot F_\Delta^c + \mathbb{H} : [e_\Delta - \Delta \lambda N], \]  
(3.43a)

\[ \Delta \lambda = \Delta t \eta \left( \frac{\varphi}{\varphi_0} \right)^N, \]  
(3.43b)

where \( e_\Delta \) is the incremental Almansi strain tensor

\[ e_\Delta = \frac{1}{2} (I - F_\Delta^{-c} \cdot F_\Delta^{-1}). \]  
(3.44)

It now remains to relate the variation of the Kirchhoff stress Eq.(3.43) to \( L^c_\delta \):

\[ \delta \tau = C : L^c_\delta. \]  
(3.45)

In the following section the consistent tangents will be derived for the elastic and the viscoplastic case.

### 3.4.3 Consistent tangent

#### Elastic increment

For a fully elastic increment, i.e. \( \varphi < 0 \) the incremental update for the Kirchhoff stress tensor reads:

\[ \tau = F_\Delta \cdot \tau^t \cdot F_\Delta^c + \mathbb{H} : e_\Delta. \]  
(3.46)

The variation of the stress then becomes:

\[ \delta \tau = \delta F_\Delta \cdot \tau^t \cdot F_\Delta^c + F_\Delta \cdot \tau^t \cdot \delta F_\Delta^c + \frac{\partial \mathbb{H}}{\partial J} : e_\Delta \delta J + \mathbb{H} : (\delta F_\Delta^{-c} \cdot F_\Delta^{-1}) : \delta e_\Delta, \]  
(3.47)

which after some manipulations, see appendix B.1, can be rewritten in the form of Eq.(3.45) with

\[ C = C_e = 2 \mathbb{S} \cdot F_\Delta \cdot \tau^t \cdot F_\Delta^c - 2 \mathbb{K} \cdot e_\Delta I + \mathbb{H} : \left( \mathbb{S} \cdot F_\Delta^{-c} \cdot F_\Delta^{-1} \right). \]  
(3.48)

#### Viscoplastic increment

For a viscoplastic increment (\( \varphi > 0 \)) the incremental update of the Kirchhoff stress tensor reads:

\[ \tau = F_\Delta \cdot \tau^t \cdot F_\Delta^c + \mathbb{H} : (e_\Delta - \Delta \lambda N). \]  
(3.49)

The hardening law is next assumed to be a function of \( \lambda \) only, i.e. the yield stress can be a function of the viscoplastic multiplier and/or its rate, \( \tau_y(\lambda, \dot{\lambda}) \). After some manipulations, see appendix B.2, the variation of the stress can be rewritten as

\[ \delta \tau = N^{-1} : (C_e + \mathbb{T}) : L^c_\delta \equiv C_{vp} : L^c_\delta, \]  
(3.50)
with $C_e$ as defined in Eq.(3.48), and the fourth order tensors $T$ and $N$ as

$$T = 2\Delta \lambda K \frac{1}{J} NI,$$  \hfill (3.51)

and

$$N = I + c_2 H : NN + H : \Delta \lambda \frac{1}{\tau_{eq}} \left( \frac{3}{2} - \frac{1}{2} II - NN \right),$$  \hfill (3.52)

where the coefficients $c_1$ and $c_2$ equal

$$c_1 = N \Delta t \frac{\eta}{\phi_0} \left( \frac{\tau_{eq} - \tau_y}{\phi_0} \right)^{N-1} \quad \text{and} \quad c_2 = \frac{c_1}{1 + \frac{\partial \tau_y}{\partial \Delta \lambda} c_1}. \hfill (3.53)$$

### 3.4.4 Stress update algorithm

Every increment is initially assumed to be fully elastic. The stresses obtained in this elastic predictor state are then used to check the yield criterion. If this has been violated return mapping has to be performed, i.e. the amount and direction of plastic flow has to be determined and the stresses should be updated accordingly. The return mapping requires the set of nonlinear Eqs.(3.43) to be solved. This will be done using the previously described standard Newton–Raphson iterative procedure.

It can be shown that for the current hyperelastic model the direction of plastic flow is fully determined by the elastic predictor state, $N = N^*$, see appendix B.3. Furthermore, for the stress update algorithm the nodal displacements are known and fixed, except for the plane stress case. Therefore the volume and the stresses of the elastic predictor will not change, i.e. $\delta J$ and $\delta^* \tau$ are zero. Decomposing system (3.43) in an iterative manner, $(\cdot)^{(i+1)} = (\cdot) + \delta(\cdot)$, thus results in

$$\mathbb{0} : \delta \tau + H : N \delta \lambda = \tau - \bar{\tau} - H : N \Delta \lambda,$$  \hfill (3.54a)

$$\delta \Delta \lambda - c_1 (N : \delta \tau - \delta \tau_y) = \Delta t \eta \left( \frac{\phi}{\phi_0} \right)^N - \Delta \lambda,$$  \hfill (3.54b)

with

$$c_1 = \frac{\Delta t \eta N \left( \frac{\phi}{\phi_0} \right)^{N-1}}{\phi_0}.$$  \hfill (3.54c)

See appendix B.3 for details. Since it is assumed that the incremental hardening law is only a function of $\Delta \lambda$ the system that needs to be solved becomes

$$\mathbb{0} : \delta \tau + H : N \delta \lambda = \tau - \bar{\tau} - H : N \Delta \lambda,$$  \hfill (3.55a)

$$-c_1 N : \delta \tau + (1 + c_1 \frac{\partial \tau_y}{\partial \Delta \lambda}) \delta \Delta \lambda = \Delta t \eta \left( \frac{\phi}{\phi_0} \right)^N - \Delta \lambda.$$  \hfill (3.55b)

The system can also be solved in an uncoupled manner, first Eq.(3.43b) is solved, next the stress is updated by evaluating the linear equation (3.43a). Linearisation of Eq.(3.43b) yields

$$\left[ 1 + c_1 \left( N : H + \frac{\partial \tau_y}{\partial \Delta \lambda} \right) \right] \delta \Delta \lambda = \Delta t \eta \left( \frac{\phi}{\phi_0} \right)^N - \Delta \lambda.$$  \hfill (3.56)
3.5 Material parameters

In order to minimise the global energy, a homogeneous Sn–Pb mixture will start to phase separate. From the free energy function for the system, see Fig. 3.2, it can be deduced that the preferred state of material is a system consisting of $\alpha$ and $\beta$ phase regions with mass fractions corresponding to the binodal points. In this state the configurational energy of the total system is at its lowest point. The binodal points can be found by constructing a double tangent to the free energy curve. For more details on this subject see Ubachs et al. [2004]. It follows that, with the exception of the interfaces, the Sn–Pb system consists only of $\alpha$ and $\beta$ phase regions, whose compositions are determined by the corresponding binodal points.

No direct mechanical data is available on the separate phases. Therefore, the required parameters are extracted from measurements performed on multiphase solder joints. First the material parameters for the bulk solder are determined. Next, a linear extrapolation is performed to find the required parameters for the $\alpha$ phase containing 0% Sn and the $\beta$ phase containing 0% Pb. To find the material parameters $\eta$, $N$, $\tau_y$, and $\varphi_0$ for the Perzyna model, saturation stress versus inelastic strain curves are reproduced from the parameters for the Anand model given in Cheng et al. [2000] and Wang et al. [2001] who exploited data provided by Darveaux and Banerji [1992]. In the experiments performed by Darveaux and Banerji [1992] shear deformations up to 50% for 60Sn–40Pb and 130% for 2.5Sn–97.5Pb were recorded before the onset of damage.

From the experimental stress strain curves at constant strain rates it can be seen that the hardening is strain rate dependent. Therefore the following evolution law for the yield stress is chosen

$$\tau_y = \tau_{y0} + h_1 \varepsilon_{vp}^{h_2} [1 - \exp(-h_3 \varepsilon_{vp})],$$

(3.57)

where $h_1$, $h_2$, and $h_3$ are hardening parameters. The derivative of the yield stress with respect to the viscoplastic multiplier needed for Eqs.(3.56) and (3.53) then reads (after applying a backward Euler time discretisation)

$$\frac{\partial \tau_y}{\partial \varepsilon_{vp}} \frac{\partial \varepsilon_{vp}}{\partial \Delta \lambda} = \frac{h_1}{\Delta t} \varepsilon_{vp}^{h_2-1} \left[ h_2 - (h_2 - h_3 \Delta \lambda) \exp(-h_3 \varepsilon_{vp}) \right].$$

(3.58)
The parameter determination will be done from stress strain curves obtained at constant strain rates. In that case the following expression for the stress during a uniaxial tensile test at constant strain rate can be found using Eqs.(3.29)–(3.31):

$$\tau_{eq} = \varphi_0 \left( \frac{\dot{\varepsilon}_{vp}}{\eta} \right)^{1/N} + \tau_{y0} + h_1 \dot{\varepsilon}_{vp} \left[ 1 - \exp(-h_3 \dot{\varepsilon}_{vp}) \right].$$

(3.59)

**Figure 3.3** — Fitting of Perzyna parameters, stress versus inelastic strain, dotted lines are fitted to the data published by Cheng et al. [2000].

Fig. 3.3 shows the material data from Cheng et al. [2000] and the fitted curves for two Sn–Pb systems with different mass compositions. First the $\eta$, $N$, $\tau_{y0}$, and $\varphi_0$ are determined by fitting. They determine the initial yield stress –indicated in the figures by a circle– which is highly dependent on the strain rate. These parameters are then held constant while the hardening parameters are fitted. The fitted parameters for both compositions are summarised in table 3.1.

Because the phase field model provides a continuous field for the mass fraction $c$ ranging from 0 to 1, material parameters need to be known for all $0 \leq c \leq 1$. In a first-order approximation, all parameters are assumed to be linearly dependent on the mass fraction $c$, e.g. $E = (1 - c) E_{Pb} + c E_{Sn}$. Parameter values for the individual components should correspond to the parameters found with the fitting procedure above when using this linear rule to calculate properties for their specific composition. This assumption then allows for extracting material parameters for the individual phases. The applied first-order rule is only valid nearby the $\alpha$ and the $\beta$ phases, which are the phases actually present in the multi-phase mixture. Note that the values for the $\alpha$ phase with 0% Sn ($\alpha_0$) and the $\beta$ phase containing 0% Pb ($\beta_0$) can differ considerably from values for the pure elements, Pb and Sn. They are however used here only to obtain the correct interpolated values. The parameters as used for the finite element calculations are shown in table 3.1.
Table 3.1 — Material parameters for the Sn–Pb solder system.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>60-40</th>
<th>0.025-97.5</th>
<th>β₀</th>
<th>α₀</th>
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<tr>
<td>Young’s modulus</td>
<td>E</td>
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<td>0.33</td>
<td>0.44</td>
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<td>1.98 · 10⁻⁵</td>
<td>5.52 · 10⁻⁶</td>
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<td>h₂</td>
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<td></td>
<td>h₃</td>
<td>[—]</td>
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<td>13.53</td>
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<td>[nm]</td>
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3.6 Numerical Results

Simulations of mechanical loading have been performed on different microstructures which are assumed not to evolve during the loading. This can be assumed for this particular test since the mechanical time scale is very small compared to the diffusion time scale. The microstructures are obtained by solving the phase field equations discussed in section 3.2. Here the results of a simulation for static ageing at 150°C have been used as microstructural input for the mechanical model. For these calculations the mechanical component of the free energy has been neglected, i.e. mechanical loading is assumed not to influence the diffusion process. The initial conditions for these calculations are taken from a scanning electron micrograph of as-cast eutectic Sn–Pb solder [Matin et al., 2004]. Quantitative comparison between the simulation and the experiment showed a good agreement [Ubachs et al., 2004]. Four different microstructures have been subjected to the mechanical loading, immediately after casting (as-cast), indicated as microstructure A and after 2, 6, and 15 hours of ageing, respectively indicated as microstructure B, C, and D. They are shown in Fig. 3.4. The dark regions correspond to the α or Pb-rich phase while the light regions correspond to the β or Sn-rich phase.

Figure 3.4 — Simulated microstructure of aged eutectic Sn–Pb aged at 150°C.
The specimens are subjected to cyclic shearing to 2% global deformation after which the direction of shearing is reversed until the 0% shear state is recovered. After three cycles the specimens are held to evaluate the relaxation behaviour. For all the microstructures a strain rate $\dot{\varepsilon}$ of $2 \cdot 10^{-3} \text{s}^{-1}$ is used. The calculations have been performed using periodic boundary conditions: The deformation of each opposing boundary pair is equal and the stress vectors are opposite in sign on each pair. The appropriate dependencies for the edges, see Fig. 3.5, are:

$$\vec{x}_R - \vec{x}_{BR} = \vec{x}_L - \vec{x}_{BL}, \quad \text{and} \quad \vec{x}_T - \vec{x}_{TL} = \vec{x}_B - \vec{x}_{BL},$$  

(3.60)

where the subscripts $L$, $R$, $T$, and $B$ denote left, right, top, and bottom respectively. A regular mesh of 50 by 50 quadratic elements has been used. In this way each interface is described by approximately 3 to 4 quadratic elements.

![Figure 3.5 — Periodic mesh and boundary conditions.](image)

**Figure 3.5** — Periodic mesh and boundary conditions.

![Figure 3.6 — Reaction force versus time for different microstructures, strain rate $2 \cdot 10^{-3} \text{s}^{-1}$. Force in horizontal direction along edge $\vec{x}_{TL} - \vec{x}_{TR}$ (see Fig. 3.5).](image)

**Figure 3.6** — Reaction force versus time for different microstructures, strain rate $2 \cdot 10^{-3} \text{s}^{-1}$. Force in horizontal direction along edge $\vec{x}_{TL} - \vec{x}_{TR}$ (see Fig. 3.5).
Fig. 3.6 shows the global reaction force as a function of time for the different microstructures. At first the maximum reaction force increases with each cycle due to hardening of the material, but after a small number of cycles a steady state is reached. This is more clearly depicted in Fig. 3.7 which shows the reaction force versus displacement curve for 12 shearing cycles performed on microstructure D with \( \dot{\varepsilon} = 2 \cdot 10^{-3} \, \text{s}^{-1} \). A dependency on the microstructure can be seen in Fig. 3.6. Ageing of the material, which leads to an increase of microstructural length scales, results in a lower yield point and lower reaction forces during loading. Microstructure A clearly shows the highest overall reaction force, Microstructures B and C clearly show a consecutive reduction of the reaction forces. However, between microstructures C and D the difference is very small. This can be explained by the fact that relatively little coarsening has occurred in this last stage of the ageing process resulting in similar microstructures.

The tendency ‘small is strong’ is also known to occur in single phase materials, which show a increasing strength as the grain size decreases. The best-known macroscopic experimental consequence is the increased flow stress on decreasing average grain size, which is expressed by the so-called Hall–Petch relation [Evers et al., 2004]. This strengthening effect is commonly attributed to stress concentrations originating from material heterogeneity, which obstruct plastic flow. Therefore, next, the strain and stress distributions within the microstructures will be investigated to find the cause behind the strengthening effect found here for the eutectic Sn–Pb system.

The development of the equivalent Cauchy stress, \( \sigma_{eq} \) for microstructure A, loaded with strain rate \( \dot{\varepsilon} = 2 \cdot 10^{-3} \, \text{s}^{-1} \), is shown in Fig. 3.8. It can be clearly seen that stress concentrations develop in the hard \( \beta \)-phase. Since the yield stress is much lower in the \( \alpha \)-phase it will start to flow plastically before the yield stress in the rest of the material has been reached. The hardening induced by the plastic flow is not significant enough to smooth the stress field. This can be observed in Fig. 3.10 which shows the development of the effective viscoplastic strain, \( \varepsilon_{vp} \). The plastic strain is almost completely confined to the \( \alpha \)-phase. It should be noted that due to the localisation of plastic straining the effective viscoplastic strain reaches values of 0.22 already after 3 cycles, which is much higher than the applied macro shear strain of 0.02. The heterogeneous plastic straining of the material results in residual stresses after relaxation, see Fig. 3.9. All the other simulations show the
same qualitative behaviour. Therefore the $\varepsilon_{\text{vp}}$ is only shown at the end of the simulation, Fig. 3.11, and the $\sigma_{eq}$ at $t = 60$ and $70$ s, that is at maximum stress levels and after relaxation, Figs. 3.12 and 3.13.

As opposed to the reaction force, the stress concentrations increase with increasing microstructural sizes. This is due to the fact that the finer microstructure allows for a better distribution of the stresses, whereas the coarser microstructure forces the stresses to localise more. The residual stresses after relaxation do not show a significant difference between the various microstructures. The effect of increasing microstructural length scale can be seen more clearly by comparing the equivalent viscoplastic strain fields, see Fig. 3.11. Increased ageing, or coarsening, leads to increased localisation behaviour and thus higher (local) values for the viscoplastic strain. Note that the maximum viscoplastic strain is in fact higher for microstructure C than for microstructure D. This is due to the higher curvature at the most prominent localisation site for the two microstructures, while the difference in the sizes of the microstructures is not pronounced.

The reason behind the strengthening effect can be seen in Fig.3.11. For the initial microstructure, with the smallest phase region size, the effective viscoplastic strain distributed relatively evenly over the material. This indicates that the deformation needed to accommodate the prescribed displacement occurred in both phases. However, as the microstructure coarsens more of the viscoplastic strain is confined to the softer Pb-rich

![Figure 3.8 — Evolution of the equivalent Cauchy stress field in MPa for microstructure A, $\dot{\varepsilon} = 2 \cdot 10^{-3}$ s$^{-1}$.](image1)

![Figure 3.9 — Residual equivalent Cauchy stress field in MPa for microstructure A after cycling, $\dot{\varepsilon} = 2 \cdot 10^{-3}$ s$^{-1}$, $t = 70$ s.](image2)
phase. Fig. 3.11 shows almost no viscoplastic deformation occurred in the Sn-rich phase in microstructure D. This means that only the soft phase has to undergo deformation which leads to a lower required force for this case compared to a fine microstructure.

To demonstrate the influence of the strain rate microstructure A is also loaded with rates of respectively $2 \cdot 10^{-4}$ and $2 \cdot 10^{-2}$ s$^{-1}$. Fig. 3.14 shows the reaction forces versus time for the three strain rates. Increasing the rate also increases the flow stress and reaction force. The stress and effective viscoplastic strain fields show similar behaviour. The localisation occurs in the same positions for all strain rates, but increases in intensity. Note that the increase of $\varepsilon_{vp}$ with increasing strain rate also results in higher residual stresses.

Figure 3.10 — Evolution of the effective viscoplastic strain field [-] for microstructure A, $\dot{e} = 2 \cdot 10^{-3}$ s$^{-1}$.

Figure 3.11 — Equivalent viscoplastic strain [-] at 70 s for the different microstructures, $dotc = 2 \cdot 10^{-3}$ s$^{-1}$. 
Figure 3.12 — Equivalent Cauchy stress in MPa at 60 s for different microstructures, $\dot{\varepsilon} = 2 \cdot 10^{-3} \text{ s}^{-1}$.

Figure 3.13 — Equivalent Cauchy stress in MPa at 70 s for different microstructures, $\dot{\varepsilon} = 2 \cdot 10^{-3} \text{ s}^{-1}$.
Figure 3.14 — Reaction force versus time for various strain rates $[\text{s}^{-1}]$ (microstructure A).

Figure 3.15 — Equivalent viscoplastic strain $[-]$ at 70 s for various strain rates (microstructure A).

Figure 3.16 — Equivalent Cauchy stress in MPa at $t = 60$ for different strain rates (microstructure A).
A model is presented to describe the mechanical behaviour of microstructure dependent materials. The model describes the microstructure using a phase field approach and employs a constitutive model dependent on the phase field to account for the underlying microstructure. It has been demonstrated by applying it to eutectic Sn–Pb solder, a material whose mechanical characteristics are strongly influenced by its continuously evolving microstructure. For the constitutive behaviour the elasto-viscoplastic Perzyna model has been adopted. The Perzyna model allows the use of different hardening functions, which makes it suitable for a large variety of materials. It is coupled to the phase field model through the model parameters which are taken dependent on the mass fraction field resulting from the solution of the phase field equations. In this way the microstructure is accounted for using a continuum mechanics approach. The approach does not require interface tracking methods since all information about the interface is included in the phase field. Furthermore, the evolution of a microstructure can be calculated using one mesh, no re-meshing needs to be done.

Two-dimensional simulations of cyclic shearing have been performed using various microstructures. These microstructures were obtained by performing a simulation of static ageing of eutectic Sn–Pb at 150°C with the phase field model. The mechanical simulations demonstrate a dependency of the material behaviour on the microstructure. Microstructures that have been aged for longer periods of time not only show larger microstructural length scales, but also a decrease in resulting reaction force, indicating microstructural softening of the material.

The difference between the various microstructures is even more pronounced when the stress field and the effective viscoplastic strain rate are examined locally. As the microstructural length scales increase, more pronounced localisation patterns are found. Both $\sigma_{eq}$ and $\varepsilon_{vp}$ locally reach significantly higher values for aged microstructures. Because of the difference between the yield stress of the two phases present in the material, most plastic flow occurs in the softer $\alpha$-phase. Locally the plastic strain can reach values much higher than the macroscopically applied strain. Due to this heterogeneous plastic straining residual stresses appear in the material after relaxation.

Frear et al. [1988] found that regions of high strain in the eutectic Sn–Pb show a higher degree of coarsening. Furthermore, it is found that these regions of inhomogeneous
Coarsening are the crack nucleation sites during thermal cycling [Frear, 1989; Hacke et al., 1998]. The cracks occur on the interface between the Sn-rich and the Pb-rich phase [Frear, 1989; Zhao et al., 2000]. The results presented in this paper demonstrate that a coarser microstructure leads to significantly higher local values of the viscoplastic strain and of the stresses. Future work will be focused on the incorporation of damage into the presented model.
A theoretical framework describing the nonlinear mechanical behaviour of multiphase solders is introduced, along with its numerical implementation. Next to viscoplasticity, damage has been included in the model using a gradient-enhanced nonlocal damage description. Microstructural data is accounted for in the model by means of a phase field. In here, the methodology followed is generic and can be readily extended to numerous multi-phase materials. The model is applied to the eutectic Sn–Pb system whose mechanical properties are known to be heavily dependent on the underlying microstructure. The results indicate that coarser microstructures exhibit a more pronounced localisation behaviour compared to finer ones leading to higher local values for the stresses and viscoplastic strains. Furthermore, different damage evolution behaviour is observed for the investigated microstructures, with a coarser microstructure leading to earlier microstructural failure.

4.1 Introduction

The ongoing miniaturisation in microelectronics is at present confronted with a number of technological issues affecting the reliability of the soldered connections. Increasing functionality and chip density on printed circuit boards, leads to higher energy fluxes and thus higher temperatures. Since soldered connections typically have low melting temperatures, they are operating at very high homologous temperatures and consequently exhibit high temperature deformation mechanisms associated with creep and relaxation. Furthermore, their size has been decreased to the extent that microstructural length scales are of the same order of magnitude as their geometrical dimensions. The microstructure plays an increasingly important role when it comes to the mechanical behaviour. Consequently, a solder joint will behave intrinsically differently from the corresponding bulk solder material [Bonda and Noyan, 1992]. Furthermore, the microstructure will change over time: the high homologous temperature at which solder materials operate provokes microstructural coarsening, resulting in a markedly different microstructure after only a short time following solidification [Conrad et al., 1999; Jung and Conrad, 2001a,b; Matin et al., 2003]. The high temperatures also provoke diffusion of external material into the joint, changing the average composition of the solder or triggering the growth of an intermetallic compound layer [Amagai et al., 2002; Miao and Duh, 2001; Stam and Davitt, 2001]. All this affects the properties of the solder joint, which consequently vary throughout its lifetime. The importance of the microstructure with respect to one of the most crucial technological properties of soldered connections, i.e. their reliability, has been recognised by various authors [Frear et al., 1997; Basaran and Chanderoy, 1998; Vianco et al., 1999; Basaran and Chanderoy, 2000].

The Sn–Pb (Tin–Lead) system has been the most widely used solder material for a long time. Although legislation in Europe is moving toward lead–free electrical and electronic equipment in 2006, exemptions will be made for applications where substitution is not possible from a scientific and technical point of view or where the negative environmental or health impacts caused by substitution are likely to outweigh the human and environmental benefits of the substitution [EU, 2003; Goodman et al., 2004]. Critical applications of electronics with possible safety constraints like e.g. in aerospace, are typical examples of expected exemptions. Since Sn–Pb is the most studied and best known solder alloy, it is used here as a reference material.

In this paper a framework will be introduced to investigate the influence of the microstructure on the mechanical response of multiphase materials. The resulting modelling approach will be demonstrated on the mechanical cycling of eutectic Sn–Pb solder.

The Sn–Pb system exhibits severe coarsening of phase regions under the influence of temperature. The microstructure and the mechanical response of Sn–Pb are known to be closely linked [Basaran and Jiang, 2002], especially when it comes to fatigue damage [Frear et al., 1988, 1997; Hacke et al., 1998; Kanchanomai et al., 2002a]. In the literature various publications can be found on the modelling of Sn–Pb solder where a single scalar quantity is incorporate in the model to account for the microstructure [Conrad et al., 1999; Guo and Conrad, 1993; Basaran and Chanderoy, 2000; Basaran and Tang, 2002; Hacke et al., 1998]. In the proposed framework, more extensive information on the microstructure is included. This is accomplished by coupling a phase field model [Cahn and Hilliard, 1958; Leo et al., 1998; Zhu et al., 1999; Dreyer and Müller, 2000; Ni et al., 2002] describing
the microstructural evolution to the parameters figuring in the constitutive equation. The phase field approach yields detailed information on the microstructure, such as the shape, size, distribution, and composition of the phase regions. The results of a static ageing simulation were used as input for mechanical analyses of the influence of the microstructure on the macroscopic response of eutectic Sn–Pb. A brief outline of the adopted phase field model is given in section 4.2; more extensive information can be found in Ubachs et al. [2004].

Because of their high homologous temperature, soldered connections are prone to high temperature deformation mechanisms. Therefore, the mechanical response of solder alloys is usually characterised by a viscoplastic material model. Various time-dependent models are available and have been employed to describe the material behaviour of solder, e.g. the Anand model [Anand, 1985; Amagai, 1999; Cheng et al., 2000; Wang et al., 2001] and the Dorn equation [Guo and Conrad, 1993; Akay et al., 1997; Zhang and Lee, 1998; Anderson et al., 2000]. Here, the viscoplastic strain rate function defined by Perzyna [1966] is selected to describe the mechanical response [Wang et al., 1997; Van der Sluis et al., 1999; Ju et al., 1994; Basaran et al., 1998]. Through the coupling with the results of the phase field model this results in a model that shows a mechanical response influenced by the microstructure. By taking the material parameters of the viscoplastic model dependent on the mass fraction field, available in the phase field model, the required microstructural information is accounted for.

In section 4.4 the model is extended to include damage initiation and propagation. Attention is given to the regularising properties of the softening model used. It is known that rate dependency (viscosity) may restore the well posedness of the initial value problem [Wang et al., 1997]. Viscosity regularises the solution by implicitly introducing a length scale into the model, which causes the localisation region to be more diffuse compared to an inviscid plastic material [Glema et al., 2000]. These viscous terms are sometimes referred to as temporal nonlocalities [Sluys and De Borst, 1994]. However, for practical purposes this effect is usually only sufficient for highly rate-sensitive materials or high loading rates and the numerical results can still show a mesh dependency [Wang and Sluys, 2000; Voyiadjis et al., 2004; Peerlings et al., 2004]. Therefore, a nonlocal approach will be used here for the incorporation of of damage; the damage is driven by a nonlocal internal variable which is the weighted average of its local counterpart over a finite domain [Steinmann, 1999; Geers et al., 2001, 2003; Boers et al., 2005].

In the literature, solder is usually considered as a single phase material material when modelling damage [Basaran and Chanderoy, 2000; Basaran and Tang, 2002; Zhang and Lee, 1998; Towashiraporn et al., 2004; Yang and Nassar, 2005]. The damage parameter is in most of these cases only influenced by the microstructure through the inclusion of an averaged grain size parameter in the constitutive model. Here, the damage evolution parameters will be coupled with the local composition of the material obtained with the phase field model. In this way damage behaviour can be specified individually for the two phases and for interfaces.

Some aspects of the numerical implementation are discussed in section 4.5 after which the choice of material parameters is dealt with in section 4.6. In section 4.7, numerical simulations of a mechanically loaded Sn–Pb solder alloy with different microstructure and strain rates are discussed, after which the conclusions will be presented in section 4.9.
4.2 Phase field model

In order to describe the microstructure and its evolution, a phase field model is used. The mass fraction \( c \) is taken as the field variable, and it is assumed that it varies continuously throughout the system. The evolution of the mass fraction field is obtained by solving the non-linear diffusion equation:

\[
\rho \frac{dc}{dt} = \nabla \cdot (\rho \mathbf{M} \cdot \nabla \delta \Phi \delta c), \tag{4.1}
\]

where \( \rho \) is the mass density, \( t \) is time, \( \mathbf{M} \) the mobility tensor and \( \Phi \) the free energy. The partial derivative of the free energy with respect to the mass fraction is the chemical potential, which is the driving force of the diffusion process since it determines how the microstructure will evolve. The phase field model will be utilised here to calculate the evolution of a microstructure during a static ageing process, without any external mechanical loading.

The free energy is assumed to consist of three parts: the configurational energy, \( \Phi_0 \), which determines the equilibrium mass fractions for the different phases, the interfacial energy, \( \Phi_\gamma \), which controls the number of interfaces, their size, and orientation, and the elastically stored energy \( \Phi_e \). However, since no mechanical boundary conditions are applied during the ageing process, the influence of the elastic part of the free energy is negligible. Therefore the elastic free energy term is omitted in the following.

The free energy function, which is the starting point for finding an expression for the mass flux is taken from Giacomin and Lebowitz [1997]. They derived a macroscopic equation describing phase segregation in microscopic model systems with long-range interactions evolving according to stochastic Kawasaki dynamics with nearest neighbour exchanges (see also Brandon [1994]). They obtained the following expression for the specific free energy:

\[
\Phi = \Phi_0(c(\vec{x})) + \frac{1}{4} \int_\Omega g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right]^2 d\vec{y}, \tag{4.2}
\]

where \( \Omega \) is the region around the material point with position vector \( \vec{x} \), \( \vec{y} \) a local coordinate vector with respect to \( \vec{x} \) pointing to neighbouring material points in \( \Omega \). \( g \) is the nonlocal kernel (a symmetric smooth function \( \mathcal{C}^\infty \)), and \( \Phi_0 \) is the equilibrium free energy density of a homogeneous system. Similar nonlocal extensions of the free energy have been used by Gajewski and Zacharias [2003] and Fosdick and Mason [1998]. The second term on the right hand side equals zero in the bulk of individual phase regions, since there \( c(\vec{x}) \) is equal to \( c(\vec{x} + \vec{y}) \). Only in the neighbourhood of interfaces will these two variables differ from each other. This term thus represents the interfacial free energy, \( \Phi_\gamma \). Where \( \Phi_0 \) causes a system to phase separate, \( \Phi_\gamma \) penalises it for the amount of interface area present, and so drives the coarsening process.

In this work, the introduced nonlocality which accounts for the finite size of the interfacial effects is assumed to be Lagrangian (material) in nature, i.e. the volume over which long range interactions occur is defined by the material configuration. Using the specific free energy (4.2) and taking the variation with respect to \( c(\vec{x}) \) in order to obtain the chemical potential \( \mu \) then gives,

\[
\mu = \frac{\delta \Phi}{\delta c(\vec{x})} = \frac{\delta \Phi_0}{\delta c} + \frac{1}{2} \int_{\Omega_0} g(\vec{y}) \left[ c(\vec{x}) - c(\vec{x} + \vec{y}) \right] d\vec{y}, \tag{4.3}
\]
which can be rewritten to yield:
\[ \mu = \frac{\partial \Phi_0}{\partial c} + \kappa [c(\bar{x}) - \bar{c}(\bar{x})], \] (4.4)

where \( \kappa \) is the interfacial tension coefficient. In here, the nonlocal mass fraction \( \bar{c} \) is defined as
\[ \bar{c}(\bar{x}) = \int_{\Omega_0} g'(\bar{y}) c(\bar{x} + \bar{y}) \, d\bar{y}, \quad \kappa = \frac{1}{2} \int_{\Omega_0} g(\bar{y}) d\bar{y}, \quad \text{and} \quad g' = g / \kappa. \] (4.5)

Eq.(4.5) can be reformulated towards an implicit Helmholtz equation [Ubachs et al., 2004] which is well suited for implementation in a finite element framework:
\[ \bar{c} - \ell_c^2 \nabla_0^2 \bar{c} = c, \] (4.6)

where \( \nabla_0^2 \) represents the Laplacian operator taken in the material (Lagrangian) configuration, and \( \ell_c \) is an internal length parameter, which, when using an isotropic weighting function, \( g(r) \), depending on a radial coordinate \( r \) only, is defined as
\[ \ell_c^2 = \frac{1}{2 \Omega_0} \int_{\Omega_0} r^2 g(r) \, dr. \] (4.7)

Note that the solution of this partial differential equation indeed leads to a strongly nonlocal solution as assumed in (4.5). Details on this particular topic have been elaborated within the context of strongly nonlocal damage [Peerlings et al., 2001]. Details on the explicit format of the nonlocal kernel (which is the result of the adopted mathematical approximation) can be found in this reference as well.

The reformulation of the integral expression (4.5) towards the differential equation (4.6) requires the introduction of boundary conditions. Either the value of \( \bar{c} \), its normal derivative, or a linear combination of these quantities must be specified on the boundary. Here the homogeneous Neumann boundary condition has been used on the entire boundary:
\[ \bar{V}_0 \bar{c} \cdot \bar{n} = 0, \] (4.8)

where \( \bar{n} \) is the normal vector of the boundary. This condition ensures the conservation of the global mass fraction throughout the entire system upon nonlocal averaging:
\[ c_{\text{total}} = \frac{1}{\Omega_0} \int_{\Omega_0} c \, d\bar{x} = \frac{1}{\Omega_0} \int_{\Omega_0} \bar{c} \, d\bar{x}. \] (4.9)

More comments on this boundary condition can be found in Peerlings et al. [2001]. The complete nonlocal phase field model now reads
\[ \rho \frac{dc}{dt} = \bar{V} \cdot (\rho \bar{M} \cdot \bar{V} \mu) \quad \text{with} \quad \mu = \frac{\partial \Phi_0}{\partial c} + \kappa [c - \bar{c}], \] (4.10a)
\[ \bar{c} - \ell_c^2 \nabla_0^2 \bar{c} = c. \] (4.10b)

The solution of the system of equations yields detailed information on the distribution of phases in the material, the position and orientation of interfaces, and the mass fractions in the separate phases. The mechanical behaviour of the material is dependent on its microstructure. More detailed information on this specific phase field model, its implementation and its application can be found in Ubachs et al. [2003, 2004].
4.3 Elasto-viscoplasticity model

4.3.1 Kinematics

Transformation from the undeformed configuration at time $t_0$ to the current configuration at time $t$ is described by the deformation gradient tensor $F$, of which the determinant $J = \det(F)$ represents the relative volume change. The conventional multiplicative decomposition is used to split the deformation into an elastic and inelastic (viscoplastic) part, denoted by subscripts $e$ and $vp$ respectively:

$$F = (\nabla_0 \tilde{x})^c = F_e \cdot F_{vp}. \quad (4.11)$$

The right and left Cauchy–Green strain tensors, $C$ and $B$, and the Green-Lagrange strain tensor $E$ are given by

$$C = F^c \cdot F, \quad B = F \cdot F^c, \quad \text{and} \quad E = \frac{1}{2} (C - I). \quad (4.12)$$

Material velocity is taken into account by the rate of deformation $D$ and the spin tensor $\Omega$, respectively being the symmetric and skewsymmetric parts of the velocity gradient tensor $L$. Decomposition (4.11) implies the introduction of elastic and viscoplastic rate tensors:

$$L = \left(\nabla \tilde{\nu}\right)^c = F \cdot F^{-1} = F_e \cdot F^{-1}_e + F_e \cdot F_{vp} \cdot F^{-1}_{vp} \cdot F^{-1}_e$$

$$= L_e + L_{vp} = (D_e + \Omega_e) + (D_{vp} + \Omega_{vp}). \quad (4.13)$$

To make the decomposition unique it is frequently assumed that the viscoplastic spin $\Omega_{vp}$ is zero. Superimposed rigid body rotations are thus fully represented in $F_e$.

4.3.2 Elasto-viscoplastic model

A hyperelastic model is chosen to describe the elastic part of the material behaviour at finite deformations. In that case, an elastic strain energy function $\Phi_e$ exists, which can be used to calculate the stresses,

$$S = \frac{1}{J} \sigma = \frac{1}{J} F^{-1} \cdot \sigma \cdot F^{-c} = \frac{1}{J} F^{-1} \cdot \tau \cdot F^{-c} = \frac{\partial \Phi_e}{\partial E} = 2 \frac{\partial \Phi_e}{\partial C}. \quad (4.14)$$

Here $S$, $\sigma$, and $\tau$ denote the second Piola-Kirchhoff, the Cauchy, and the Kirchhoff stress tensor, respectively. Since the constitutive relation is needed in a rate form, the Truesdell objective time derivative of the Kirchhoff stress $\nabla \tau$ is used, which is constitutively related to the rate of deformation tensor by

$$\nabla \tau = \mathbb{H} : D, \quad (4.15)$$

The elastic strain energy function is given by

$$\Phi_e = \frac{1}{2} G [C : I - 3 - 2 \ln(J)] + \frac{1}{2} K \ln^2(J), \quad (4.16)$$

where $G$ and $K$ are Lamé’s constants defined as

$$G = \frac{E}{2(1 + v)} \quad \text{and} \quad K = \frac{\nu E}{(1 + v)(1 - 2v)} , \quad (4.17)$$
with $E$ the Young's modulus and $\nu$ the Poisson's ratio. This leads to the following expression for the fourth-order material tensor $\mathbb{H}$,

$$\mathbb{H} = 2 \left[ G - K \ln(J) \right] \mathbb{I} + K \mathbb{I} \mathbb{I}.$$  \hfill (4.18)

The material is assumed to behave viscoplastically once a threshold is exceeded, i.e. after an equivalent stress measure exceeds the current yield stress $\tau_y$. Here the Von Mises equivalent stress is used, which expressed in terms of the Kirchhoff stress tensor $\tau$ reads

$$\tau_{eq} = \sqrt{\frac{3}{2} \tau^d : \tau^d}, \quad \text{with} \quad \tau^d = \tau - \frac{1}{3} \tau : \mathbb{I}.$$ \hfill (4.19)

The evaluation of the occurrence of yield is then evaluated through a yield criterion $\varphi$, which represents a yield surface in stress space.

The classical Von Mises-Huber yield function $\varphi$ is used to evaluate the stress state and to check whether the deformation is purely elastic ($\varphi < 0$) or viscoplastic ($\varphi \geq 0$):

$$\varphi(\tau, \varepsilon_{vp}, \dot{\varepsilon}_{vp}) = \tau_{eq} - \tau_y(\tau_{y0}, \varepsilon_{vp}, \dot{\varepsilon}_{vp}).$$ \hfill (4.20)

In here, $\tau_y$ may incorporate a linear, a non-linear hardening, or a softening response. The initial yield stress is denoted by $\tau_{y0}$ while the hardening behaviour is related to the effective viscoplastic strain measure $\varepsilon_{vp}$ and its rate $\dot{\varepsilon}_{vp}$. In contrast with conventional rate-independent plasticity theory, stress states outside the yield surface are allowed, hence the name overstress model which is sometimes used for these types of models.

During viscoplastic deformation the yield stress will change as a function of the effective viscoplastic strain $\varepsilon_{vp}$ and its rate $\dot{\varepsilon}_{vp}$. Plastic flow does not induce any stresses in the material, therefore the constitutive model becomes

$$\nabla \tau = \mathbb{H} : D_e = \mathbb{H} : (D - D_{vp}),$$ \hfill (4.21)

With ongoing viscoplastic deformation the direction of the viscoplastic strain rate $D_{vp}$ is defined by the normality or associative flow rule which states that the strain rate is directed along the normal to the yield surface and denoted by $\partial \varphi / \partial \tau$. Its magnitude is characterised by the rate of the viscoplastic multiplier $\dot{\lambda}$. Thus the expression for the viscoplastic strain rate can be formulated as [Perzyna, 1985]:

$$D_{vp} = \dot{\lambda} \mathbb{N}, \quad \text{where} \quad \mathbb{N} = \frac{\partial \varphi}{\partial \tau} = \frac{3}{2} \frac{\tau^d}{\tau_{eq}}, \quad \text{and} \quad \dot{\lambda} = \eta \varphi(\varphi),$$ \hfill (4.22)

with $\eta$ a fluidity parameter which determines the magnitude of the viscoplastic strain rate and $\varphi(\varphi)$ the overstress function.

A power law is chosen for the overstress function [Perzyna, 1971]:

$$\varphi(\varphi) = \left( \frac{\varphi}{\varphi_0} \right)^N,$$ \hfill (4.23)

with $N$ the rate sensitivity parameter and $\varphi_0$ is a scaling parameter. A higher value of $N$ reduces the strain rate sensitivity of the model.
The rate of the hardening variable $\dot{\varepsilon}_{vp}$ is directly coupled to the rate of the viscoplastic multiplier $\dot{\lambda}$ by $\dot{\varepsilon}_{vp} = \dot{\lambda}$. The definition of $\dot{\varepsilon}_{vp}$ then follows as

$$\dot{\varepsilon}_{vp} = \dot{\lambda} = \sqrt{\frac{2}{3} D_{vp} : D_{vp}}. \quad (4.24)$$

More details on this particular viscoplastic model and its application can be found in Ubachs et al. [2005a].

### 4.4 Elasto-viscoplastic damage

To account for the effect of damage, an isotropic damage parameter $\omega$ is introduced. This damage parameter can be interpreted as the reduction of effective volume of the material due to void initialisation, growth, and coalescence. For the undamaged material $\omega$ is zero, and for completely failed material $\omega$ equals one. A linear relation is assumed between the volume reduction and the elastically stored free energy. In the definition for the stress, Eq. (4.14), $\Phi_e$ is then replaced by $(1 - \omega)\Phi_e$. Differentiating with respect to time, and transforming the result to the current configuration, similar to the procedure in section 4.3.2 yields for the rate of stress

$$\nabla \tau = (1 - \omega) H : (D - D_{vp}) - \frac{\tau}{(1 - \omega)} \dot{\omega}, \quad \text{with} \quad D_{vp} = \dot{\lambda} N, \quad (4.25)$$

where the second term originates from time differentiation of the damage term. As damage increases within a material volume it gradually looses its ability to store energy elastically but also to dissipate it plastically. The damage parameter is therefore also present in the viscoplastic part of the model,

$$\dot{\lambda} = \eta \left( \frac{(1 - \omega)^{-1} \tau_{eq} - \tau_y}{\phi_0} \right)^N = \eta \left( \frac{\tau_{eq} - (1 - \omega) \tau_y}{(1 - \omega) \phi_0} \right)^N. \quad (4.26)$$

The term $(1 - \omega)^{-1} \tau_{eq}$ can be considered as the effective stress, i.e. the actual stress that has to be carried by the undamaged material after correcting for the loss of cross sectional area due to damage.

#### 4.4.1 Gradient enhanced damage

To restore the well-posedness of the governing partial differential equations and avoid problems with mesh dependency coupled to that, the damage parameter is related to a nonlocal variable $\bar{z}$ which is defined as

$$\bar{z}(\vec{x}) = \int_{\Omega_0} G(\vec{y}) z(\vec{x} + \vec{y}) \, d\vec{y} \quad (4.27)$$

where $z$ is a local quantity which acts as the damage driving variable, $G$ a weighting function, $\Omega_0$ is the region around the material point with position vector $\vec{x}$, and $\vec{y}$ a local position vector defined relative to $\vec{x}$. The damage parameter $\omega$ is computed with a damage evolution law from a history parameter $Z$,

$$Z = \{ \max[\bar{z}(t')] \mid t' \in [0, t] \} \quad (4.28)$$
The history parameter represents the largest value of $\bar{z}$ attained during the complete deformation history of a material point, so $\omega = \omega(Z)$. With respect to the history parameter the following set of Kuhn-Tucker conditions holds

$$\dot{Z} \geq 0, \quad \bar{z} - Z \leq 0, \quad \dot{Z}(\bar{z} - Z) = 0, \quad \text{with initial condition } Z_0 = Z_i$$  \hspace{1cm} (4.29)

Here $Z_i$ is the damage initialisation threshold, below this value no damage occurs.

Eq.(4.27) can be rewritten in a differential form [Peerlings et al., 1996; Geers et al., 2003]:

$$\bar{z} - \ell_\omega^2 \nabla_0^2 \bar{z} = z,$$  \hspace{1cm} (4.30)

where the subscript $o$ indicates the material (initial undeformed) configuration, i.e. the Laplacian is taken with respect to the initial configuration. Usually, the Neumann boundary condition $\nabla_0 \bar{z} \cdot \vec{n}$ is applied. A partial justification for this boundary condition is given in Peerlings et al. [2001]. By deriving the solution of Eq.(4.30) using Green's functions, Peerlings et al. [2001] have shown in the context of quasi-brittle damage that such an approach is strictly nonlocal in the sense of Bažant and Pijaudier-Cabot [1988].

This particular damage model has recently been applied successfully in the modelling of metal forming processes [Boers et al., 2005; Mediavilla et al., 2005a,b,c]

The parameter $\ell_\omega$ is a material length parameter accounting for microstructural and micromechanical events. Because it is defined in the material configuration, averaging is always performed over the same material volume when $\ell_\omega$ is taken constant. It is also possible to write Eq.(4.30) in the spatial (current deformed) configuration. In this case, when taking the length parameter constant, averaging is done over the same spatial volume. More information on this subject can be found in Geers et al. [2003] were it is shown that a material nonlocal formulation always leads to an equivalent spatial nonlocal formulation where the nonlocal kernel depends on the deformation. It appears that this spatial length scale evolves monotonically with the deformation. See also Steinmann [1999] who derives and investigates four alternative formulations for gradient enhanced damage in a geometrically nonlinear framework.

**Damage driving variable**

For the damage driving variable various potential choices can be made a priori, e.g. the plastic strain, a combination of stress and strain, the stored or dissipated energy, or the entropy [Basaran and Tang, 2002]. However, every material has characteristic damage mechanisms, which may vary under different loading conditions. No direct experimental information is available related to the damage process of the Sn–Pb alloy that permits to qualify the correct damage driving variable. Therefore, the effective viscoplastic work $W_{vp}$ per unit of reference volume is chosen as the driving variable,

$$z = W_{vp} = \int_0^{t+\Delta t} \left[ \frac{\sigma : D_{vp}}{1 - \omega} \right] dt' = \int_0^{t+\Delta t} \frac{\tau_{eq} \dot{\lambda}}{1 - \omega} dt'.$$  \hspace{1cm} (4.31)

The consideration leading to this choice of damage driving variable is as follows. Because of the varying properties of the two phases in the material under investigation, the stresses and the viscoplastic strains occurring in the individual phases differ significantly and cannot be readily compared. Taking the plastic work as the damage driving variable
implicitly introduces a distinct damage behaviour for different strain rates. Because plastic work depends both on stress and strain, the damage process will be affected by the strain rate. For increased rates, damage will occur at lower strains, and subsequently propagate faster. Note that hydrostatic stresses, which are known to play an important role in ductile damage initiation for metals [Oyane et al., 1980; Rice and Tracey, 1969], and for void nucleation and growth in general [Gurson, 1977; Tvergaard, 1990; Needleman et al., 1992], do not influence the damage process for this particular choice.

**Damage evolution law**

Reflecting an initial rapid damage growth, the relation between the damage variable $\omega$ and its driving parameter is described by the following exponential relation,

$$
\omega = 1 - \exp\left[-b(Z - Z_i)\right]
$$

(4.32)

Here $b$ is the rate at which damage develops. The exponential form tends to values of $\omega$ close yet smaller than unity, preventing the stiffness matrix from becoming singular.

### 4.5 Finite element implementation

The complete elasto-viscoplastic damage model in a rate form now consists of the constitutive equation Eq.(4.25), the expressions for the rate of the magnitude of viscoplastic flow Eq.(4.26) and the rate of the damage driving parameter Eq.(4.30), the nonlocal averaging equation (4.30), and the damage evolution law Eq.4.32. Whether the behaviour will be elastic or elasto-viscoplastic is determined by evaluation of the yield function, Eq.(4.20), and whether damage increases is evaluated by use of the Kuhn-Tucker conditions given in Eq.(4.29).

The system of equations will be solved within a finite element framework. Displacements and nonlocal variables are taken as degrees of freedom and hence solved in the global iteration loop. Therefore, the following two equations have to be solved, the momentum balance and the nonlocality equation,

$$
\bar{\nabla} \cdot \sigma = \bar{0},
$$

(4.33a)

$$
\bar{z} - \ell^2 \bar{\nabla}_o^2 \bar{z} = \bar{z}.
$$

(4.33b)

Implementing this coupled problem in a finite element program for finite deformations follows standard routines: time discretisation of the equations, elaboration of the weak form of the governing equations, linearisation and discretisation. The system is then solved in an iterative manner.

After each global iteration, the new residual stresses are calculated. During this step the viscoplastic multiplier increments $\Delta \lambda$ are also updated if necessary. For each integration point, first the increment is assumed to be entirely elastic. The stresses obtained in this elastic predictor state are then used to check the yield criterion. If this is violated, the return mapping is performed, i.e. the amount and direction of plastic flow is determined and the stresses are updated accordingly. The return mapping solves the set of non-linear equations, Eqs. (4.25) and (4.26) through an iterative procedure. For this iterative procedure
at the integration point level, a combination of the bisection method and the secant method has been used, as the standard Newton–Raphson scheme does not converge adequately. Accurately solving the stress update algorithm is essential for achieving the correct solution of the system.

The global iteration loop is solved using a Newton–Raphson iteration scheme. For the speed and stability of this method, the proper use of a consistent tangent is of great importance, ensuring quadratic convergence. In this section a brief summary of the finite element implementation is given, details are provided in appendices C.2 and C.3.

### 4.5.1 Incremental elasto-viscoplastic nonlocal damage model

Since the system of equations cannot be integrated analytically, an incremental time integration scheme will be used. Performing a backward Euler discretisation scheme, 

\[
a = a^{t+\Delta t} = a^t + \frac{da}{dt}\big|_{t+\Delta t} \Delta t,
\]

yields for the viscoplastic multiplier

\[
\Delta \lambda = \lambda - \lambda^t = \Delta t \eta \left( \frac{\tau_{eq} - (1 - \omega) \tau_Y}{(1 - \omega) \phi_0} \right)^N,
\]

and for the damage driving parameter

\[
z = z^t + \frac{\tau_{eq} \Delta \lambda}{1 - \omega}.
\]

The constitutive equation for the stress first needs to be formulated in terms of invariant variables, which remain unaltered under superposed spatial rigid body motions, thereby preserving the principle of objectivity. This ensures that no spurious stresses are generated due to rigid body motions. Performing the time discretisation, see appendix C.1, then results in

\[
\tau = \frac{(1 - \omega)}{1 - \omega_t} \left[ F_{\Delta} \cdot \tau_t \cdot F_{\Delta}^c + (1 - \omega) \mathbb{H} : (e_{\Delta} - \Delta \lambda \mathbb{N}) \right],
\]

with \( F_{\Delta} = F \cdot [F^t]^{-1} \) the incremental deformation gradient tensor and \( e_{\Delta} \) the incremental Almansi strain tensor

\[
e_{\Delta} = \frac{1}{2} \left( I - F_{\Delta}^{-c} \cdot F_{\Delta}^{-1} \right).
\]

Only the nonlocality equation and the damage evolution law remain to complete the system:

\[
\ddot{z} - \ell^2 \omega \nabla^2 \tilde{z} = z,
\]

\[
\omega = 1 - \exp \left[ -b (Z - Z_t) \right], \quad \text{with} \quad Z = \{\max | \tilde{z}(t') | | t' \in [0, t] \}.
\]
4.5.2 Weak formulation and linearisation

Momentum equation

Multiplying Eq.(4.33a) with a vectorial weighting function $\mathbf{\tilde{w}}$ and integrating over the domain $\Omega$ with a boundary $\Gamma$ whose unit outward normal is $\mathbf{n}$, yields

$$\int_{\Omega} \mathbf{\tilde{w}} \cdot (\nabla \cdot \mathbf{\sigma}) \, d\Omega = 0 \quad \forall \mathbf{\tilde{w}}. \quad (4.35)$$

Applying the divergence theorem, using $\mathbf{\sigma} = \mathbf{\sigma}^c$ and applying the natural boundary condition $\mathbf{\sigma} \cdot \mathbf{n} = \mathbf{\tilde{p}}$ on $\Gamma$ leads to

$$\int_{\Omega} \mathbf{\tilde{w}}^c : \mathbf{\sigma} \, d\Omega = \int_{\Gamma} \tilde{w} \cdot \tilde{p} \, d\Gamma. \quad (4.36)$$

Because of the nonlinear and time dependent nature of the stress this equation needs to be solved iteratively by using a standard Newton–Raphson iteration procedure. First the left hand side is transformed to the last known equilibrium configuration at time $t$ according to $\mathbf{\tilde{V}} = \mathbf{F}_L \mathbf{c}^{-1} \cdot \mathbf{\tilde{V}}_t$ (with $\mathbf{F}_L = \mathbf{F} \cdot [\mathbf{F}^t]^{-1}$ the incremental deformation gradient tensor) and $d\Omega = (J/J^t) \, d\Omega^t$, and using $\mathbf{\tau} = J \mathbf{\sigma}$ results in

$$\int_{\Omega^t} (\mathbf{\tilde{V}}_t \mathbf{\tilde{w}})^c : \mathbf{F}_L^{-1} \cdot \mathbf{\tau} \frac{1}{J^t} \, d\Omega^t = \int_{\Gamma} \tilde{w} \cdot \tilde{p} \, d\Gamma. \quad (4.37)$$

Next, the unknown Kirchhoff stress tensor and incremental deformation gradient tensor are decomposed in an iterative manner,

$$\mathbf{\tau}^{(i+1)} = \mathbf{\tau}^{(i)} + \delta \mathbf{\tau} \quad \text{and} \quad \mathbf{F}_L^{(i+1)} = \mathbf{F}_L^{(i)} + \delta \mathbf{F}_L, \quad (4.38)$$

leading to the following linearised weak formulation of the equilibrium condition, where the superscript $(i)$ has been omitted,

$$\int_{\Omega^t} \left[(\mathbf{\tilde{V}}_t \mathbf{\tilde{w}})^c : (\delta \mathbf{F}_L^{-1} \cdot \mathbf{\tau} + \mathbf{F}_L^{-1} \cdot \delta \mathbf{\tau})\right] \frac{1}{J^t} \, d\Omega^t = \int_{\Gamma} \tilde{w} \cdot \tilde{p}^{(i+1)} \, d\Gamma - \int_{\Omega^t} (\mathbf{\tilde{V}}_t \mathbf{\tilde{w}})^c : (\mathbf{F}_L^{-1} \cdot \mathbf{\tau}) \frac{1}{J^t} \, d\Omega^t. \quad (4.39)$$

Introducing the variational deformation gradient tensor $\mathbf{L}_\delta^c$, allows for rewriting of $\delta \mathbf{F}_L^{-1}$ as follows,

$$\mathbf{L}_\delta^c = (\mathbf{\tilde{V}} \delta \mathbf{x})^c = \delta \mathbf{F}_L \cdot \mathbf{F}_L^{-1} \quad \rightarrow \quad \delta \mathbf{F}_L^{-1} = -\mathbf{F}_L^{-1} \cdot \delta \mathbf{F}_L \cdot \mathbf{F}_L^{-1} = -\mathbf{F}_L^{-1} \cdot \mathbf{L}_\delta^c. \quad (4.40)$$

Transformation back to the last known configuration then results in the linearised weak form of the momentum equation:

$$\int_{\Omega} (\mathbf{\tilde{V}} \mathbf{\tilde{w}})^c : \left[-\mathbf{\tau} : \mathbf{L}_\delta^c + \delta \mathbf{\tau}\right] \frac{1}{J} \, d\Omega = \int_{\Gamma} \tilde{w} \cdot \tilde{p}^{i+1} \, d\Gamma - \int_{\Omega} (\mathbf{\tilde{V}} \mathbf{\tilde{w}})^c : \mathbf{\tau} \, d\Omega. \quad (4.41)$$

Here $\Gamma$ is the boundary of the domain $\Omega$ with outward normal $\mathbf{\tilde{n}}$, and $\mathbf{L}_\delta^c$ is the variational displacement gradient tensor defined as $(\mathbf{\tilde{V}} \delta \mathbf{x})^c$. To obtain a consistent tangent, the variation of the Kirchhoff stress needs to be found. For the case presented here, the tangent relates this variation to $\mathbf{L}_\delta^c$ and $\delta \mathbf{z}$, which can be written as

$$\delta \mathbf{\tau} = \mathbf{K}_{uu}^x \cdot \mathbf{L}_\delta^c + \mathbf{K}_{uz}^x \cdot \delta \mathbf{z}, \quad (4.42)$$
where the superscript $x$ is either $e$ or $vp$ depending on whether the material behaves elastic or viscoplastic.

Derivation of the tensors $\kappa_{uu}^x$ and $K_{ug}^x$, which are the constitutive tangent operators, is outlined in appendix C.2. In case of elastic behaviour they read:

$$
\kappa_{uu}^e = \frac{1-\omega}{1-\omega^t} \left[ 2\mathbf{F}_\Delta : \mathbf{F}_\Delta^t : \mathbf{F}_\Delta^c - (1-\omega) \left\{ 2K\mathbf{F}_\Delta : \mathbf{e}_\Delta + \mathbf{H} : \left( \mathbf{F}_\Delta : \mathbf{F}_\Delta^c \right)^{-1} \right\} \right],
$$

(4.43)

and

$$
K_{uz}^e = -\frac{1}{1-\omega^t} \left[ \mathbf{F}_\Delta : \mathbf{F}_\Delta^t : \mathbf{F}_\Delta^c + 2(1-\omega)\mathbf{H} : \mathbf{e}_\Delta \right] \frac{\partial\omega}{\partial \bar{z}}.
$$

(4.44)

In the case of viscoplastic behaviour they become:

$$
\kappa_{uu}^p = \mathcal{A} : \kappa_{uu}^e + \frac{(1-\omega)^2}{(1-\omega^t)} \left( 2K\Delta\mathbf{F}_\lambda \mathbf{N} - \mathbf{H} : \mathbf{N} \mathbf{K}_{\gamma u} \right),
$$

(4.45)

and

$$
K_{uz}^p = \mathcal{A} : K_{uz}^e + 2\Delta\lambda\mathbf{H} : \mathbf{N} \frac{(1-\omega)}{(1-\omega^t)} \frac{\partial\omega}{\partial \bar{z}} - \frac{(1-\omega)^2}{(1-\omega^t)} \frac{\partial\tau_y}{\partial \bar{z}} \mathbf{N} \mathbf{K}_{\gamma z},
$$

(4.46)

with

$$
\mathcal{A} = \mathbb{I} - \frac{(1-\omega)^2}{(1-\omega^t)} \mathbf{H} : \mathbf{\tau}_{eq} \left( \frac{3}{2} - \frac{1}{2} \mathbb{I} - \mathbf{N} \right),
$$

(4.47)

$$
\mathbf{K}_{\gamma u} = \frac{c_1}{c_2} \left[ \mathbf{N} : \kappa_{uu}^e + 3\Delta\lambda \frac{(1-\omega)^2}{1-\omega^t} \mathbf{K}_I \right],
$$

(4.48)

and

$$
\mathbf{K}_{\gamma z} = \frac{c_1}{c_2} \left[ \mathbf{N} : \mathbf{K}^e_{uz} + \frac{\mathbf{\tau}_{eq} + \frac{3}{2} c_3 \Delta\lambda}{1-\omega} \frac{\partial\tau_y}{\partial \bar{z}} \right].
$$

(4.49)

The coefficients $c_1, c_2$, and $c_3$ are defined as follows,

$$
c_1 = \Delta\mathbf{F} \frac{\eta N}{(1-\omega)\phi_0} \left( \frac{\phi}{(1-\omega)\phi_0} \right)^{N-1},
$$

(4.50)

$$
c_2 = 1 + \frac{3}{2} c_1 c_3 + 3 \left( 1-\omega \right) \frac{\partial\tau_y}{\partial \bar{z}},
$$

(4.51)

$$
c_3 = \frac{(1-\omega)^2}{1-\omega^t} 2 \left[ G - K \ln(J) \right].
$$

(4.52)

**Nonlocal damage driving variable**

Multiplication of the nonlocality equation with a weighting function $w_z$ and integration of this product over the domain yields:

$$
\int_\Omega w_z \left( \bar{z} - \mathbf{t}_\omega^2 \nabla_\omega^2 \bar{z} \right) d\Omega = \int_\Omega w_z z d\Omega \quad \forall \quad w_z
$$

(4.53)
Applying partial differentiation and the Gauss divergence theorem results in
\[ \int_{\Omega} w_{z} \dd z \; d\Omega + \int_{\Omega} \tilde{\nabla} w_{z} \cdot \ell_{\omega}^{2} \tilde{\nabla} \dd z \; d\Omega = \int_{\Gamma} w_{z} \dd z \; d\Omega + \int_{\Gamma} w_{z} \ell_{\omega}^{2} \tilde{\nabla} \dd \Gamma \]  
(4.54)

The last term on the right hand side disappears after applying the usual homogeneous Neumann boundary condition \( \tilde{\nabla} \dd z \cdot \dd n = 0 \). Subsequent linearisation gives:
\[ \int \Omega w_{z} (\dd z - \delta z) \; d\Omega + \int \Omega \tilde{\nabla} w_{z} \cdot \ell_{\omega}^{2} \tilde{\nabla} \dd z \; d\Omega = -f_{\dd z} \]  
(4.55)

with
\[ f_{\dd z} = \int \Omega w_{z} (\dd z - z) \; d\Omega + \int \Omega \tilde{\nabla} w_{z} \cdot \ell_{\omega}^{2} \tilde{\nabla} \dd z \; d\Omega \]  
(4.56)

The variation of the damage driving variable, Eq.(4.34b), can be expressed in terms of \( L_{\dd}^{C} \),
\[ \delta z = K_{zu} L_{\dd}^{C} + K_{zz} \delta \dd z, \]  
(4.57)

where \( K_{zu} \) and \( K_{zz} \) are the following tangent operators,
\[ K_{zu} = \frac{1}{1 - \omega} \left( \Delta \lambda \mathbf{N} : \kappa_{uu}^{X} + \left[ \tau_{eq} + \frac{\tau_{eq} \Delta \lambda}{1 - \omega} \frac{\partial \omega}{\partial \lambda} \right] K_{\gamma u} \right), \]  
(4.58)

and
\[ K_{zz} = \frac{1}{1 - \omega} \left( \Delta \lambda \mathbf{N} : \mathbf{K}_{zz}^{X} + \left[ \tau_{eq} + \frac{\tau_{eq} \Delta \lambda}{1 - \omega} \frac{\partial \omega}{\partial \lambda} \right] K_{\gamma z} \right), \]  
(4.59)

leading to the following linearised weak form,
\[ \int_{\Omega_{0}} \left( w_{z} [1 - K_{zz}] \delta \dd z + \ell_{\omega}^{2} \tilde{\nabla}_{w} w_{z} \cdot \tilde{\nabla}_{w} \delta \dd z \right) d\Omega_{0} - \int_{\Omega_{0}} w_{z} K_{zu} L_{\dd}^{C} d\Omega_{0} = f_{\dd z}. \]  
(4.60)

### 4.5.3 Discretisation

In order to obtain a finite element formulation the equations are next discretised. The domain is subdivided into \( n \) elements. The unknown fields, \( u \) and \( \dd z \), are approximated by \( C^{0} \) continuous interpolation functions, \( N_{uu} \) and \( N_{\dd z} \), within each element. The finite element approximations for the unknown fields and their derivatives are written as
\[ \dd u = \sum_{A=1}^{n_{u}} N_{uu}^{A} \dd u_{A}, \quad \text{and} \quad \tilde{\nabla} \dd u = \sum_{A=1}^{n_{u}} \tilde{\nabla} N_{uu}^{A} \dd u_{A} \quad \text{with} \quad \tilde{\nabla} \dd u = \tilde{\nabla} N_{uu}^{A} \]  
(4.61)

\[ \dd \dd z = \sum_{A=1}^{n_{\dd z}} N_{\dd z}^{A} \dd \dd z_{A}, \quad \text{and} \quad \tilde{\nabla} \dd \dd z = \sum_{A=1}^{n_{\dd z}} \tilde{\nabla} N_{\dd z}^{A} \dd \dd z_{A} \quad \text{with} \quad \tilde{\nabla} \dd \dd z = \tilde{\nabla} N_{\dd z}^{A} \]  
(4.62)

where \( n_{u} \) and \( n_{\dd z} \) are the considered number of nodes used to represent the corresponding field variables in the element. The capital Latin subscripts indicate a nodal quantity. Following the standard Galerkin approach, the interpolation functions for the field variables are also used for the corresponding weighting functions.

Now a Cartesian basis in \( \mathbb{R}^{3} \) is chosen, with respect to which the equations are written in column and matrix format, which follows standard procedures. Further details are therefore omitted.
Table 4.1 — Phase field model parameters for the Sn–Pb solder system.

<table>
<thead>
<tr>
<th>parameter</th>
<th>symbol</th>
<th>unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>density $\alpha_0$</td>
<td>$\rho^\alpha$</td>
<td>[kg/m$^3$] 11300</td>
</tr>
<tr>
<td>density $\beta_0$</td>
<td>$\rho^\beta$</td>
<td>[kg/m$^3$] 7260</td>
</tr>
<tr>
<td>mobility $\alpha_0$</td>
<td>$M^\alpha I$</td>
<td>[m$^5$/(Js)] 2.2 \cdot 10^{-25} I</td>
</tr>
<tr>
<td>mobility $\beta_0$</td>
<td>$M^\beta I$</td>
<td>[m$^5$/(Js)] 0</td>
</tr>
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<td>diffusion length parameter</td>
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<td>[nm] 25</td>
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<td>$g_2$</td>
<td>[GJ/m$^3$] $-1.5263$</td>
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<td></td>
<td>$g_5$</td>
<td>[GJ/m$^3$] 0.8750</td>
</tr>
</tbody>
</table>

4.6 Material parameters and phase field coupling

Using a phase field approach, it is assumed that each phase obeys the same constitutive law, yet with different material parameters. Phase mixtures with different concentrations are then easily dealt with. The material parameters are therefore, as a first approximation, taken to be dependent on the mass fraction field which can be obtained by evaluation of the phase field model. In this way extensive information on the microstructure is included in a straightforward manner in the constitutive model. In previously published work [Ubachs et al., 2005a] this approach has already been applied for mechanical loading of eutectic Sn–Pb specimens, however, without the inclusion of damage.

The attention is focused on the influence of the microstructure through the phase field on the mechanical response of the material. Therefore, the microstructures used for the simulations were obtained through simulations of static ageing without any mechanical constraints, so the elastically stored energy could be omitted (section 4.2). Furthermore, the simulations of mechanical tests, which will be presented in the following, all take place within a short enough time period, such that diffusion processes can be neglected.

Figure 4.1 — Configurational free energy $\Phi_0$ for the $\alpha$ and $\beta$-phase at 150°C and the fitted single free energy curve as a function of the tin mass fraction $c$. 

![Configurational free energy](image-url)
Table 4.2 — Perzyna parameters for the Sn–Pb solder system.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>$\alpha_0$</th>
<th>$\beta_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young's modulus</td>
<td>$E$</td>
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<td>41</td>
</tr>
<tr>
<td>Poisson's ratio</td>
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<td>0.33</td>
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<td>$\tau_y$</td>
<td>[MPa]</td>
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</tr>
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<td>fluidity</td>
<td>$\eta$</td>
<td>[1/s]</td>
<td>$5.52 \cdot 10^{-6}$</td>
<td>$1.98 \cdot 10^{-5}$</td>
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<td>2.30</td>
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<td>[-]</td>
<td>13.53</td>
<td>21.67</td>
</tr>
</tbody>
</table>

The eutectic Sn–Pb system has been selected as the material to be used for the simulations. When cooling the Sn–Pb system from above its melting temperature, it solidifies and two phases separate out, the $\alpha$ phase, which contains mostly Pb and the $\beta$ phase, containing mostly Sn. The phase separation occurs because it lowers the total energy of the system. The equilibrium $c$ values for the phases after separation, the binodal points, can be found in a classical manner by constructing a double tangent to the free energy curve, see Fig. 4.1. In order to calculate the mass fraction fields, the following configurational free energy function has been used,

$$\Phi_0 = g_1 c + g_2 (1 - c) + g_3 c \ln(c) + g_4 (1 - c) \ln(1 - c) + g_5 c (1 - c)$$ (4.63)

The parameters needed for the phase field model are summarised in Table 4.1.

Although most of the material consists of either phase $\alpha$ or $\beta$, the interfaces are mixtures of these and the composition of the phases changes with temperature. Parameters are thus needed for the entire mass fraction range. The easiest way to accomplish this is by adopting a linear relation between the mass fraction and the material parameters,

$$P(c) = c \left( P^\beta - P^\alpha \right) + P^\alpha,$$ (4.64)

where $P$ is any material parameter and the subscripts $\alpha$ and $\beta$ refer to one of the phases present. The required parameters are found by fitting the material model to two Sn–Pb systems with different compositions, and thus different phase volume fractions. It is assumed that the bulk material properties are found by averaging the material properties of the individual phases using the rule of mixtures. The parameters can then be extrapolated to mass fraction values zero and one, denoted as $\alpha_0$ and $\beta_0$ respectively. The parameters used for the Perzyna model are listed in Table 4.2.

Using this linear interpolation approach implies that the interface behaviour would be the average of the individual phases next to it. An interface, however, may behave substantially different from the bulk material. The Sn–Pb system consists of two phases with different crystallographic structures, which gives rise to many crystallographic distortions in the interfacial regions. Unfortunately, quantitative data at the interface is largely missing. Therefore, these particular characteristics of the interface are neglected here, and the interfacial parameters are assumed to vary linearly from one phase to the next.

Determination of quantitative values for the damage parameters is difficult, considering the limited amount of experimental quantitative data at this scale of observation. Nevertheless, an attempt is made to capture the damage behaviour qualitatively.
From experimental evidence it appears that the $\alpha$ phase is capable of undergoing large amounts of plastic deformation without damaging. Voids and cracks are mainly seen to occur in the more brittle $\beta$ phase and along colony and phase boundaries [Kanchanomai et al., 2002a,b; Liu and Plumbridge, 2003; Ding et al., 2004; Shen et al., 2004; Kanchanomai and Mutoh, 2004]. Liu and Plumbridge [2003] also found fracture to occur occasionally through the $\alpha$ phase particles. Based on these experimental findings the damage parameters are specified independently in the $\alpha$ and $\beta$ phase. It is assumed that damage is concentrated in the $\beta$ phase, where a constant value for the damage parameters is specified. For the $\alpha$ phase damage is prohibited, for which the parameters are taken equal to zero for all the mass fractions below the eutectic composition ($c = 0.63$).

It is clear that interfaces are the preferred locations for damage initiation. Not only do stress concentrations often arise there, they are also more susceptible to damage because of the higher concentration of dislocations present and the misorientation between the crystal lattices. The phase field approach followed here, easily allows to account for this increased damage susceptibility at the interfaces. The location of the interfaces is known from the local and the nonlocal mass fraction fields, Fig. 4.2. In the bulk they will have equal values, but at the interfaces they will differ from one another. This information is next used to include the interfacial influence in the damage characteristics of the material. A weighting factor $w_\omega$ is introduced, which penalises the presence of interfaces, increasing the local driving force in their vicinity,

$$z_{\omega}(c, \bar{c}) = [1 + w_\omega(c - \bar{c})] z,$$

where $z_{\omega}$ is the weighted local damage driving variable, to be used in Eq.(4.34e).

When traversing the $\beta$ phase, cracks tend to follow grain boundaries. The current model does not take this explicitly into account. However, experimental observations of fracture regions in the $\beta$ phase indicate recrystallisation to occur in the neighbourhood of the cracks [Rynemark et al., 1993; Kanchanomai et al., 2002a].

The damage parameters are listed in Table 4.3. The specific choice of the values will be discussed in section 4.7.3, based on a preliminary investigation of the material behaviour without damage.
Table 4.3 — Damage parameters for the Sn–Pb solder system.

<table>
<thead>
<tr>
<th>parameter</th>
<th>symbol</th>
<th>unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>damage initialisation $\alpha_0$</td>
<td>$Z^\alpha_i$</td>
<td>$[\text{MJ/m}^3]$</td>
</tr>
<tr>
<td>damage initialisation $\beta_0$</td>
<td>$Z^\beta_i$</td>
<td>$[\text{MJ/m}^3]$</td>
</tr>
<tr>
<td>damage rate $\alpha$</td>
<td>$b^\alpha$</td>
<td>$[-]$</td>
</tr>
<tr>
<td>damage rate $\beta$</td>
<td>$b^\beta$</td>
<td>$5 \cdot 10^5$</td>
</tr>
<tr>
<td>damage weighting</td>
<td>$w_\omega$</td>
<td>$[-]$</td>
</tr>
<tr>
<td>damage length parameter</td>
<td>$\ell_\omega$</td>
<td>$[\text{nm}]$</td>
</tr>
</tbody>
</table>

4.6.1 Nonlocality and lengthscale

The nonlocal damage approach introduces a length scale in the model which is related to the distance at which material points influence each other. It should be emphasised that for high strain rates this nonlocal regularisation is not necessary. Mesh dependency will in that case be avoided through a sort of temporal nonlocality [Sluys and De Borst, 1994] introduced by the viscous nature of the material. However, as seen in Fig. 4.4, for the eutectic tin-lead alloy even at moderate strain rates the damage process shows a mesh dependency. The nonlocal regularisation effectively averages a local quantity over a finite domain. As is well-known for these types of nonlocal models, the element size should be smaller than the length scale parameter, which determines the localisation width. Lacking clear experimental input, the appropriate choice for the value of the internal length scale remains a point of discussion. For further calculations $\ell_\omega$ is chosen to be 200 nm, eight times the distance used for the interface length scale.

Figure 4.3 — Boundary conditions.

(a) $\ell_\omega = 0 \text{ nm}$, 20x5 elements. (b) $\ell_\omega = 0 \text{ nm}$, 40x10 elements.

(c) $\ell_\omega = 100 \text{ nm}$, 20x5 elements. (d) $\ell_\omega = 100 \text{ nm}$, 40x10 elements.

Figure 4.4 — Damage distributions for tensile test on near eutectic Sn–Pb solder, $c = 0.6$, using different length scales and meshes, strain rate $\dot{\varepsilon} = 0.002 [1/\text{s}]$. 

\[
\begin{align*}
\ell &= 4 \mu\text{m} \\
h &= 1 \mu\text{m}
\end{align*}
\]
4.7 Simulated microstructure evolution and material degradation

4.7.1 Microstructure evolution

Earlier results obtained with the phase field model of the coarsening process of eutectic Sn–Pb under static ageing conditions have been presented in Ubachs et al. [2004], where also a quantitative comparison with experimental observations was made. The results were used in Ubachs et al. [2005a], to perform a numerical investigation of the material behaviour of eutectic Sn–Pb solder, under the influence of an ageing microstructure. However, the RVEs used in that paper showed, for the aged microstructures, an α phase region traversing the complete domain. This may affect the simulations in a way that the as-cast microstructure, which does not have such a percolated microstructure, cannot be compared accurately with the aged microstructures. For this reason a new static ageing simulation has been performed, where the size of the domain has been increased to prevent percolation in the RVEs. The same parameters as used in Ubachs et al. [2004] have been used. They are summarised in Table 4.1. The initial condition was taken from a micrograph of eutectic Sn–Pb solder taken immediately after casting and quenching in liquid nitrogen. Fig. 4.5 shows the microstructure of the sample at various stages during the simulation of a static ageing process at 150°C.

4.7.2 Mechanical response without damage

The data obtained from the thermal ageing simulation is now used to investigate the response to mechanical loading of the as-cast microstructure and the microstructure after thermal ageing for 84 hours. A cycled uniaxial tensile test is performed using periodic boundary conditions, see Fig. 4.6. The prescribed displacement of node $\vec{x}_{br}$ ranges from 0 to 0.2 $\mu$m, amounting to a maximum of 1% macro strain. The strain is applied at a rate of 0.002 [1/s].

The resulting reaction force curves are depicted in Fig. 4.7. There is no significant difference
between the two curves, microstructural sizes do not seem to influence the macroscopic response of the material significantly. This is not consistent with the results found in Ubachs et al. [2005a], however, as noted before, the RVEs used there are not representative enough, whereas here the specimens are larger and therefore more appropriate as RVEs. It does once again indicate the importance of the microstructure, especially for very small solder joints. Both the microstructure show a gradual, transient, increase in their reaction force, caused by the increase of the yield stress through isotropic hardening. However, the microstructure does have an impact on the distribution of the stresses and strains. During each cycle, when excluding damage, the spatial distribution of the field variables evolve qualitatively similar, only their magnitudes change, therefore, results are only shown for one specific time instance.

Fig. 4.8 shows $\tau_{eq}$ after 5 cycles for both microstructures. Note that since the elastic deformation is small and the plastic flow is isochoric the Kirchhoff stress will differ only slightly from the Cauchy stress. Both microstructures provide qualitatively the same results, with higher stresses appearing in the $\beta$ phase. However, the aged microstructure induces higher stress concentrations, while the as-cast microstructure shows a more diffuse stress field. A similar picture is found for $\varepsilon_{vp}$ (Fig. 4.9), for which the highest values appear within the $\alpha$ phase. Here, the aged microstructure also exhibits more intense localisation.

The plastic work, Fig. 4.10 also exhibits a more pronounced localisation behaviour, but it

Figure 4.7 — Macroscopic stress versus displacement for the cyclic tensile test excluding damage.

Figure 4.6 — Dimensions and boundary conditions used for cyclic tensile loading
Figure 4.8 — Equivalent Kirchhoff stress distributions [MPa] after 5 mechanical cycles, excluding damage.

Figure 4.9 — Effective plastic strain distributions [-] after 5 mechanical cycles, excluding damage.

Figure 4.10 — Plastic work distributions [MJ/m³] after 5 mechanical cycles, excluding damage.
is not confined to one of the phases. Concentrations of equal magnitude can be found in both the $\alpha$ and $\beta$ phase. Clearly there seem to be preferential concentrations at the level of the interfaces. Again the coarser, aged microstructure yields the highest local values.

### 4.7.3 Damage caused by cyclic loading

Since the distribution of the field variables does not change under the influence of mechanical cycling, the amount of cycling prior to damage onset will have a negligible influence on the damage process, qualitatively speaking. For this reason, the initiation threshold for damage is chosen very low, since this convenient choice does not influence the qualitative description of the damage process. The damage initiation parameter $Z_i$ is set to 1 [MJ/m$^3$], and the damage rate $b$ is set to $5 \cdot 10^5$, large enough for the damage process to require several cycles to go from damage initiation to complete damage.

It has been shown before that when excluding damage, higher local values where found in the coarser microstructure for the stresses, the plastic strain, and most importantly, the damage driving variable, the plastic work. This would suggest that the aged microstructure would be the weaker one, since the damage threshold will be passed here first. Fig. 4.12

![Figure 4.12 — Average applied stress versus displacement including damage (cyclic shearing).](image-url)
Figure 4.13 — Damage distributions after 2 cycles.

Figure 4.14 — Damage distributions after 7 cycles.

Figure 4.15 — Damage distributions after 10 cycles.
shows the macroscopic stress versus the cumulative displacement resulting from the cyclic loading simulation incorporating the damage evolution. Initially, no significant difference in the damage evolution can be discerned between the as-cast and the aged microstructures from this curve. After a sufficient number of cycles however, the macroscopic stress for the aged microstructures drops below the one for the as-cast microstructure, indicating the damage evolution to proceed more rapidly. For the final number of cycles the level of the macroscopic stress is of the coarsest (aged 84 hours) microstructure is the lowest and that of the finest (as-cast) the highest, with the intermediate microstructure (aged 24 hours) yielding a response between these two.

Additional calculations were performed with the microstructures rotated over 90 degrees. Similar results were obtained, confirming the observed trend is caused by the level of coarseness of the microstructure.

In the following a more detailed comparison is made between the as-cast and the microstructure aged for 84 hours, referred to as the aged microstructure.

Damage occurs in the $\beta$ phase in such a way that the weakened zones line up with ductile $\alpha$ phase regions to form localisation bands, see Figs. 4.13–4.15, resulting in shear bands at an angle of $45^\circ$ with the loading direction, see Fig. 4.16. These bands of increased damage and viscoplastic strain unload the rest of the material, lowering the stress throughout the microstructure. Damage is observed to initiate at interfaces and subsequently propagate rapidly through the $\beta$ phase towards an adjacent interface. Fig. 4.15 shows the damage field after 7 cycles, when the aged microstructure has a macroscopic stress that dropped below the stress of the as-cast microstructure. The reason for this more rapid decrease in the aged microstructure is the propagation of a band of nearly complete damage traversing the entire specimen.

Similar to the simulations without damage, the field variables present a more diffuse view in the as-cast microstructure. This also holds for the damage, which is the reason why the overall response of the aged microstructure does not differ significantly from that of the as-cast one. Fig. 4.17 shows the normalised damage distribution curves during the cycling process. They are found by calculating the amount of area with a certain amount damage
Figure 4.17 — Development of the damage distribution during mechanical cycling in tensile for the as-cast and the aged microstructure.

and dividing it by the total area. Here, the impact of the microstructure on the damage evolution is more clearly visible. At any time instance during the cycling, the as-cast microstructure has a larger area where damage is low compared to the aged microstructure. After the damage evolution has been initiated, this area remains constant during the cycling process. The situation is different for areas where the damage is critical (close to one). This area expands during loading, and grows faster for the aged microstructure compared to the as-cast one.

4.8 Discussion

Through numerical simulations it was found that the coarseness of the microstructure influences the distribution of stresses, strains, and damage within the microstructure. Initially, the average stress during mechanical cycling is not significantly different for the two microstructures. This is consistent with the analysis performed by Shen et al. [2001], assuming a perfect plastic material behaviour. Their results suggest that a coarser microstructure is not stronger or weaker than its finer counterpart. Yet, plastic strain accumulated faster in the coarser structure under the same loading conditions.

It has been found experimentally that during thermomechanical cycling of a eutectic Sn–Pb solder joint the coarsening process will not be homogeneous. Regions of high shear strain within an eutectic Sn–Pb connection appear to coarsen more rapidly under the influence of mechanical loading and high homologous temperature [Frear et al., 1988]. These regions of inhomogeneous coarsening are known to be crack nucleation and propagation sites [Frear et al., 1988, 1997; Hacke et al., 1998; Shen et al., 2004; Rynemark et al., 1993]. Results presented here show stresses and strains to localise more in a coarser microstructure,
eventually leading to earlier initiation of damage compared to a finer microstructure, which is consistent with the experimental observations.

Furthermore, experimental observations show damage predominately to occur along interfaces between colonies and phase regions and between \( \beta \) grains [Frear, 1989; Kanchanomai et al., 2002a,b; Liu and Plumbridge, 2003; Ding et al., 2004; Kanchanomai and Mutoh, 2004]. In accordance with these observations, the calculations show plastic work to localise preferably near interfaces, causing damage to be initiated in these regions and subsequently propagating through the \( \beta \) phase. This supports the choice of the damage driving variable and the damage evolution parameters.

### 4.9 Conclusions

The influence of thermal ageing on the damage characteristics of Sn–Pb has been investigated by comparing the damage initiation and propagation in two microstructures, one as-cast microstructure and the other resulting from thermal ageing of the former. A viscoplastic material model was used to describe the creep deformation behaviour observed in solders in combination with a nonlocal continuum damage model. The plastic work was chosen as the damage driving variable. Through the use of a phase field model, microstructural information has been incorporated in the formulation. Material parameters are assigned to the different phases and an additional penalty parameter is introduced to account for the higher damage susceptibility of interfaces.

In the current model the material is modelled with a viscoplastic constitutive equation in which damage evolution is included. It is shown that after a sufficient number of cycles a stress decrease, triggered by microstructural failure of the specimen, occurs earlier for the coarse microstructure than for the fine microstructure.

The drop in average stress is attributed to a band of nearly completely damaged material \( (\omega > 0.95) \) traversing the entire specimen. This is more rapidly accomplished in a coarser microstructure since the path the damage can take is less obstructed. For a fine microstructure, the ductile \( \alpha \) phase regions tend to interrupt preferential damage growth directions and spread the damage over a larger area of material.

Stresses, strains, and plastic work are more evenly distributed in a finer microstructure as opposed to the coarse microstructure where the local variables reach higher values. In the fine microstructure, a larger area of low damage is found during the entire cycling process. The coarse microstructure however, shows a larger area of material to be critically damaged after sufficient cycling. Furthermore, the fraction of heavily damaged material also increases more rapidly in the case of a coarser microstructure.

In conclusion, the presented model is able to predict microstructural dependent damage evolution in eutectic Sn–Pb, indicating coarser microstructures to be more susceptible to damage than finer ones. Qualitative comparison with experimental observations in the literature shows an adequate agreement.
A finite element analysis has been performed of the thermal cycling of Sn–Ag–Cu solder. Experimental observations revealed damage to occur at grain boundaries even in the absence of external mechanical loading. The effect of the anisotropy of the material has been investigated through a three-dimensional simulation of the experiment. Anisotropy was included in the model in the elastic and thermal expansion properties. Stress concentrations arise at grain boundaries, dependent on the orientation of the connected grains and their interface. Qualitative comparison of the numerical results with the experimental observations indicated a reasonable agreement. A more accurate prediction requires three-dimensional experimental data on the grain structure of the sample.

5.1 Introduction

The Sn–Pb (tin–lead) system has been a widely used solder material in microelectronics for a long time. Legislation in Europe however, imposes the manufacturing of lead–free electrical and electronic equipment in 2006, with some exceptions. Many Sn based Pb–free solder alloys have been proposed as a replacement for the eutectic Sn–Pb, e.g. Sn–Ag, Sn–Bi, Sn–Zn, Sn–Zn–Bi, Sn–Ag–Bi.

Recently, industry has focused its interest on a near-eutectic Sn–Ag–Cu alloy (SAC) because of its comparatively low melting temperature, competitive price, and good mechanical properties [Suhling et al., 2004; Suganuma, 2001; Miric and Grusd, 1998; Harrison et al., 2001]. Research on the microstructure of SAC [Salam et al., 2004; Lehman et al., 2004], its evolution [Allen et al., 2004a,b], and mechanical properties [Amagai et al., 2002; Kim et al., 2002, 2003; Korhonen et al., 2004; Pang et al., 2004; Plumbridge et al., 2001; Xiao et al., 2004; Yu et al., 2004; Guo et al., 2001] is starting to appear in the literature, but more is needed to gain a thorough understanding of the behaviour of the alloy. Some modelling work has also been performed, usually considering the material as a single phase, isotropic material [Kim and Jung, 2004; Lau et al., 2004]. Erinc et al. [2004] simulate fatigue damage in a SAC solder joint taking into account grain boundaries using cohesive zones.

It has been reported in literature that the anisotropic character of the $\beta$-Sn crystal structure (BCT) can lead to stress concentrations at Sn grain boundaries during thermal cycling [Subramanian and Lee, 2004; Matin et al., 2005]. Furthermore, it has also been shown that localised deformation occurs at the grain boundaries of Sn-rich solders upon thermal loading [Telang et al., 2004]. Recently, experimental evidence has been presented that these stress concentrations can actually lead to damage, purely due to the influence of thermal cycling without any external mechanical constraints [Matin et al., 2005].

The SAC alloy is not the only material were damage tends to nucleate at the grain boundaries. Bieler et al. [2005] investigate the effect of crystallographic texture on the nucleation and growth of cavities in Ti–6Al–4V alloy and also found preferential damage evolution at grain boundaries with a high misorientation angle. Similar results were presented by Xun et al. [2005] for an Al–Li alloy. They argued that grain boundary sliding and cavitation occurs at high angle grain boundaries between groups of grains with a similar orientation which slide as a unit. Chen and Li [1998] showed, using an anisotropic elastic finite element model of a multicrystal system, that stress concentrations arise at certain grain boundaries and triple junctions. Furthermore, Vehoff et al. [2004] showed, using an elastically anisotropic model, that predicted stress concentrations arising at grain boundaries of Fe–2.7Si correspond well with experimentally observed crack nucleation sites. Matin et al. [2005] showed in a comparison between experimentally obtained data and a simulation for a thermally cycled SAC specimen, using an elastic analysis including anisotropic elastic and thermal expansion properties, that the locations of predicted stress concentrations correlated reasonably with damaged areas found in the experimental observations.

In this paper, an analysis of the damage process of a near-eutectic SAC solder due to thermal cycling is presented, with a particular emphasis on the influence of the SAC microstructure on the mechanical response. Simulations of a cyclic thermal loading experiment of a SAC specimen have been performed using the finite element method (FEM). The required experimental information on the microstructure and the damage evolution of the specimen
has been obtained using Orientation Imaging Microscopy (OIM) and optical microscopy Matin et al. [2005].

Mechanical and thermal anisotropy of the material are accounted for in the model through the elastic constants and the coefficients of thermal expansion, respectively. Furthermore, since soldered connections usually operate at high homologous temperatures, they are prone to high temperature deformation mechanisms. Therefore, the mechanical response is captured by a viscoplastic material model based on the viscoplastic strain rate function defined by Perzyna [1966]. Next, the model is extended to include damage initiation and propagation. To ensure the well-posedness of the problem and mesh insensitivity the damage is driven by a nonlocal quantity: the weighted average of its local counterpart over a finite domain.

To accurately incorporate the anisotropic behaviour of the material the FEM model has been implemented in 3D. Boundary conditions associated with a two-dimensional implementation, either plane strain or plane stress, would be too strict, leading to incorrectly predicted stress states.

The paper presents a numerical analysis of a thermal cycling experiment, previously published by Matin et al. [2005]. There a numerical analysis was also performed, including elastic and thermal anisotropy. The results showed a correlation existed between areas where stress concentrations were predicted and areas where microstructural damage was observed experimentally. However, no direct relation was found between the magnitude of the predicted stress concentrations and experimentally observed damage. In this contribution, possible reasons for this are investigated. The influence of the boundary conditions and the grain boundary orientation on the stress concentrations is examined and a more accurate description of the material behaviour is used. The temperatures at which the thermal cycling is performed are close to the liquidus point of the SAC solder alloy, which causes the material to demonstrates time-dependent plastic behaviour, e.g. creep. Since this might influence the response of the material, here, an elasto-viscoplastic material model is used to describe the mechanical behaviour of the solder. Furthermore, the local occurrence of damage influences the stress and strain field in the neighbouring material, which might exhibit relaxation. Therefore, a continuum damage formulation is included in the model.

First, in section 5.2, the experimental procedures and observations are recalled. Next, details on the material model are presented in section 5.3, after which simulations are presented in section 5.4 and the results are confronted with the experimental findings. Finally, conclusions are formulated in section 5.5.

5.2 Experimental thermal cycling results

5.2.1 Sample preparation and procedure

Bulk SAC solder specimens were prepared from a commercial solder paste Sn–96L–NS (Balverzinn, Germany). The alloy was sealed in a cylindrical quartz ampule of 10 mm diameter and 50 mm length under a vacuum of $10^{-4}$ Pa. The ampules were superheated to 100 K above the eutectic temperature in a furnace at a heating rate of 10 K/min and held there for
5 min. To ensure the homogeneity of the alloy, the ampules were carefully shaken before quenching them in liquid nitrogen to 77 K. The purpose of quenching was to obtain bulk specimens with a fine microstructure, which is characteristic for a solder interconnection used in microelectronics. Solder joints cool rapidly due to the small size and furthermore have a large surface to volume ratio, providing many nucleation sites for grains. A piece was cut from the sample, which was next ground using silicon carbide polishing paper with grit sizes 1000 to 2400, followed by fine polishing with diamond suspensions of 6, 3, and 1 µm. Finally, mechanical polishing was performed with a solution of 0.05 µm colloidal silica. The resulting specimen had a height of approximately 4 mm and a diameter of approximately 10 mm.

After preparation, the alloy was thermally cycled between 293 K and 353 K with 5 minutes hold at low temperature and 15 minutes hold at high temperature. Temperature changes were applied with a ramp rate of 30 K/min using a heating-cooling stage (LINKAM LTS-350) with a temperature accuracy of 1 K. The specimen was subjected to 1000 thermal cycles.

Polarisation microscopy was used for the characterisation of the microstructure on the plane section of specimens before and after thermal cycling. Back-Scattered Electron (BSE) images (FEI Sirion HR-SEM) were taken from selected areas on the plane section of samples before and after thermal cycling to evaluate the microscopic deformation mechanisms.

Orientation Imaging Microscopy (TSL OIM detector) was performed to obtain local orientation information by collecting Electron Back Scattering Diffraction (EBSD) patterns. A 30 kV beam with a current intensity of about 8 nA was used. Crystallographic orientation data of the sample was collected before and after thermal cycling.

5.2.2 Microstructure and evolution

The crystallography of the sample was studied in detail using Orientation Imaging Microscopy (OIM). Because of the size of the sample, several OIM scans were made of parts of the sample, which were then combined to obtain the full sample data. Fig. 5.1a shows the [001] inverse pole figure (IPF) map of the sample. A clean-up routine has been applied to the data, sub-micron size Ag₃Sn and Cu₆Sn₅ particles have been removed, and each individual grain has been identified with a single orientation.

Above 13°C the crystal structure of Sn is Body Centred Tetragonal (β-Sn), with lattice parameters [100] = [010] = 0.5632 nm and [001] = 0.3182 nm. From the OIM data the angle between the [001] directions belonging to all pairs of adjacent measurements was calculated. Fig. 5.1b shows a map of these misorientation angles, ranging from 0 to 90°. The locations of the grain boundaries can be distinguished clearly.

A comparison of OIM data for the sample before and after the thermal cycling did not show a noticeable change in the grain geometry or orientation, although microstructural changes were observed related to damage.

Figs. 5.2a and 5.2b show polarisation micrographs of the as-solidified SAC alloy, and the same specimen after the treatment described in section 5.2.1, respectively. In each case several micrographs were combined to represent the entire specimen surface. The individual grains can be distinguished in the micrographs.

Thermal cycling results in significant microstructural changes, which are shown in Fig. 5.2b.
Viscoplastic nonlocal damage modelling of thermal fatigue

(a) [001] IPF intensity map showing the average crystal orientation per grain.
(b) Misorientation angles between the [001] crystal axes.

**Figure 5.1** — Microstructural information obtained by OIM.

(a) Before cycling.
(b) After 1000 thermal cycles.

**Figure 5.2** — Optical micrographs of the SAC specimen before and after thermal cycling between 293 and 353 K [Matin et al., 2005].
Sharp damage patterns have developed at or in the neighbourhood of the grain boundaries. Their appears to be a preference for grain boundaries with a larger mismatch angle, although it is not obvious that a larger angle results systematically in more damage. For example, grain $g_4$ is damaged mostly at its boundary with $g_3$, but it has the largest mismatch angle with $g_2$. Also, no damage is visible at some of the grain boundaries with a high mismatch angle, e.g. between $g_1$ and $g_2$ and the right and lower boundary of $g_{15}$. For a more detailed analysis on the damage development see Matin et al. [2005].

In the following sections, a material model will be introduced, which is used to investigate the microstructural evolution of the SAC specimen. Information obtained by OIM is used as input for the model in order to incorporate the anisotropic heterogeneous properties of the microstructure.

### 5.3 Anisotropic elasto-viscoplastic gradient enhanced damage model

An elasto-viscoplastic material model is selected to account for the time-dependent behaviour of the material. Damage initiation and propagation are accounted for by a gradient-enhanced continuum damage description. The elastic material properties are taken to be anisotropic, while the viscoplastic behaviour remains isotropic. More detailed information on the model and its implementation in a FEM framework can be found in Ubachs et al. [2005b]. A summary of the essential features is presented below.

#### 5.3.1 Kinematics

Transformation from the undeformed configuration at time $t_0$ to the current configuration at time $t$ is described by the deformation gradient tensor $F$, the determinant of which, $J = \text{det}(F)$, represents the relative volume change. The conventional multiplicative decomposition is used to split the deformation into an elastic and an inelastic (viscoplastic) part, denoted by subscripts $e$ and $vp$ respectively:

$$F = (\tilde{\nabla}_0 \tilde{x})^c = F_e \cdot F_{vp}.\quad (5.1)$$

The right and left Cauchy–Green strain tensors, $C$ and $B$, and the Green-Lagrange strain tensor $E$ are given by

$$C = F^c \cdot F, \quad B = F \cdot F^c, \quad \text{and} \quad E = \frac{1}{2}(C - I).\quad (5.2)$$

Material velocity is accounted for by the deformation rate tensor $D$ and the spin tensor $\Omega$, respectively, being the symmetric and skewsymmetric parts of the velocity gradient tensor $L$. Decomposition (5.1) implies the introduction of elastic and viscoplastic rate tensors:

$$L = (\tilde{\nabla}_0 \tilde{v})^c = \dot{F} \cdot F^{-1} = \dot{F}_e \cdot F_e^{-1} + \dot{F}_{vp} \cdot F_{vp}^{-1} \cdot F_e^{-1}$$

$$= L_e + L_{vp} = (D_e + \Omega_e) + (D_{vp} + \Omega_{vp}).\quad (5.3)$$

To make the decomposition unique it is commonly assumed that the viscoplastic spin $\Omega_{vp}$ is zero. Superimposed material rotations are thus fully represented in $F_e$. 


5.3.2 Elasto-viscoplasticity

The Kirchhoff stress tensor is defined as

$$\tau = 2F \cdot \frac{\partial \Phi}{\partial C} \cdot F^c,$$

where $\Phi$ is the free energy, i.e. elastically stored energy, in the system. Since a time-dependent material model will be used, the elastic constitutive relation should be formulated in a rate form. Transforming Eq.(5.4) to the reference configuration, performing time differentiation, and transforming the result back to the current configuration, results in the following expression for the objective Truesdell derivative of the Kirchhoff stress,

$$\nabla \tau = 4 \left[ F \cdot \left( F \cdot \frac{\partial^2 \Phi}{\partial C^2} \cdot P^c \right)^{lc,rc} \cdot F \right] : D = \mathbb{H} : D.$$ (5.5)

Here $\mathbb{H}$ is a fourth order material tensor and the superscript $lc,rc$ stands for the left and right conjugate of a tensor, i.e. $A^{lc,rc} = A_{ijkl} \tilde{e}_j \tilde{e}_i \tilde{e}_k \tilde{e}_l$.

The problem under consideration focusses on the evolution of damage due to thermal loading. Accounting for thermal strains resulting from temperature variations in the constitutive relation yields

$$\nabla \tau = \mathbb{H} : (D - D_T), \quad \text{with} \quad D_T = \alpha \widetilde{T}.$$ (5.6)

The thermal rate of deformation is characterised by $\alpha$, a second-order tensor containing the crystallographic coefficients of thermal expansion (CTEs), and $\widetilde{T}$ the rate of change of the temperature.

The BCT crystal structure of Sn results in anisotropic mechanical and thermal material properties. This anisotropy is accounted for in the description of the elastic behaviour and in the expansion of the material due to thermal changes. Since $\mathbb{H}$ is a three-dimensional fourth-order tensor it has $3^4 = 81$ components, from which only 21 are independent in the general case.

For a tetragonal material there are no coupling terms between extensional strains and shear stresses ($H_{ijkl} = H_{ijkk} = 0$), and shear strains in one plain do not cause shear stresses in other planes. Furthermore, because of the symmetry of the crystal some of the remaining components are equal. Writing the material tensor $\mathbb{H}$ in matrix format, taking the crystal axes as the reference configuration, then results in

$$\begin{bmatrix}
H_{1111} & H_{1122} & H_{1133} & 0 & 0 & 0 \\
H_{1122} & H_{1111} & H_{1133} & 0 & 0 & 0 \\
H_{1133} & H_{1133} & H_{3333} & 0 & 0 & 0 \\
0 & 0 & 0 & H_{2323} & 0 & 0 \\
0 & 0 & 0 & 0 & H_{2323} & 0 \\
0 & 0 & 0 & 0 & 0 & H_{1212} \\
\end{bmatrix}.$$ (5.7)

This system contains 6 independent elasticity components only.

The thermal anisotropy is expressed by the second-order tensor $\alpha$. When the frame of reference coincides with the crystallographic axes only the 11, 22, and 33 components, respectively belonging to lattice directions [100], [010], and [001] are nonzero.
The material behaves elastically until the equivalent stress $\tau_{eq}$ exceeds the current yield stress $\tau_y$. Whether the viscoplastic response is activated or not is evaluated using the Von Mises-Huber yield function $\varphi$,

$$\varphi = \tau_{eq} - \tau_y \leq 0 \quad \text{with} \quad \tau_{eq} = \sqrt{\frac{3}{2} \tau^d : \tau^d}, \quad (5.8)$$

where the yield stress $\tau_y$ generally accounts for the linear or nonlinear hardening or even softening response due to damage. Here, it is a function of the initial yield stress $\tau_{y0}$, the effective viscoplastic strain measure $\varepsilon_{vp}$ and its rate $\dot{\varepsilon}_{vp}$.

During viscoplastic deformation the yield stress will change as a function of the effective viscoplastic strain. Plastic flow does not induce any stresses in the material, therefore the constitutive model becomes

$$\varv = \mathbb{H} : (\mathbf{D} - \mathbf{D}_T - \mathbf{D}_{vp}). \quad (5.9)$$

With ongoing viscoplastic deformation, the direction of the viscoplastic strain rate $\mathbf{D}_{vp}$ is defined by the commonly used normality or associative flow rule, which states that the strain rate is directed along the normal to the yield surface and denoted by $\partial \varphi / \partial \tau$. Its length is characterised by the magnitude of the rate of the viscoplastic multiplier $\dot{\lambda}$. Thus the expression for the viscoplastic strain rate can be formulated as [Perzyna, 1985]:

$$\mathbf{D}_{vp} = \dot{\lambda} \mathbf{N}, \quad \text{where} \quad \mathbf{N} = \frac{\partial \varphi}{\partial \tau} = \frac{3}{2} \frac{\tau^d}{\tau_{eq}}, \quad \text{and} \quad \dot{\lambda} = \eta \varphi(\varphi), \quad (5.10)$$

with $\eta$ a fluidity parameter which determines the magnitude of the viscoplastic strain rate and $\varphi(\varphi)$ the overstress function for which a power law is adopted [Perzyna, 1971]:

$$\varphi(\varphi) = \left(\frac{\tau_{eq} - \tau_y}{\varphi_0}\right)^N = \left(\frac{\varphi}{\varphi_0}\right)^N, \quad (5.11)$$

with $N$ the rate sensitivity parameter and $\varphi_0$ a scaling parameter. Higher values of $N$ reduce the strain rate sensitivity of the model [Perić, 1993].

The rate of the hardening variable $\varepsilon_{vp}$ and of the viscoplastic multiplier $\dot{\lambda}$ are coupled in a straightforward manner, $\dot{\varepsilon}_{vp} = \dot{\lambda}$. From Eq. (5.10) the definition of $\dot{\varepsilon}_{vp}$ then follows,

$$\dot{\varepsilon}_{vp} = \dot{\lambda} = \sqrt{\frac{2}{3} \mathbf{D}_{vp} : \mathbf{D}_{vp}} \quad \text{and} \quad \varepsilon_{vp} = \int_0^t \dot{\varepsilon}_{vp} \, dt. \quad (5.12)$$

### 5.3.3 Damage

To account for the effect of damage, an isotropic damage parameter $\omega$ is introduced. This damage parameter can be interpreted as the reduction of the effective volume of the material due to void nucleation, growth, and coalescence. It reduces the amount of elastic free energy that can be stored in the material volume. For the undamaged material $\omega$ is zero, and for completely failed material $\omega$ equals one. To incorporate the effect of damage on the elastic response of the material, in Eq.(5.4) $\Phi$ is replaced by $(1 - \omega)\Phi_e$. 
Performing differentiation with respect to time, and applying a push forward to the current configuration, similar to the procedure in section 5.2, now yields the rate of stress,

$$\nabla \dot{\tau} = (1 - \omega) H : \left( D - D_T - D_{vp} \right) - \frac{\tau}{(1 - \omega)} \dot{\omega},$$  \hspace{1cm} (5.13)

where the second term originates from time differentiation of the damage term. As damage increases in a material volume it gradually loses its ability to store elastic energy but also to dissipate energy plastically. Therefore the damage parameter is also incorporated in the viscoplastic part of the model,

$$\dot{\lambda} = \eta \left( \frac{(1 - \omega) - \tau_{eq}}{\varphi_0} \right)^N \left( \frac{\tau_{eq} - (1 - \omega) \tau_y (1 - \omega) \varphi_0}{(1 - \omega) \varphi_0} \right)^N.$$  \hspace{1cm} (5.14)

The term $(1 - \omega)^{-1} \tau_{eq}$ can be considered to be the effective stress, i.e. the actual stress that is in fact carried by the undamaged material after correcting for the loss of cross sectional area due to damage.

### 5.3.4 Gradient enhanced damage

It is known that rate dependency (viscosity) may restore the well posedness of the initial value problem [Wang et al., 1997]. Viscosity regularises the solution by implicitly introducing a length scale into the model, which causes the localisation region to be more diffuse compared to an inviscid plastic material [Glema et al., 2000]. These viscous terms are sometimes referred to as temporal nonlocalities [Sluys and De Borst, 1994]. However, for practical purposes this effect is usually only sufficient for highly rate-sensitive materials or high loading rates and the numerical results can still show a mesh dependency [Wang and Sluys, 2000; Voyiadjis et al., 2004; Peerlings et al., 2004]. Therefore, a nonlocal approach will be used here to incorporate damage; the damage is driven by a nonlocal internal variable $\bar{z}$ which is the weighted average of its local counterpart over a finite domain [Bažant and Pijaudier-Cabot, 1988; Steinmann, 1999; Geers et al., 2001, 2003; Boers et al., 2005],

$$\bar{z}(\bar{x}) = \int_{\Omega_0} g(\bar{y}) z(\bar{x} + \bar{y}) d\bar{y},$$  \hspace{1cm} (5.15)

where $z$ is a local quantity which acts as the damage driving variable, $\Omega_0$ is the region around the material point with position vector $\bar{x}$ in the undeformed state and $\bar{y}$ a local position vector with respect to $\bar{x}$.

Eq. (5.15) can be rewritten in a differential form [Peerlings et al., 1996, 2001; Geers et al., 2003]:

$$\bar{z} - \ell_\omega^2 \nabla^2_{\Omega_0} \bar{z} = z,$$  \hspace{1cm} (5.16)

where the subscript $\Omega_0$ indicates the material (initial undeformed) configuration, i.e. the Laplacian is taken with respect to the initial configuration. Usually the Neumann boundary condition $\nabla_{\Omega_0} \bar{z} \cdot \vec{n}$ is applied. The parameter $\ell_\omega$ is a material length parameter accounting for microstructural and micromechanical events. Because it is defined in the material configuration, averaging is always performed over the same material volume when $\ell_\omega$ is taken constant.
The damage parameter $\omega$ is related to a history parameter $Z$, which is the largest value of $\ddot{z}$ attained during the complete deformation history of a material point,

$$Z = \{\max[\ddot{z}(t') | t' \in [0, t)]\}.$$  (5.17)

With respect to the history parameter the following set of Kuhn-Tucker conditions hold,

$$\dot{Z} \geq 0, \quad \ddot{z} - Z \leq 0, \quad \dot{Z}(\ddot{z} - Z) = 0, \quad \text{and} \quad Z(t = 0) = Z_i,$$  (5.18)

and $Z_i$ is the initial damage threshold.

It now remains to chose a damage evolution function which relates $Z$ to $\omega$. When the material is undamaged, the damage variable equals zero and for completely damaged material it equals one. The following evolution law is used,

$$\omega = 1 - \exp\{-b(Z - Z_i)\}.$$  (5.19)

where $b$ determines the rate of damage.

### 5.3.5 Crystal orientation

The crystal structure of $\beta$-Sn is Body Centred Tetragonal. This means that its mechanical response will be anisotropic. Here this anisotropic behaviour is included in the thermal properties by the tensor $\alpha$, Eq.(5.6), representing the thermal expansion properties, and in the mechanical properties by the fourth-order material tensor $\mathbb{H}$. The viscoplastic and damage properties are assumed to be isotropic.

From Orientation Imaging Microscopy the Euler angles $(\phi_1, \theta, \phi_2)$ characterising the crystallites are obtained. The first rotation is around the $z$-axis, resulting in a new axes system $(x'', y'', z'')$, see Fig. 5.3. The second rotation is performed about the new $x''$-axis, resulting in the $(x', y', z')$ axes system. The third and final rotation is about the new $z''$-axis, resulting in the crystal axes system $(x', y', z')$.

To incorporate the Euler angles in the finite element code, they need to be captured in a rotation tensor $R$. Each consecutive rotation maps an orthonormal coordinate system onto another one and can be captured by a rotation tensor. These three rotation tensors, $R_{\phi_1}$, $R_{\theta}$, and $R_{\phi_2}$, can be combined to one tensor which transforms a vector between the crystal axes system and the laboratory axes system:

$$R = R_{\phi_2} \cdot R_{\theta} \cdot R_{\phi_1}.$$  (5.20)
In matrix notation the rotation tensors $R_{\phi_1}$, $R_{\theta}$, and $R_{\phi_2}$ are respectively,

$$
\begin{bmatrix}
\cos(\phi_1) & \sin(\phi_1) & 0 \\
-sin(\phi_1) & \cos(\phi_1) & 0 \\
0 & 0 & 1
\end{bmatrix}, \quad
\begin{bmatrix}
0 & 0 & 1 \\
0 & \cos(\theta) & \sin(\theta) \\
0 & -\sin(\theta) & \cos(\theta)
\end{bmatrix}, \quad
\begin{bmatrix}
\cos(\phi_2) & \sin(\phi_2) & 0 \\
-sin(\phi_2) & \cos(\phi_2) & 0 \\
0 & 0 & 1
\end{bmatrix}.
$$

(5.21)

The rotation tensor $R$ is inserted in the material model to account for the crystal orientation. It maps the orthonormal base vectors of the global axes ($\vec{e}_i$) to a set of orthonormal base vectors linked to the crystallographic axes ($\vec{e'}_i$),

$$
R = \vec{e'}_i \cdot \vec{e}_i^\prime = \vec{e'}_i \cdot \vec{e}_i^\prime \cdot \vec{e}_i^\prime \cdot \vec{e}_i
$$

(5.22)

The anisotropic material properties, the coefficients of thermal expansion and the elastic properties, which are contained within respectively a second-order ($\alpha$) and a fourth order tensor ($H$), need to be transformed accordingly,

$$
\alpha = R \cdot \alpha' \cdot R,
$$

(5.23)

and

$$
H = H_{ijkl} = \left[ R \cdot \left( R \cdot \left( \frac{1}{1-\omega} \cdot R \right) \right) \right]_{lc,rc}.
$$

(5.24)

### 5.3.6 Incremental description

The complete elasto-viscoplastic model in its rate form is now determined by Eqs. (5.13), (5.14), (5.16) and (5.19). Since this system can not be integrated analytically, an incremental time integration scheme will be used. Using a backward Euler discretisation scheme,

$$
a = a^t + \Delta t = a^1 + \left. \frac{da}{dt} \right|_{t+\Delta t} \Delta t,
$$

applied to the viscoplastic multiplier gives

$$
\Delta \lambda = \lambda - \lambda^t = \Delta t \eta \left( \frac{\tau_{eq} - (1-\omega)\tau_y}{(1-\omega)\phi_0} \right)^N.
$$

(5.25)

The constitutive equation for the stress first needs to be formulated in terms of invariant variables, which remain unaltered under superposed spatial rigid body motions [e.g. Marsden and Hughes, 1983], thereby preserving the principle of objectivity. Performing the time discretisation then gives,

$$
\tau = \left( \frac{1-\omega}{1-\omega^t} \right) \left[ F_\Delta \cdot \tau^t \cdot F_\Delta^c + (1-\omega) H : (e_\Delta - \Delta \lambda N - \alpha \Delta t) \right]
$$

(5.26)

with $F_\Delta = F \cdot [F^t]^{-1}$ the incremental deformation gradient tensor and $e_\Delta$ the incremental Almansi strain tensor

$$
e_\Delta = \frac{1}{2} \left( I - F_\Delta^{-c} \cdot F_\Delta^{-1} \right).
$$

(5.27)
### Table 5.1 — Elastic constants [Mason and Bömmel, 1956] and coefficients of thermal expansion of BCT Sn [Pearson, 1958].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
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<tr>
<td>Elastic constants</td>
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<tr>
<td></td>
<td>$H_{111}$</td>
<td>[GPa]</td>
<td>73.5</td>
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<tr>
<td></td>
<td>$H_{333}$</td>
<td>[GPa]</td>
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</tr>
<tr>
<td></td>
<td>$H_{232}$</td>
<td>[GPa]</td>
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</tr>
<tr>
<td></td>
<td>$H_{121}$</td>
<td>[GPa]</td>
<td>22.65</td>
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<tr>
<td></td>
<td>$H_{112}$</td>
<td>[GPa]</td>
<td>23.4</td>
</tr>
<tr>
<td></td>
<td>$H_{113}$</td>
<td>[GPa]</td>
<td>28.0</td>
</tr>
<tr>
<td>Coefficients of thermal expansion</td>
<td>$a_{11}(30\degree C)=a_{22}(30\degree C)$</td>
<td>[K$^{-1}$]</td>
<td>$16.5 \cdot 10^{-6}$</td>
</tr>
<tr>
<td></td>
<td>$a_{33}(30\degree C)$</td>
<td>[K$^{-1}$]</td>
<td>$32.4 \cdot 10^{-6}$</td>
</tr>
<tr>
<td></td>
<td>$a_{11}(130\degree C)=a_{22}(130\degree C)$</td>
<td>[K$^{-1}$]</td>
<td>$20.2 \cdot 10^{-6}$</td>
</tr>
<tr>
<td></td>
<td>$a_{33}(130\degree C)$</td>
<td>[K$^{-1}$]</td>
<td>$41.2 \cdot 10^{-6}$</td>
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</tbody>
</table>

### 5.4 Simulations

#### 5.4.1 Parameters

The material under investigation is a near-eutectic Sn–Ag–Cu alloy. Since the material consists mostly out of $\beta$-Sn grains, it is assumed that the elastic properties are dominated by those of pure Sn. As discussed in section 5.2, due to symmetry, only 6 parameters are needed to describe the anisotropic elastic behaviour of Sn. Thermal anisotropy is caused by the thermal expansion coefficients, which in the short BCT axis deviates from the long BCT axes by almost a factor of two. The CTEs are assumed to vary linearly with temperature for the range used in the experiment. The elastic and thermal parameters are summarised in Table 5.4.1 (with two temperatures for the CTEs).

The viscoplastic properties are taken isotropic. The plastic properties of a Sn based solder are heavily affected by the addition of alloying metals, especially around the grain boundaries. These locations are also the typical spots where the Cu$_6$Sn$_5$ and Ag$_3$Sn intermetallics can be found in the SAC solder. Therefore, these parameters are found by fitting to material data on SAC found in literature [Amagai et al., 2002] instead of assuming them to be equal to those of pure Sn. The hardening law that has been used reads

$$\tau_y = \tau_{y0} + h_1 \left(1 - \exp[-h_2\varepsilon_{vp}]\right) + h_3\varepsilon_{vp}. \quad (5.28)$$

The plastic work has been selected as the source term of Eq.(5.16) which determines the nonlocal damage driving variable. The simulation of the thermal cycling process using viscoplastic material behaviour, but excluding damage, revealed a steady state distribution of the field variables to arise after only a limited number of cycles. After this situation has been reached the values of the stresses, viscoplastic strains, and plastic work increase proportionally in the specimen with additional cycles. To describe damage evolution in a qualitative way, it does therefore not matter when damage initiates, as long as it does so after the steady state distributions have been reached. The occurrence of damage in particular locations of the specimen can cause the neighbouring material to unload by allowing a redistribution of the deformation. To adequately capture this phenomenon qualitatively, the rate at which damage develops in the simulations should not be too fast.
The choice of the length parameter is based on the distance between defects found in the specimen after thermal cycling, see Fig. 5.4.

The viscoplastic and damage parameters used for the simulations are listed in Table 5.2

### 5.4.2 Fully elastic simulation

First, a simulation of thermal loading of the sample was performed where the material is assumed to be fully elastic. In this case, cycling will have no influence on the stress and strain distributions. During the simulation, the temperature of the sample is increased homogeneously from 20 to 80°C. A finite element mesh of the complete sample is created, whereby the data obtained from the OIM in the vicinity of grain boundaries is used to refine the mesh locally. All the grain boundaries are assumed to be perpendicular to the surface. The crystal orientation is accounted for by assigning the Euler angles from the OIM measurements to the integration points.

This simulation differs from the one published by Matin et al. [2005] in that different boundary conditions are used, which will be discussed next, and the temperature dependency of the CTEs is accounted for.

No mechanical constraints are applied to the specimen, only rigid body motions are constraint. Another possibility for the boundary conditions would be to constrain all the

<table>
<thead>
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<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
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<tr>
<td>initial yield stress</td>
<td>( \tau_{y0} )</td>
<td>[MPa]</td>
<td>8</td>
</tr>
<tr>
<td>fluidity</td>
<td>( \eta )</td>
<td>[1/s]</td>
<td>( 2 \times 10^{-4} )</td>
</tr>
<tr>
<td>rate sensitivity</td>
<td>( N )</td>
<td>[-]</td>
<td>4</td>
</tr>
<tr>
<td>hardening parameters</td>
<td>( h_1 )</td>
<td>[MPa]</td>
<td>12</td>
</tr>
<tr>
<td></td>
<td>( h_2 )</td>
<td>[-]</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>( h_3 )</td>
<td>[MPa]</td>
<td>20</td>
</tr>
<tr>
<td>damage rate</td>
<td>( b )</td>
<td>[-]</td>
<td>( 10 \times 10^6 )</td>
</tr>
<tr>
<td>damage initialisation</td>
<td>( Z_i )</td>
<td>[MJ]</td>
<td>0.04</td>
</tr>
<tr>
<td>length parameter</td>
<td>( \ell_0 )</td>
<td>[nm]</td>
<td>200</td>
</tr>
</tbody>
</table>
bottom nodes in the $z$-direction. Obviously, this would introduce additional stresses in the specimen, as the boundary condition would constrain the grains. Although, the grains modelled are indeed connected to underlying grains, there is no experimental information on the depth at which the associated grain boundaries are located. Furthermore, the stress concentrations resulting from an interface are restricted to its vicinity, i.e. if the grains observed in the $xy$-plane are large enough in the $z$ direction, the stresses resulting from the interface would not have a significant contribution at the surface. No information is available on the orientation of the grain boundaries in the out of plane direction ($z$-axis), therefore they are assumed to be perpendicular to the $xy$-plane.

The resulting stress field at $80^\circ$C is depicted in Fig. 5.5. Stress concentrations arise at the grain boundaries, but indeed there is no direct relation between the magnitude of the misorientation angle and the stress. Maximum stresses are found at the grain boundaries $g_1/g_2$ and $g_7/g_{12}$ where the misorientation angle is about $90^\circ$, but at boundaries $g_2/g_3$ and $g_1/g_8$ with a similar misorientation angle, the stresses do not reach the same values. In the next section the grain boundary orientation will be shown to have a significant impact on the resulting stress concentrations, which explains the lack of correlation between the magnitude of the misorientation angle and the magnitude of the stress concentration.

However, there is a good correlation between grain boundaries where stress concentrations appear in the simulation and those where damage is observed experimentally, e.g. around $g_{12}$, $g_1/g_5$, $g_{11}/g_{13}$, and $g_7/g_{16}$. Also, grain boundaries where stress concentrations are absent did not show damage experimentally, e.g. $g_6/g_7$. However, stress concentrations also arise at places where no damage is visible, e.g. inside $g_{11}$ and in the vicinity of $g_8$.

### 5.4.3 Misorientation angle versus stress concentrations

The absence of a proportional relation between the mismatch angle and the magnitude of the stress concentrations at the grain boundaries is further investigated in this section. The samples used for this theoretical analysis are rectangular volumes with a thickness of 400 µm, and a width and length of 800 µm. They are thermally loaded by raising the temperature homogeneously from $20^\circ$C to $80^\circ$C, without any mechanical constraints, i.e. they are free to expand in all directions.

![Figure 5.5 — Kirchhoff stresses [MPa] after raising the temperature from 20°C to 80°C. Fully elastic material behaviour.](image-url)
Fig. 5.6 shows that the magnitude of the stress concentrations arising at an interface is not uniquely determined by the misorientation angle between the grains. For these calculations the misorientation angle between the c-axes of the crystals is 90° and the grain boundary is vertical, located in the middle of the sample. After increasing the temperature stress concentrations arise. However, for different combinations of Euler angles different stress fields are obtained, ranging from a maximum stress concentration for the set (0,0,0)-(0,90,0) to a completely stress-free sample for the set (90,−45,0)-(90,45,0). Rather than depending on the mismatch angle, the stress concentrations depend on the difference between the projections of the [001]-axes of the two neighbouring crystals on their mutual grain boundary. This can be quantified by introducing the mismatch factor,

\[ M = \| (\vec{c}_1 - \vec{c}_2) \cdot (I - \vec{\hat{n}}\vec{\hat{n}}) \|, \] (5.29)

where \( \vec{c}_i \) are the [001]-axes of the two crystals and \( \vec{\hat{n}} \) the normal to the grain boundary plain. Although for the set (90,−45,0)-(90,45,0) the mismatch angle is 90°, \( M \) is zero, and no stresses arise. Maximum stress concentrations are found for the set (0,0,0)-(0,90,0), which correlates with the maximum difference between the projections, \( M = \sqrt{2} \approx 1.4142 \).

Changing the orientation of the grain boundary to an angle of 45° with the vertical direction,

**Figure 5.7** — Influence of the grain boundary orientation on the equivalent Kirchhoff stresses in [MPa] caused by thermal expansion (sideview). Grain boundary is oriented at 45° with the vertical. (Deformations are magnified).
see Fig. 5.7, shows that maximum stress concentrations are no longer found for the set 
(0,0,0)-(0,90,0) \( (M = 1.2247) \), but instead for the Euler angle set \((0,0,0)\)\(-(-45,90,0)\) \( (M = 1.4142) \).

From the above it is clear that the orientation of the grain boundaries has a significant 
influence on the stress concentrations that develop. The assumption that all the grain 
boundaries are perpendicular to the \(xy\)-plane, therefore, introduces a source of uncertainty 
on the predicted stress fields (the true 3D normals of the grain boundaries and their spatial 
variation are not known).

### 5.4.4 Elasto-viscoplastic analysis

The thermal cycling experiment was next simulated using the elasto-viscoplastic material 
model. The same boundary conditions were used as for the elastic analysis. No damage 
was allowed to develop in this stage.

Fig. 5.8 shows the resulting equivalent stress field during the 8th thermal cycle. The 
viscoplastic behaviour causes residual stresses to be present in the material at the begin-
ing of the cycle. Heating the specimen causes the stresses to increase, but their 
distribution remains qualitatively the same. During heating, the yield stress is exceeded 
at the grain boundaries where stress concentrations occur. Here the material deforms 
plastically and the yield stress increases locally through hardening. Subsequently holding 
the specimen at the high temperature causes viscoplastic deformation, accompanied by a 
gradual relaxation of the stresses. During this creep stage, the material still undergoes local 
plastic deformation until the stress drops below the yield stress. Due to the relaxation of the 
stresses at the high holding temperature, cooling the specimen back to room temperature 
causes the stresses to increase again towards comparable values as after the heating stage, 
after which they relax again during the hold period similarly to the holding stage at the high 
temperature. Between the stress state before and after the cycle, a small increase in the 
residual stresses occurs, accompanied with an increase in both the effective plastic strain 
and plastic work. In Fig. 5.9, the accumulated effective viscoplastic strain and the plastic 
work done after 8 cycles is displayed. The spatial distribution of these field variables does 
do not change quantitatively with ongoing cycling, their values only increase.

Compared to the stresses obtained for the fully elastic material, the stresses for the 
viscoplastic material behaviour are much lower. The distribution of the stress field is not 
influenced significantly by the viscoplastic behaviour, except for the fact that cooling does 
not lead to a stress-free configuration, but residual stresses remain present in the material.

### 5.4.5 Damage analysis

Finally, a calculation including the damage analysis has been performed using the same 
boundary and loading conditions as before. The results are displayed in Fig. 5.10. The 
distribution of the damage correlates well with the distribution fields of the other field 
variables obtained for the purely elastic case and the elasto-viscoplastic case without 
damage. Areas where stress and viscoplastic strain concentrations are predicted clearly 
display the highest damage values.
Viscoplastic nonlocal damage modelling of thermal fatigue

Figure 5.8 — Development of the equivalent Kirchhoff stress during a thermal cycle between 20°C and 80°C, elasto-viscoplastic material behaviour.

Figure 5.9 — Results of thermal cycling between 20°C and 80°C after 8 cycles, elasto-viscoplastic material behaviour.

(a) Effective viscoplastic strain [-].
(b) Plastic work [MJ/m^3].
The regions were damage is predicted numerically and where it actually occurred experimentally correspond well, supporting the theory that the anisotropy of Sn contributes to cyclic fatigue damage in SAC solder.

The only major difference is visible around grain $g_4$, where the damage is predicted numerically at the grain boundary with $g_2$, while the experimental results indicate damage at the boundary with $g_3$. This discrepancy can easily be explained by the lack of information available on the shape of the grains in the third direction and consequently the differences in stress concentrations associated to this (see section 5.4.3). Especially, since the 3D misorientation angles of the two grain boundaries in question are almost equal, the orientation of the grain boundaries will determine where the highest stress concentrations occur.

The local differences between the numerical results and the experimental observations have a number of potential origins. First, as shown in section 5.4.3, the orientation of
the grain boundaries in the \( z \) direction can have a significant influence on the stress concentrations that appear at the boundary. Second, the used boundary conditions assume that the grains visible in the experimentally analysed \( xy \)-plane traverse the entire height of the specimen. This is however not the case; the height of the specimen is larger than the average grain diameter observed in a sectioning plane, see Fig. 5.1a, indicating that multiple grains can be found in the \( z \) direction. Third, the material is modelled as a single phase, filtering out the contribution of the \( \text{Cu}_6\text{Sn}_5 \) and \( \text{Ag}_3\text{Sn} \) intermetallics, which might influence the resulting stress fields.

5.5 Conclusions

A three-dimensional elasto-viscoplastic damage model has been used to investigate the possible influence of the mechanical and thermal anisotropy of Sn on fatigue damage. The elastic constants and the thermal expansion coefficients were taken anisotropic, whereas the viscoplastic and damage behaviour were considered isotropic. A gradient-enhanced continuum damage description was used, which properly regularises the solution and avoids mesh dependency.

Experimental observations [Matin et al., 2005] indicated damage to appear in the vicinity of grain boundaries during unconstraint thermal cycling of commercial SAC solder between 20\(^\circ\)C and 80\(^\circ\)C. OIM information on the examined sample was used as the geometrical input for the adopted finite element model, thereby incorporating the orientations of the individual grains.

As opposed to the elastic analysis presented in Matin et al. [2005] here also the time-dependent material behaviour and the influence of damage have been accounted for.

Also, the employed boundary conditions have been reevaluated.

The simulations showed stress and viscoplastic strain concentrations to arise at grain boundaries, dependent on the misorientation between the grains and their orientation with respect to the grain boundary.

A consistent agreement was found between the numerically obtained damage distribution field and the damaged areas identified experimentally. Differences between the simulations and the experimental results could be explained by the missing data on the 3D grain boundary orientation through the sample thickness. It can be concluded that the anisotropy indeed plays an important part in determining the thermal fatigue life of SAC solder.
The objective of this thesis has been the investigation of thermomechanical fatigue of soldered connections. Initially the project was started with the eutectic Sn–Pb alloy in mind, but due to the recent push towards Pb–free solders, the work was concluded by an analysis on one of the most promising alloys, i.e. a near-eutectic Sn–Ag–Cu alloy.

For the Sn–Pb solder alloy fatigue damage is closely linked to the microstructure and its evolution. In Ubachs et al. [2004] (chapter 2), an extension of the Cahn–Hilliard model is presented to describe the microstructure evolution through diffusion. The driving force of this diffusion is found in the spatial gradients of the free energy, which consists of a configurational energy, elastically stored energy, and interfacial energy. The latter is accounted for by introducing a strongly nonlocal parameter, i.e. the weighted average of the mass fraction over a finite domain. The model is implemented in a two-dimensional finite element framework, which is used to perform a simulation of a static thermal ageing experiment.

With respect to the resulting microstructure evolution model the following conclusions are drawn:

- In the classical Cahn–Hilliard theory, the interfacial energy is dependent on spatial derivatives which are per definition confined to an infinitesimal neighbourhood, whereas here a strongly nonlocal measure is used.

- By introducing the strongly nonlocal measure in the free energy, this particular implementation has the advantage that higher level derivatives are absent in the formulation, facilitating the numerical solution.

- The nonlocal extension introduces an additional parameter affecting the interfaces in the model, compared with the Cahn–Hilliard theory. The two parameters which influence the interface, the internal length scale \( \ell_c \) and the interface tension coefficient \( \kappa \), can be used to separately define the interfacial width and the area of influence of an interface. These distinct features, of which the relevance is illustrated, cannot be obtained with a classical Cahn–Hilliard theory.

- Elastically stored energy causes a shift of the mass fraction equilibrium values and
decreases the interfacial width. Furthermore, it increases the decomposition rate and the coarsening rate.

- Qualitatively, the model is able to predict break-up, coalescence, growth and dissolution of phase regions, similar to what is observed experimentally.
- The total surface length per unit area is used to quantitatively compare the predicted microstructure evolution with the experimental results. An adequate agreement was found.

The next two chapters [Ubachs et al., 2005a,b] deal with the influence of the microstructure on the material response during mechanical cycling.

In Ubachs et al. [2005a], chapter 3, an elasto-viscoplastic material model has been used to describe the time dependent deformation mechanisms associated in metals operating at high homologous temperatures such as solders. Through the use of a phase field model, microstructural information has been incorporated in the formulation. Material parameters are assigned to the individual phases by means of the local mass fraction values. In this way the microstructure is accounted for using a continuum mechanics approach.

A two-dimensional finite element implementation of the model has been employed to simulate mechanical cycling tests of Sn-Pb solder. Calculations were performed on different microstructures, which were obtained through a static thermal ageing simulation employing the phase field model. The main conclusions related to this part are:

- The approach does not require interface tracking methods since all information about the interface is included in the phase field. Furthermore, the evolution of a microstructure can be calculated using a single mesh, no remeshing needs to be done.
- Viscoplastic deformation is mostly restricted to the Pb-rich phase, due to its lower yield stress. The highest stresses are found in the Sn-rich phase.
- Within the phases, the field variables, stresses and strain, are more homogeneously for finer microstructures. A coarser microstructure leads to more pronounced localisation behaviour.
- Locally, stresses and strain reach higher values for a coarse microstructure compared to a fine microstructure. In both cases the local values exceed the macroscopic values significantly.
- Due to heterogeneous plastic straining, residual stresses appear in the material after unloading.

In Ubachs et al. [2005b] the material model is extended to include damage initiation and evolution. To restore the well-posedness of the governing partial differential equations, a gradient-enhanced continuum damage model has been implemented. Microstructural dependency is here also included by means of the phase field model. Different damage parameters are assigned to the individual phases. Additionally, a penalty parameter is introduced to account for interfacial damage, the existence of which was emphasized by experimental observations published in literature. The plastic work was chosen as the damage driving variable. The main conclusions are:
As is the case for the stresses and strains, the damage parameter values are more homogeneously distributed for a fine microstructure compared to a coarse one.

A fine microstructure exhibits a larger area of low damage during mechanical cycling compared to a coarse microstructure, which has relatively a larger area that is critically damaged.

After a sufficient number of cycles, the stress decreases through microstructural failure of the specimen, which occurs earlier for a coarse microstructure than for a fine microstructure. This is attributed to the preferential damage path that is obstructed more in a fine microstructure than in a coarse one.

The presented model predicts microstructural dependent damage evolution in eutectic Sn–Pb, indicating coarser microstructures to be more prone to damage than finer ones. Qualitative comparison with experimental observations in the literature shows an adequate agreement.

In the last paper, Ubachs et al. [2005c], the elasto-viscoplastic nonlocal damage model is used to investigate the influence of anisotropy on the fatigue behaviour of a Sn–Ag–Cu solder. To this end, the anisotropic elastic properties and the anisotropic thermal expansion coefficients are accounted for in a three-dimensional FEM implementation of the model. The viscoplastic and damage properties are considered to be isotropic.

A simulation of a thermal cycling experiment of a Sn–Ag–Cu specimen has been performed and a qualitative analysis was made. Data regarding the crystal orientation was obtained by orientation imaging microscopy and used as input for the calculations, resulting in the following conclusions:

Experimental observations indicated defects to appear in the vicinity of grain boundaries during unconstraint thermal cycling of commercial SAC solder between 20°C and 80°C.

A fully elastic analysis showed stress concentrations appearing at grain boundaries. The magnitude of these stresses is shown to depend on the misorientation angle between the grains and their orientation with respect to the grain boundary.

A very good agreement was found between the numerically obtained damage distribution field and the damaged areas identified experimentally.

A more accurate prediction of damage initiation and propagation sites requires three-dimensional experimental data on the grain structure of the sample, in particular the out-of-plane geometry of the grain boundaries.

The anisotropic properties of Sn–Ag–Cu solder have an important contribution to the thermal fatigue life of SAC solder.

The small size at which solder joints are being produced, and the high temperatures at which they operate today, introduces new technological challenges. Joint dimensions are approaching typical microstructural sizes, causing the microstructure to substantially influence the properties of a connection. A phase field approach has been introduced that
provides a powerful means of tracking microstructural changes, while its coupling to a
time-dependent nonlocal damage material model efficiently incorporates microstructure
dependence in the description of the mechanical response of the material. Furthermore,
investigation of the influence of grain orientation within a solder on thermal fatigue
properties indicated the importance of including detailed microstructural information in
the analysis. The work presented has shown that numerical tools are most helpful in gaining
an understanding of the material behaviour at small scales.
Appendices
A. Strongly nonlocal parameter

The nonlocal mass fraction will be defined as

$$\tilde{c}(\bar{x}) = \frac{1}{\kappa} \int_\Omega g'(\bar{y}) \ c(\bar{x} + \bar{y}) \ d\bar{y}$$  \hspace{1cm} (A.1)

where $\Omega$ is the region around the material point with position vector $\bar{x}$, $\bar{y}$ a local coordinate vector with respect to $\bar{x}$ pointing to points in $\Omega$, and $g'(\bar{y})$ a normalised weighting function which has to satisfy

$$\int_\Omega g'(\bar{y}) \ d\bar{y} = 1$$  \hspace{1cm} (A.2)

This integral formulation can also be written in a differential form [Peerlings et al., 2001]. To this purpose the local mass fraction at $\bar{x} + \bar{y}$ is expanded in a Taylor series around $\bar{x}$

$$c(\bar{x} + \bar{y}) = c(\bar{x}) + \nabla c(\bar{x}) \cdot \bar{y} + \frac{1}{2!} \nabla^2 c(\bar{x}) : \bar{y} \bar{y}$$  \hspace{1cm} (A.3)

$$+ \frac{1}{3!} \nabla^3 c(\bar{x}) : \bar{y} \bar{y} \bar{y} + \frac{1}{4!} \nabla^4 c(\bar{x}) : \bar{y} \bar{y} \bar{y} \bar{y} + \ldots$$  \hspace{1cm} (A.4)

Substituting this into the integral definition of the nonlocal mass fraction, equation (A.1), gives

$$\tilde{c}(\bar{x}) = \int_\Omega g'(\bar{y}) \ c(\bar{x}) \ d\bar{y} + \int_\Omega g'(\bar{y}) \ \nabla c(\bar{x}) \cdot \bar{y} \ d\bar{y} + \frac{1}{2} \int_\Omega g'(\bar{y}) \ \nabla^2 c(\bar{x}) : \bar{y} \bar{y} \ d\bar{y}$$

$$+ \frac{1}{3!} \int_\Omega g'(\bar{y}) \ \nabla^3 c(\bar{x}) : \bar{y} \bar{y} \bar{y} \ d\bar{y} + \frac{1}{4!} \int_\Omega g'(\bar{y}) \ \nabla^4 c(\bar{x}) : \bar{y} \bar{y} \bar{y} \bar{y} \ d\bar{y} + \ldots$$  \hspace{1cm} (A.5

If an isotropic weighting function is used, which is only dependent on a radial coordinate $r$, odd derivatives vanish upon integration, whereas terms involving $\nabla \nabla$ condensate to a single term involving $\nabla^4$, and its derivatives. Substituting

$$\tilde{c}(\bar{x}) = c(\bar{x}) + \ell^2_c \nabla^2 c(\bar{x}) + k^4 \nabla^4 c(\bar{x}) + \ldots$$  \hspace{1cm} (A.7)

with

$$\ell^2_c = \frac{1}{2} \int_\Omega r^2 g'(r) \ dr \quad \text{and} \quad k^4 = \frac{1}{24} \int_\Omega r^4 g'(r) \ dr$$  \hspace{1cm} (A.8)

If only second-order gradient terms are retained, equation (A.7) yields an explicit approximation of integral (A.1) since $\tilde{c}$ is given explicitly in terms of $c$ and its derivatives. Substituting this into the diffusion equation (2.27a) yields

$$\rho \frac{dc}{dt} = \nabla \cdot \left[ \rho \ M \cdot \nabla \left( \frac{\partial \Phi_0}{\partial c} - \kappa \ell^2_c \nabla^2 c + \psi \right) \right]$$  \hspace{1cm} (A.9)

which is identical to the Cahn–Hilliard theory with $\gamma_{CH} = \kappa \ell^2_c$.

A strongly nonlocal formulation can be found by applying the Laplacian operator to equation (A.7) and multiplying it with $\ell^2_c$. Subtracting this from (A.7) gives the following relation:

$$\tilde{c}(\bar{x}) - \ell^2_c \nabla^2 \tilde{c}(\bar{x}) = c(\bar{x}) + (k^4 - \ell^2_c) \nabla^4 c(\bar{x}) + \ldots$$  \hspace{1cm} (A.10)
Derivatives of fourth-order and higher now vanish naturally through the use of a specific weighting function $g^*(\mathbf{y})$, which leads to the following exact reformulation of equation (A.1):

$$\bar{c} - \ell_c^2 \nabla^2 \bar{c} = c$$  \hspace{1cm} (A.11)

The weighting function that enforces the cancelling out of all higher-order terms in equation (A.10) ($k^4 = \ell_c^2$) is in fact the Green’s function associated to equation (A.11) [Myint-U and Debnath, 1987]. See Peerlings et al. [2001] for more details on this particular strongly nonlocal reformulation.

The nonlocal mass fraction $\bar{c}$ thus is the solution of a boundary value problem consisting of the Helmholtz equation (A.11) and appropriate boundary conditions. The resulting gradient formulation is sometimes referred to as implicit. The equation also gives rise to an additional parameter in the system: the internal length parameter $\ell_c$. It is a measure for the size of the region around a material point (in this case the interface) that influences the associated nonlocal variable and in this case the resulting spatial distribution of the surface energy. For the limit case $\ell_c \rightarrow 0$ the nonlocal field $\bar{c}(\mathbf{x})$ equals the local field $c(\mathbf{x})$ and the classical Cahn–Hilliard equations will be obtained again.
B. Details on the finite element implementation of the elasto-viscoplastic model

B.1 Linearisation of the stress for an elastic increment

For a fully elastic increment, i.e. \( \varphi < 0 \), the incremental update for the Kirchhoff stress tensor reads:

\[
\tau = F_0 \cdot \tau' \cdot F_0^c + \mathbb{H}(J) : e_0
\]  

(B.12)

The variation of the stress then becomes:

\[
\delta \tau = \delta F_0 \cdot \tau' \cdot F_0^c + F_0 \cdot \tau' \cdot \delta F_0^c + \frac{\partial \mathbb{H}}{\partial J} : e_0 \delta J + \mathbb{H}(J) : \delta e_0
\]  

(B.13a)

\[
\begin{align*}
\delta \tau &= \delta F_0 \cdot F_0^{-1} \cdot F_0^c \cdot \tau' \cdot F_0^c + F_0 \cdot \tau' \cdot F_0^c \cdot F_0^{-c} \cdot \delta F_0^c \\
&\quad + J \frac{\partial \mathbb{H}}{\partial J} : e_0 F^{-1} : \delta F - \mathbb{H} : \frac{1}{2} (\delta F_0^{-c} \cdot F_0^{-1} + F_0^{-c} \cdot \delta F_0^{-1})
\end{align*}
\]  

(B.13b)

\[
\begin{align*}
\delta \tau &= L_0^c \cdot F_0 \cdot \tau' \cdot F_0^c + F_0 \cdot \tau' \cdot F_0^c \cdot L_0^c + \left[ J \frac{\partial \mathbb{H}}{\partial J} : e_0 I \right] : L_0^c \\
&\quad + \mathbb{H} : \frac{1}{2} \left[ F_0^{-c} \cdot \delta F_0^c \cdot F_0^{-c} \cdot F_0^{-1} + F_0^{-c} \cdot F_0^{-1} \cdot \delta F_0 \cdot F_0^{-1} \right]
\end{align*}
\]  

(B.13c)

\[
\begin{align*}
\delta \tau &= \left[ 2I^S \cdot F_0 \cdot \tau' \cdot F_0^c \right] : L_0^c + \left[ J \frac{\partial \mathbb{H}}{\partial J} : e_0 I \right] : L_0^c \\
&\quad + \mathbb{H} : \frac{1}{2} \left[ L_0^c \cdot F_0^{-c} \cdot F_0^{-1} + F_0^{-c} \cdot F_0^{-1} \cdot L_0 \right]
\end{align*}
\]  

(B.13d)

\[
\begin{align*}
\delta \tau &= \left[ 2I^S \cdot F_0 \cdot \tau' \cdot F_0^c + J \frac{\partial \mathbb{H}}{\partial J} : e_0 I + \mathbb{H} \cdot \left( [I^S \cdot F_0^{-c} \cdot F_0^{-1}] \right) \right] : L_0^c
\end{align*}
\]  

(B.13e)

\[
\delta \tau = C : L_0^c
\]  

(B.13f)

with

\[
C = 2I^S \cdot F_0 \cdot \tau' \cdot F_0^c + J \frac{\partial \mathbb{H}}{\partial J} : e_0 I + \mathbb{H} \cdot \left( [I^S \cdot F_0^{-c} \cdot F_0^{-1}] \right)
\]  

(B.13g)

where use has been made of

\[
\delta \det(A) = \frac{\partial \det(A)}{\partial A} : \delta A = \det(A) A^{-1} : \delta A
\]  

(B.14)

The derivative of the material tensor \( \mathbb{H} \), Eq.(3.27), with respect to \( J \) reads:

\[
\frac{\partial \mathbb{H}}{\partial J} = -2K \frac{\partial \ln(J)}{\partial J} \mathbb{I}^c = -2K \frac{1}{J} \mathbb{I}^c
\]  

(B.15)
B.2 Linearisation of the stress for an elasto-viscoplastic increment

The variation of the stress tensor in the case of elasto-viscoplastic behaviour, Eq.(3.43a), reads

\[ \delta \tau = \delta^* \tau - \delta \mathbb{H} : \Delta \lambda N - \mathbb{H} : \delta \Delta \lambda N - \mathbb{H} : \Delta \lambda \delta N, \] (B.16)

where \( \delta^* \tau \) is the variation of the Kirchhoff stress tensor in case of purely elastic deformation. Variation of the material tensor \( \mathbb{H} \), Eq.(3.27), the viscoplastic multiplier \( \Delta \lambda \), Eq.(3.43b), and the plastic flow direction \( N \) respectively read

\[ \delta \mathbb{H} : \Delta \lambda N = -2K \frac{1}{J} I I : L^c_{\theta}^0 : \Delta \lambda N = -2K \Delta \lambda N : L^c, \] (B.17)

\[ \delta \Delta \lambda = A \eta \left( \frac{\tau_{eq} - \tau_y}{\varphi_0} \right)^N = N A \eta \left( \frac{\tau_{eq} - \tau_y}{\varphi_0} \right)^{N-1} (\delta \tau_{eq} - \delta \tau_y) \]
\[ = c_1 \left( N : \delta \tau - \frac{\partial \tau_y}{\partial \Delta \lambda} \delta \Delta \lambda \right) = c_2 N : \delta \tau, \] (B.18)

with

\[ c_1 = N A \eta \left( \frac{\tau_{eq} - \tau_y}{\varphi_0} \right)^{N-1} \] and \[ c_2 = \frac{c_1}{1 + \frac{\partial \tau_y}{\partial \epsilon_{vp}} \frac{\partial \epsilon_{vp}}{\partial \Delta \lambda} c_1}, \] (B.19)

and

\[ \delta N = \frac{3}{2} \frac{\tau_{eq}}{\tau_{eq}} \delta \tau^d - \frac{3}{2} \frac{\tau_{eq}}{\tau_{eq}} \delta \tau_{eq} = \frac{3}{2} \frac{\tau_{eq}}{\tau_{eq}} \left( \delta \tau - \frac{1}{3} I I : \delta \tau \right) - \frac{1}{\tau_{eq}} N N : \delta \tau \]
\[ = \frac{1}{\tau_{eq}} \left( \frac{3}{2} - \frac{1}{2} I I - N N \right) : \delta \tau. \] (B.20)

Substituting Eqs.(B.17), (B.18), and (B.20) in Eq.(B.16) then results in

\[ \delta \tau = N^{-1} : (C + T) : L^c, \] (B.21)

with

\[ T = 2 \Delta \lambda K N I, \] (B.22)

and

\[ N = I + c_2 \mathbb{H} : N N + \mathbb{H} : \Delta \lambda \frac{1}{\tau_{eq}} \left( \frac{3}{2} - \frac{1}{2} I I - N N \right). \] (B.23)
B.3 Stress update algorithm

It can be shown that for the current elastic model the direction of plastic flow is fully
determined by the elastic predictor state, \( N = ^*N \). The viscoplastic stress is found by
applying a plastic correction to the elastic predictor,

\[
\tau = ^*\tau - \frac{(1-\omega)^2}{1-\omega} H : \Delta\lambda N
\]  
(B.24)

For this particular case of \( H \) this reduces to,

\[
\tau = ^*\tau - c_3 \Delta\lambda N \quad \text{with} \quad c_3 = \frac{(1-\omega)^2}{1-\omega^2} - 2[G - K\ln(J)] \]  
(B.25)

Collecting terms, using the definition of \( N \) (4.22), and taking the deviatoric part,

\[
\tau^d \left(1 + \frac{3}{2} c_3 \frac{\Delta\lambda}{\tau_{eq}}\right) = ^*\tau^d
\]  
(B.26)

Taking the equivalent value \( (A_{eq} = \sqrt{2/3}A^d : A^d) \) of the tensors on both sides of the equation
then results in,

\[
\tau_{eq} + \frac{3}{2} c_3 \Delta\lambda = ^*\tau_{eq} \quad \text{or} \quad \tau_{eq} = ^*\tau_{eq} - \frac{3}{2} c_3 \Delta\lambda
\]  
(B.27)

From Eqs. (B.26) and (B.27) it follows that,

\[
N = \frac{3/2}{\tau_{eq}} \tau^d = \frac{3/2}{(1 + 3/2 c_3 \frac{\Delta\lambda}{\tau_{eq}})\tau_{eq}} = \frac{3/2}{\tau_{eq} + \frac{3}{2} c_3 \Delta\lambda} = \frac{3/2}{^*\tau_{eq}} = ^*N
\]  
(B.28)

Furthermore, for the stress update algorithm the nodal displacements are known and fixed,
except for the plane stress case. Therefore the volume and the stresses of the elastic
predictor will not change, i.e. \( \delta J \) and \( \delta ^*\tau \) are zero. To use the Newton–Raphson method
system (3.43) is first rewritten into

\[
\tau - ^*\tau + H : N\Delta\lambda = 0,
\]  
(B.29a)

\[
\Delta\lambda - \Delta t \eta \left( \frac{\phi}{\phi_0} \right)^N = 0.
\]  
(B.29b)

Decomposing these equations in an iterative manner results in

\[
\tau + \delta\tau - ^*\tau + H : N(\Delta\lambda + \delta\Delta\lambda) = 0,
\]  
(B.30a)

\[
\Delta\lambda + \delta\Delta\lambda - \Delta t \eta \left( \frac{\phi}{\phi_0} \right)^N - \frac{\Delta t \eta N}{\phi_0} \left( \frac{\phi}{\phi_0} \right)^{N-1} \delta\phi = 0.
\]  
(B.30b)

Substituting the definition of the yield function \( \varphi = \tau_{eq} - \tau_y \) and using the variation of \( \tau_{eq} \),

\[
\delta\tau_{eq} = \frac{1}{2} \left( \frac{3}{2} \tau^d : \tau^d \right)^{-1/2} \left( \frac{3}{2} \tau^d : \delta\tau^d \right) = \frac{3}{2} \frac{1}{\tau_{eq}} \tau^d : \delta\tau - \tau^d : \frac{1}{3} I : \delta\tau
\]  
(B.31)

\[
= \frac{3}{2} \frac{1}{\tau_{eq}} \left( \tau^d - \tau^d : \frac{1}{3} I \right) : \delta\tau = \frac{3}{2} \frac{1}{\tau_{eq}} \delta\tau = N : \delta\tau,
\]  
(B.32)
then results in

\[
\mathbb{I} : \delta \tau + \mathbb{H} : N \delta \Delta \lambda = \tau^* - \tau - \mathbb{H} : N \Delta \lambda, \quad (B.33a)
\]

\[
\delta \Delta \lambda - c_1 (N : \delta \tau - \delta \tau_y) = \Delta t \eta \left( \frac{\varphi}{\varphi_0} \right)^N - \Delta \lambda \quad (B.33b)
\]

with

\[
c_1 = \Delta t \eta N \left( \frac{\varphi}{\varphi_0} \right)^{N-1} \quad (B.34)
\]

Assuming the incremental hardening law to be a function of \( \Delta \lambda \) only, i.e. the yield stress can be a function of the viscoplastic multiplier and/or its rate the system that needs to be solved then becomes

\[
\mathbb{I} : \delta \tau + \mathbb{H} : N \delta \Delta \lambda = \tau^* - \tau - \mathbb{H} : N \Delta \lambda, \quad (B.35a)
\]

\[
- c_1 N : \delta \tau + \left( 1 + c_1 \frac{\partial \tau_y}{\partial \Delta \lambda} \right) \delta \Delta \lambda = \Delta t \eta \left( \frac{\varphi}{\varphi_0} \right)^N - \Delta \lambda. \quad (B.35b)
\]
C. Details on the finite element implementation of the elasto-viscoplastic nonlocal damage model

C.1 Time Discretisation

Since the time discretisation has to be performed using invariant tensors, the starting point is the pull-back of Eq.(4.25),

\[ F^{-1} \cdot \dot{\gamma} \cdot F^{-c} = \dot{S} = 2(1 - \omega) \frac{\partial^2 \dot{U}_e}{\partial C_e^2} : (\dot{C} - \dot{\lambda} F^c \cdot N \cdot F) - \frac{S}{(1 - \omega)} \dot{\omega}. \]  
(C.36)

Applying a backward Euler time integration scheme results in,

\[ S - S^t = S \frac{(\omega - \omega^t)}{(1 - \omega)} + 2(1 - \omega) \frac{\partial^2 \dot{U}_e}{\partial C_e^2} : (C - C^t - 2F^c \cdot \Delta \lambda N \cdot F). \]  
(C.37)

Collecting the deformation gradient tensors,

\[ S = S^t - S \frac{(\omega - \omega^t)}{(1 - \omega)} + 4(1 - \omega) \frac{\partial^2 \dot{U}_e}{\partial C_e^2} \cdot \left\{ F^c \cdot \left[ \frac{1}{2} \left( I - F_{\Delta}^{-c} \cdot F_{\Delta}^{-1} \right) - \Delta \lambda N \right] \cdot F \right\}. \]  
(C.38)

Pushing forward to the current configuration,

\[ F \cdot S \cdot F^c = F_{\Delta} \cdot F^c \cdot S^t \cdot F^c + 4(1 - \omega) \frac{\partial^2 \dot{U}_e}{\partial C_e^2} \cdot \left\{ F^c \cdot \left[ \frac{1}{2} \left( I - F_{\Delta}^{-c} \cdot F_{\Delta}^{-1} \right) - \Delta \lambda N \right] \cdot F \right\}. \]  
(C.39)

Rewriting in terms of the Kirchhoff stress,

\[ \tau = F_{\Delta} \cdot \tau^t \cdot F_{\Delta}^{-c} - \tau \frac{(\omega - \omega^t)}{(1 - \omega)} + 4(1 - \omega) F \cdot \left\{ F \cdot \frac{\partial^2 \dot{U}_e}{\partial C_e^2} \cdot F^c \right\}^{lc,rc} \cdot F^c : (e_{\Delta} - \Delta \lambda N). \]  
(C.40)

Collecting \( \tau \) on the left hand side and using Eq.(4.15),

\[ \tau + \tau \frac{(\omega - \omega^t)}{(1 - \omega)} = F_{\Delta} \cdot \tau^t \cdot F_{\Delta}^{-c} + (1 - \omega) \mathbb{H} : (e_{\Delta} - \Delta \lambda N). \]  
(C.41)

The expression for the Kirchhoff stress finally yields,

\[ \tau = \frac{(1 - \omega)}{(1 - \omega^t)} \left[ F_{\Delta} \cdot \tau^t \cdot F_{\Delta}^{-c} + (1 - \omega) \mathbb{H} : e_{\Delta} \right]. \]  
(C.42)

C.2 Linearisation of the stress for an elastic increment

The Kirchhoff stress in case of a fully elastic material behaviour reads

\[ \tau = * \tau = \frac{1 - \omega}{1 - \omega^t} \left[ F_{\Delta} \cdot \tau^t \cdot F_{\Delta}^{-c} + (1 - \omega) \mathbb{H} : e_{\Delta} \right]. \]  
(C.43)
Variation yields (see appendix B.1):

\[
\delta^*\tau = \frac{-1}{1-\omega} \left[ F_A \cdot \tau^f \cdot F_A^c + (1-\omega)\mathbb{H} : e_A \right] \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z} - \frac{1-\omega}{1-\omega} \mathbb{H} : e_A \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z} + \frac{1-\omega}{1-\omega} \left[ 2 \gamma^S \cdot F_A \cdot \tau^f \cdot F_A^c (1-\omega) \left( -2K^{-c} : e_A I + \mathbb{I} : [\gamma^S \cdot F_A^{-c} \cdot F_A^{-1}] \right) \right] : L_\delta^c,
\]

which can be written as,

\[
\delta^*\tau = K_{uz} \delta \bar{z} + \kappa_{uu} : L_\delta^c,
\]

with

\[
K_{uz}^c = \frac{-1}{1-\omega} \left[ F_A \cdot \tau^f \cdot F_A^c + 2(1-\omega)\mathbb{H} : e_A \right] \frac{\partial \omega}{\partial \bar{z}},
\]

and

\[
\kappa_{uu}^c = \frac{1-\omega}{1-\omega} \left[ 2 \gamma^S \cdot F_A \cdot \tau^f \cdot F_A^c (1-\omega) \left( -2K^{-c} : e_A I + \mathbb{I} : [\gamma^S \cdot F_A^{-c} \cdot F_A^{-1}] \right) \right].
\]

### C.3 Linearisation of the stress for an elasto-viscoplastic increment

**Viscoplastic multiplier**

The incremental viscoplastic multiplier reads

\[
\Delta \lambda = \Delta t \eta \left( \frac{\tau_{eq} - (1-\omega)\tau_y}{(1-\omega)\varphi_0} \right)^N.
\]

Making use of Eq.(B.27) the variation becomes

\[
\delta \Delta \lambda = c_1 \left[ \delta^* \tau_{eq} - \frac{3}{2} \Delta \lambda \delta c_3 - \frac{3}{2} c_3 \delta \Delta \lambda + \tau_y \delta \omega - (1-\omega) \delta \tau_y + \frac{\varphi}{1-\omega} \delta \omega \right],
\]

with

\[
c_1 = \Delta t \frac{\eta N}{(1-\omega)\varphi_0} \left( \frac{\varphi}{(1-\omega)\varphi_0} \right)^{N-1}.
\]

Expanding and using Eq.(C.45) gives

\[
c_2 \delta \Delta \lambda = c_1 \left[ N : (\kappa_{uu}^c : L_\delta^c + K_{uz}^c \delta \bar{z}) + 3 \Delta \lambda \frac{(1-\omega)^2}{1-\omega} - K I : L_\delta^c \right.
\]

\[
+ 6 \Delta \lambda \frac{1-\omega}{1-\omega} \left[ G - K \ln(J) \right] \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z} + \tau_y \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z} + \frac{\varphi}{1-\omega} \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z} \bigg],
\]

with

\[
c_2 = 1 + \frac{3}{2} c_1 c_3 + c_1 (1-\omega) \frac{\partial \tau_y}{\partial \lambda}.
\]
Collecting terms,
\[
\delta \Delta \lambda = \frac{c_1}{c_2} \left[ N : \mathbf{K}_{uu}^e + 3 \Delta \lambda \frac{(1 - \omega) - KI}{1 - \omega^t} \right] : L_\delta^e \\
+ \frac{c_1}{c_2} \left[ N : \mathbf{K}_{uz}^e + \frac{\tau_{eq} - \frac{3}{2} c_3 \Delta \lambda}{1 - \omega} \frac{\partial \omega}{\partial \bar{z}} + 3 \Delta \lambda \frac{c_3}{1 - \omega} \frac{\partial \omega}{\partial \bar{z}} \right] \delta \bar{z}
\]
(C.54)

Finally this results in
\[
\delta \Delta \lambda = \mathbf{K}_{\gamma u} : L_\delta^e + \mathbf{K}_{\gamma z} \delta \bar{z}
\]
(C.55)

with
\[
\mathbf{K}_{\gamma u} = \frac{c_1}{c_2} \left[ N : \mathbf{K}_{uu}^e + 3 \Delta \lambda \frac{(1 - \omega) - KI}{1 - \omega^t} \right],
\]
(C.56)

and
\[
\mathbf{K}_{\gamma z} = \frac{c_1}{c_2} \left[ N : \mathbf{K}_{uz}^e + \frac{\tau_{eq} + \frac{3}{2} c_3 \Delta \lambda}{1 - \omega} \frac{\partial \omega}{\partial \bar{z}} \right].
\]
(C.57)

**Flow direction**
\[
\delta N = \frac{3}{2} \frac{1}{\tau_{eq}} \delta \mathbf{\tau}^d - \frac{3}{2} \frac{\tau^d}{\tau_{eq}^2} \delta \tau_{eq} = \frac{3}{2} \frac{1}{\tau_{eq}} \left( \delta \mathbf{\tau} - \frac{1}{3} II : \delta \mathbf{\tau} \right) - \frac{1}{\tau_{eq}} NN : \delta \mathbf{\tau}
\]
\[
= \frac{1}{\tau_{eq}} \left( \frac{3}{2} \mathbb{M} - \frac{1}{2} II - NN \right) : \delta \mathbf{\tau} = \frac{1}{\tau_{eq}} \mathbb{M} : \delta \mathbf{\tau}.
\]
(C.58)

with
\[
\mathbb{M} = \frac{3}{2} \mathbb{I} - \frac{1}{2} II - NN.
\]
(C.59)

It can be shown that \( N = \mathbb{M} \) which means that the variation of the flow direction only depends on the elastic predictor state:
\[
\delta N = \frac{1}{\tau_{eq}} \mathbb{M} : \delta \mathbf{\tau}, \quad \text{with} \quad \mathbb{M} = \frac{3}{2} \mathbb{I} - \frac{1}{2} II - \mathbb{N}^* \mathbb{N}.
\]
(C.60)

**Stress**

The stress tensor in the case of elasto-viscoplastic behaviour with nonlocal damage included reads
\[
\mathbf{\tau} = \frac{1 - \omega}{1 - \omega^t} \left[ \mathbf{F}_\Delta : (\mathbf{\tau}^f : (1 - \omega) \mathbb{H} : (\mathbf{e}_\Delta - N \Delta \lambda)) \right].
\]
(C.61)

The variation then becomes
\[
\delta \mathbf{\tau} = \delta \mathbf{\tau} - \frac{(1 - \omega)}{(1 - \omega^t)} \left( \frac{\partial \mathbb{H}}{\partial J} : \Delta \lambda \mathbb{H} + \mathbb{H} : \mathbf{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda + \mathbb{H} : \Delta \lambda \mathbb{H} : \mathbb{N} \Delta \lambda \right)
\]
\[
+ 2 \Delta \lambda \mathbb{H} : \mathbf{N} \frac{(1 - \omega)}{(1 - \omega^t)} \frac{\partial \omega}{\partial \bar{z}} \delta \bar{z},
\]
(C.62)
where $\delta^*\tau$ is the variation of the Kirchhoff stress tensor in case of purely elastic deformation, Eq.(4.42).

Using $\delta J = J I : L_6$ and substituting Eqs.(C.55) and (C.60) gives,

$$
\delta \tau = \delta^* \tau - \frac{(1 - \omega)^2}{1 - \omega^t} \left( \frac{\partial H}{\partial J} : \Delta \lambda N I : L_6^c + H : N \left[ K_{\gamma u} : L_6^c + K_{\gamma z} \delta \bar{z} \right] \right.
\left. + H : \Delta \lambda \frac{1}{*_{\tau eq}} M : \delta^* \tau \right)
$$

(C.63)

Collecting terms,

$$
\delta \tau = \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\Delta \lambda}{*_{\tau eq}} [ ] \right) : \delta^* \tau - \frac{(1 - \omega)^2}{1 - \omega^t} \left[ \frac{\partial H}{\partial J} : \Delta \lambda N I + H : N K_{\gamma u} \right] : L_6^c
$$

$$
+ \left[ 2 \Delta \lambda H : N \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\partial \omega}{\partial \bar{z}} - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{H : N K_{\gamma z}}{N} \right) \delta \bar{z} \right]
$$

(C.64)

Substituting Eq.(C.45) for the variation of the elastic predictor,

$$
\delta \tau = \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\Delta \lambda}{*_{\tau eq}} [ ] \right) : \left( K_{u\bar{z}}^p \delta \bar{z} + K_{u\bar{u}}^p \right) : L_6^c
$$

$$
- \frac{(1 - \omega)^2}{1 - \omega^t} \left[ \frac{\partial H}{\partial J} : \Delta \lambda N I + H : N K_{\gamma u} \right] : L_6^c
$$

$$
+ \left[ 2 \Delta \lambda H : N \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\partial \omega}{\partial \bar{z}} - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{H : N K_{\gamma z}}{N} \right) \delta \bar{z} \right]
$$

(C.65)

Finally collecting terms again leads to,

$$
\delta \tau = K_{u\bar{u}}^p : L_6^c + K_{u\bar{z}}^p \delta \bar{z},
$$

(C.66)

with

$$
K_{u\bar{u}}^p = A : K_{u\bar{u}}^c + \frac{(1 - \omega)^2}{1 - \omega^t} \left( 2K \Delta \lambda N I - H : N K_{\gamma u} \right)
$$

(C.67)

$$
K_{u\bar{z}}^p = A : K_{u\bar{z}}^c + 2 \Delta \lambda H : N \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\partial \omega}{\partial \bar{z}} - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{H : N K_{\gamma z}}{N} \right)
$$

(C.68)

and

$$
A = \left( 1 - \frac{(1 - \omega)^2}{1 - \omega^t} \frac{\Delta \lambda}{*_{\tau eq}} [ ] \right)
$$

(C.69)


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