Microstructural Evolution during High-Frequency Post Weld Impact Treatments for High-Strength Steels
This research was carried out under Project No. M32.8.09333 in the framework of the Research Program of the Materials innovation institute (M2i) in the Netherlands (www.m2i.nl).

The research described in this thesis was performed in the department of Materials Science and Engineering, Faculty of 3mE, Delft University of Technology, Mekelweg 2, 2628CD, Delft, The Netherlands.
Microstructural Evolution during High-Frequency Post Weld Impact Treatments for High-Strength Steels

PROEFSCHRIFT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus prof.ir. K.C.A.M. Luyben,
voorzitter van het College voor Promoties,
in het openbaar te verdedigen op woensdag 28 januari 2015 om 12.30 uur

doors

Rangan Kaushik DUTTA

Master of Science in Material Science and Engineering
Delft University of Technology - Delft, The Netherlands
Geboren te Kolkata, West Bengal, India
Dit proefschrift is goedgekeurd door de promotor:
Prof.dr. I.M. Richardson

Samenstelling promotiecommissie:

Rector Magnificus, voorzitter
Prof.dr. I.M. Richardson, Technische Universiteit, Delft, promotor
Dr.ir. M.J.M. Hermans, Technische Universiteit, Delft
Prof.dr.ir. R. H. Petrov, Universiteit Gent, Gent
Prof.dr.ir. S. van der Zwaag, Technische Universiteit, Delft
Prof.dr.ir. L.A.I. Kestens, Technische Universiteit, Delft
Dr. B. Hu, Allseas Engineering, R&D, Delft
Prof.dr. S. Godet, Université Libre de Bruxelles, Brussels

Dr.ir. M.J.M. Hermans heeft als promotiebegeleider in belangrijke mate aan de totstandkoming van dit proefschrift bijgedragen.

Microstructural Evolution during High-Frequency Post Weld Impact Treatments for High-Strength Steels
R. K. Dutta
Ph.D. thesis of Delft University of Technology - with summary in Dutch
ISBN 978-94-91909-14-6

Key words: Quenched and tempered high strength structural steel (S690QL1), Welding, Thermal and mechanical anisotropy, Microstructural evolution, Ultrasonic impact treatment, Surface nanocrystallisation

Copyright © 2015 by R.K. Dutta
rangan.dutta@gmail.com

All rights reserved. No part of the material protected by this copyright notice may be reproduced or utilised in any form or by any means, electronic or mechanical, including photocopying, recording or by any information storage and retrieval system, without written permission from the author.

Printed by: Proefschriftmak.nl||Uitgeverij BOXPress.
# Contents

1 Introduction

1.1 Research objective .................................................. 2
1.2 Research approach and outline of the thesis ....................... 3

2 Background

2.1 Acoustic hardening and softening ................................ 5
2.1.1 Experimental observations ..................................... 5
2.1.2 Discussion ......................................................... 10
2.2 Surface severe plastic deformation ................................. 11
2.2.1 Experimental observation ....................................... 12
2.2.2 Discussion ........................................................ 13
2.3 Post weld treatment techniques ................................... 14
2.3.1 Modification of weld toe geometry ............................. 16
2.3.2 Modification of residual stress distribution ................. 16
2.3.3 Ultrasonic technologies ......................................... 18
2.4 Summary ...................................................................... 20

3 Characterisation of S690 steel base metal ......................... 21

3.1 Composition of S690QL1 ............................................ 21
3.2 Microstructural characterisation .................................... 23
3.2.1 Optical microscopy ................................................ 23
3.2.2 Scanning electron microscopy ................................... 23
3.2.3 Results ................................................................. 24
3.3 Mechanical and thermal properties ................................ 25
3.3.1 Micro-hardness ...................................................... 26
3.3.2 Room temperature tensile strength ............................. 26
3.3.3 Elevated temperature tensile strength ......................... 28
3.3.4 Dilatation properties .............................................. 31
3.4 Concluding remarks ................................................... 35
4 Welding and characterisation of weld metal
  4.1 Gas metal arc welding (GMAW) ........................................... 37
    4.1.1 Specimen size ....................................................... 39
    4.1.2 Clamping system .................................................... 39
    4.1.3 Welding equipment .................................................. 40
    4.1.4 Filler wire .......................................................... 41
    4.1.5 Welding condition ................................................... 41
  4.2 Welding thermal cycle ..................................................... 43
  4.3 Microstructural characterisation of the welded S690QL1 steel plates .... 44
    4.3.1 Inclusion formation in fusion zones ................................ 45
  4.4 Micro-hardness variation in welded S690QL1 steels .......................... 49
  4.5 Summary ............................................................... 50

5 In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1
  5.1 Experimental procedure .................................................. 52
    5.1.1 Sample preparation ................................................ 52
    5.1.2 In-situ synchrotron diffraction .................................. 53
    5.1.3 Characterisation of diffraction peaks ......................... 55
    5.1.4 Peak searching .................................................... 55
    5.1.5 Peak fitting ...................................................... 55
    5.1.6 Implementation and workflow .................................... 56
  5.2 Temperature dependent plane specific elastic constants in S690QL1 steel .. 57
    5.2.1 Introduction ...................................................... 57
    5.2.2 Thermal mechanical cycles ...................................... 58
    5.2.3 Data analysis .................................................... 58
    5.2.4 Results .......................................................... 59
  5.3 Anisotropy in thermal expansion of bainitic ferrite ....................... 63
    5.3.1 Introduction ...................................................... 63
    5.3.2 Thermal mechanical cycles ...................................... 64
    5.3.3 Data analysis .................................................... 64
    5.3.4 Results .......................................................... 67
  5.4 Discussion ............................................................. 69
    5.4.1 Discussion: Diffraction elastic constant ....................... 69
    5.4.2 Discussion: Anisotropy in thermal expansion of bainitic ferrite ........ 69
  5.5 Conclusion ............................................................. 70

6 The effect of tensile deformation with in-situ ultrasonic treatment on the microstructure of low carbon steel
  6.1 Introduction ............................................................. 71
  6.2 Experimental ........................................................... 73
## CONTENTS

6.2.1 Sample preparation ........................................... 74
6.2.2 Tensile deformation with\textit{in-situ} ultrasonic treatment .... 74
6.2.3 Microstructure characterisation ............................... 76

6.3 Methodology of calculation ...................................... 77
   6.3.1 Calculating geometrically necessary dislocation (GND) densities from EBSD data ............................. 77
   6.3.2 Evaluation of dislocation density from X-ray diffraction profiles .......................... 78

6.4 Results .......................................................... 79
   6.4.1 Microstructure and mechanical properties .................... 79
   6.4.2 Orientation gradients and GNDs ................................. 82
   6.4.3 Determination of dislocation contrast factors and density of dislocation by modified Williamson-Hall plot .......................... 86

6.5 Discussion ....................................................... 87

6.6 Summary .......................................................... 89

7 Accommodation of plastic deformation by ultrasound induced grain rotation 91
   7.1 Introduction ..................................................... 91
   7.2 Experimental .................................................... 91
   7.3 Results and discussion ......................................... 92
      7.3.1 Microstructure evolution .................................... 92
      7.3.2 Conceptual modelling of microstructure evolution .......... 99
      7.3.3 Recrystallised grain diameter ............................... 99
   7.4 Summary .......................................................... 100

8 Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment 101
   8.1 Introduction ..................................................... 101
   8.2 Experimental .................................................... 103
      8.2.1 Microstructure characterisation ............................... 105
      8.2.2 Criterion for grain boundary characterisation ............. 106
      8.2.3 Post processing methodology by TSL\textsuperscript{®} - OIM\textsuperscript{TM} ........................................ 107
   8.3 Results .......................................................... 108
      8.3.1 TEM investigations of post weld ultrasonic impact treatment ........................................ 108
   8.4 Discussion ....................................................... 119
   8.5 Summary .......................................................... 124

9 Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld 127
   9.1 Introduction ..................................................... 127
   9.2 Experimental .................................................... 127
      9.2.1 Microstructure characterisation ............................... 129
      9.2.2 Mechanical properties ........................................ 130
9.3 Results ................................................................. 132
  9.3.1 Microstructure evolution due to post weld ultrasonic impact
treatment ......................................................... 132
  9.3.2 Deformation microtexture ...................................... 138
  9.3.3 Mechanical properties ........................................ 142
9.4 Discussion .......................................................... 145
  9.4.1 Microstructural characterisation and texture analysis .... 145
  9.4.2 Mechanical response ........................................... 147
  9.4.3 Improvement in fatigue strength of welded components due to
        UIT ................................................................. 148
9.5 Conclusions .......................................................... 149

10 Conclusion and recommendations for future work .............. 151
  10.1 General conclusions .............................................. 151
  10.2 Recommendations for future work ............................. 153

References .............................................................. 155

Summary ................................................................. 181

Samenvatting ........................................................... 185

Acknowledgement ....................................................... 189

List of publications ................................................... 191

CV ................................................................. 195
Chapter 1

Introduction

Allseas Engineering bv has designed and built a new vessel, the "Pieter Schelte" (see figure 1.1), a unique special purpose ship, designed for pipe laying, single-lift installation and removal of large offshore oil and gas platform topsides and jacket structures and installation of oil and gas pipelines. Due to her large lifting capacity, Pieter Schelte drastically reduces the amount of offshore work associated with platform installation or decommissioning, moving this work to shore where it is less costly, faster, cleaner and safer.

Figure 1.1: Model of Pieter Schelte.

Because of the necessity to design and build lifting structures and equipment of a tremendously high capacity (up to 48,000 ton), the demand for the use of high-strength steels in welded structures has increased. A post weld treatment is advised in order to reduce the chances of weld cracking and to improve the fatigue life of the structures. This research is focused on the evaluation of material changes introduced during ultrasonic impact treatments and the contributions of the ultrasonic energy in order to determine whether ultrasonic techniques offer technological advantages for such
applications.

Cracks are considered to be the most dangerous welding defects. Small cracks in the weld propagate very fast, leading to complete failure of the structure. For this reason, all welding standards show zero tolerance towards cracks, whereas other defects can be accepted within certain limits, depending on the application. The weld toe and the weld root are the most critical parts with respect to crack initiation in the weld. To decrease the susceptibility of fatigue crack initiation and propagation, post weld treatment is advised. The post weld treatments can be broadly classified under Post Weld Heat Treatment (PWHT) (also called thermo-mechanical treatment) and volume and/or surface treatment. In practice, PWHT for stress mitigation have limited applicability to higher strength steels (>345 MPa) due to the risk of thermal damage.

In the class of mechanical post weld treatment, the most recent development is in the field of novel high-frequency peening, particularly ultrasonic methods. Although previous studies report a considerable increase in the the fatigue life [1–16], the detailed changes induced in the treated material, the mechanisms by which such changes occur and particularly the influence of the ultrasonic component, are poorly understood. Before introducing ultrasonic impact treatments in the process chain, it is necessary to understand the effects of the treatment on microstructure (evolution) and the mechanical behaviour of welded components. This forms the basis for the current research project.

1.1 Research objective

The aim of this research is two-fold: (1) to improve our understanding of the effect of tensile deformation with in-situ ultrasonic treatment on the microstructure of the material (low carbon steel in the present case); and (2) to improve our understanding of the microstructural evolution during welding and post weld high frequency impact treatments of the high strength S690QL1 steel. To achieve this, the following tasks were undertaken:

- Microstructure characterisation and thermal and mechanical property evaluation of the high strength S690QL1 steel.
- Welding of the steel and characterisation of the welding process and the weld zone.
- Ultrasonic impact treatment at the toe of the S690QL1 steel weld.
- Characterisation of the microstructural gradient induced by the ultrasonic impact treatment at the toe of the weld.
- Mechanical response of the ultrasonic impact treated weld toe.
1.2 Research approach and outline of the thesis

Chapter 2 provides an introduction to acoustic hardening and softening, microstructural changes due to severe surface plastic deformation and post weld treatment techniques.

The research work begins with an assessment of the microstructure and mechanical properties of the base material, which is a quenched and tempered high strength structural steel, S690QL1 [17]. Chapter 3 provides a detailed characterisation of the microstructure and temperature dependent mechanical properties of the steel.

Localised heating and melting of a work piece during welding leads to build up of residual stresses. When distortion is prevented due to constraints in structures, or due to clamping, stress levels will be high and may exceed the yield strength. Due to welding, the microstructure of the weld metal and the heat affected zone is different from that of the base material. In combination with construction details and weld geometry, the mechanical properties will vary considerably. Hence it is very important to characterise the microstructural and hardness gradients that are introduced due to welding. Chapter 4 discusses the welding procedure, thermal gradients introduced during multi-pass welding, microstructural characterisation and hardness gradients from the fusion zone to the base material.

The mechanical behaviour and in particular the fatigue performance, is influenced to a great extent by the presence of residual stresses [18–20]. Hence, it is important to quantify the residual stress state within a component with accuracy. The application of such methods almost always necessitates the estimation of the residual stresses from some measure of the local elastic and thermal strain field. Estimation of the residual stress state from lattice strains associated with a specific \( \{hkl\} \) reflection demands the use of the corresponding plane specific diffraction elastic constants (DECs) and thermal expansion coefficients, which in general differ from the bulk properties. This is particularly important for materials that are elastically anisotropic and phases that are tetragonal. For this purpose, \textit{in-situ} tensile tests and thermal-mechanical treatments have been carried out at different temperatures in a high energy synchrotron X-ray diffractometer, the details of which are described in chapter 5.

The softening effect in metals due to ultrasonic vibration is used in many industrial applications. The existing understanding of such an acoustoplastic effect is one in which the ultrasonic treatment either imposes additional stress waves to supplement the quasi-static applied load or causes heating of the metal. In both cases the intrinsic deformation resistance and/or mechanisms of the metal are assumed to be unaltered by the ultrasound. In chapter 6, the effect of an \textit{in-situ} ultrasonic treatment on the microstructure of low-carbon steel (Fe-0.051C-0.002Si-0.224Mn-0.045Al (wt.\%)) under tensile deformation is reported. Detailed microstructural analyses reveal that the ultrasonic treatment intrinsically alters the deformation characteristics of the metal.
The deformation microstructure underneath the area of treatment in the deformed samples was investigated by a combination of optical microscopy, scanning electron microscopy, crystal orientation mapping by electron backscattered diffraction and X-ray diffraction. A bimodal grain size distribution is observed with relatively small equiaxed grains with an average diameter of 10 µm at the grain boundaries of large elongated grains. Chapter 7 discusses the formation of these relatively small equiaxed grains in terms of dynamic recrystallisation by lattice and sub-grain rotation.

Surface modification by the generation of a nanostructured surface layer induced via ultrasonic impact treatment was performed at the weld toe of a welded high strength quenched and tempered structural steel, S690QL1. Such high frequency peening techniques are known to improve the fatigue life of welded components. The nanocrystallised structure as a function of depth from the top treated surface was characterised via a recently developed automated crystal orientation mapping technique in transmission electron microscopy and is presented in chapter 8. Based on the experimental observations, a grain refinement mechanism induced by plastic deformation during the ultrasonic impact treatment is proposed. It involves the formation of low angle misoriented lamellae displaying a high density of dislocations followed by the subdivision of microbands into blocks and the resulting formation of polygonal submicronic grains. These submicronic grains further breakdown into nano grains. The results show the presence of retained austenite even after severe surface plastic deformation. The average grain size of the retained austenite and martensite is \( \sim 17 \) nm and \( \sim 35 \) nm respectively. The in-grain deformation mechanisms are different in larger and smaller grains. Larger grains show long range lattice rotations while smaller grains show deformation through grain rotation. Also the smaller nano grains exhibit the presence of short range disorder. Surface nanocrystallisation also leads to an increased fraction of low-angle and low energy coincident site lattice (CSL) boundaries especially in the smaller grains (D<50 nm).

While chapter 8 discussed the formation of the nanocrystalline structure along the rolling/transverse plane, chapter 9 focusses on the microstructural gradient induced along the normal/transverse plane. The nanocrystalline microstructure and texture was characterised as a function of depth from the surface via high-resolution electron backscatter diffraction (EBSD). Results indicate that the top 20 µm undergoes dynamic recrystallisation. The processes occurring in 20-175 µm depth are explained by progressive subgrain misorientation (PriSM). Recrystallisation results in lowering of hardness in the zone 0–20 µm from the top surface.

Each chapter has its own discussions and conclusions or summary; in addition, the conclusions derived from the entire work are enumerated in chapter 10 along with suggestions for further investigations.
Chapter 2

Background

In this chapter, an introduction to acoustic hardening and softening, microstructural changes due to severe surface plastic deformation and post weld treatment techniques is provided.

2.1 Acoustic hardening and softening

The physical properties of materials are influenced by mechanical vibrations of sufficient amplitude. This was first reported by Blaha and Langenecker [21] in 1955. They probed the softening effect of superimposed ultrasonic vibrations on zinc single crystals which were undergoing tensile deformation. Since then much research has been focused on the influence of superimposed ultrasonic vibrations on materials, leading to the development of a number of practical industrial applications. The observed effects of ultrasonic vibrations can be broadly classified into two categories, namely volume effects and surface effects. Volume effects include all processes which influence the internal friction of the test specimen whereas, surface effects include all processes which influence the external friction between the test specimen and a forming tool.

2.1.1 Experimental observations

The first evidence of the influence of mechanical vibrations on the physical properties of materials dates back to 1955 when Blaha et al. [21] insonated (exposed to ultrasound) zinc single crystals with ultrasound at 800 kHz. The ultrasound was generated by an ultrasonic generator with a maximum power output of 25 W (2 W cm$^{-2}$). The result are shown in figure 2.1. Curves A and B represent intermittent and continuous application of ultrasound respectively. Curve A shows that intermittent application reduces the tensile stress by about 40 % but upon termination of the ultrasound, the tensile stress returns to the value it had prior to insonation. Curve B shows the
lowering of the tensile stress throughout the entire tensile test. This softening effect upon application of the ultrasound is referred to as the “Blaha effect”. Blaha et al. reasoned the softening effect was due to the additional energy supplied by the ultrasound causing the dislocations to move more readily.

**Figure 2.1:** Effect of superimposed 800 kHz ultrasound on the tensile deformation of zinc single crystals. Specimen coupled to an ultrasonic generator by immersion in an insonated liquid bath. A - Intermittent application of ultrasound with constant amplitude; B - continuous application of ultrasound [21].

Further investigation was carried by Nevill et al. [22] to study the effect of mechanical vibrations in the frequency range of 15 to 80 kHz on the tensile elongation of low carbon steel wires. The vibrating mechanism used by Nevill et al. was different than the one used by Blaha et al. [21]. The experimental results are shown in figure 2.2. The intermittent mechanical vibrations caused the tensile stress to decrease by an amount which is directly proportional to the amplitude of vibrations. The continuous vibrations also resulted in a reduction of tensile stresses but unlike the results obtained previously by Blaha et al. [21], the mechanical vibrations showed no effect at stresses below the macroscopic yield point of low carbon steel specimens.

Nevill et al. [22] showed that the reduction in stress was proportional to the amplitude of vibrations but independent of the frequency in the range of 15 to 80 kHz. The stress decrease was also independent of prior strain for values up to 15 % and temperature in the range of 30 °C to 500 °C. They concluded that the mechanisms by which dislocations can absorb energy from vibrations, namely resonance, relaxation and hysteresis does not provide satisfactory explanation to justify the observed results. However, they were able to explain their observed results by a macroscopic mechanism of superposition of steady and alternating stresses.
2.1 Acoustic hardening and softening

Figure 2.2: Effect of superimposed mechanical vibrations of 15 to 80 kHz on the tensile deformation of low-carbon steel wires. Upper curve - intermittent application of vibrations; lower curve - continuous application of vibrations [22].

Langenecker et al. [23–25] carried out further investigation on the influence of ultrasound on the physical properties of materials. They observed that, although the low-amplitude ultrasonic waves caused no permanent effect on the stress-strain curve of the zinc crystal, ultrasonic vibrations having pressure amplitudes larger than $2.5 \times 10^6$ N m$^{-2}$ caused pronounced work hardening, which was directly proportional to the pressure amplitude over a considerable range, decreased in amount following repeated applications of ultrasound at a constant amplitude, and eventually levelled off at a saturation level corresponding to about $5.5 \times 10^6$ N m$^{-2}$ pressure amplitude.

Figure 2.3 shows the experimentally obtained stress-strain curves of aluminum single crystal during tensile deformation at room temperature with various amplitudes of ultrasound applied or without ultrasound at various test temperatures. Similar results were reported for zinc, stainless steel and beryllium polycrystalline specimens. Langenecker [25] observed that in the case of aluminum crystals an ultrasonic energy of $10^{15}$ eV cm$^{-3}$ was sufficient to reduce the stress to nearly zero, while $10^{22}$ eV cm$^{-3}$ of thermal energy was required to cause the same amount of stress reduction.

Langenecker [25] also compared the stresses created by ultrasound with the measured yield stresses of various metals. For an ultrasonic wave possessing a power level of about 50 W cm$^{-2}$, he calculated that the acoustic stresses for zinc, aluminum, beryllium, iron, and tungsten ranged from $5.3 \times 10^6$ N m$^{-2}$ to $10^7$ N m$^{-2}$, while the measured yield stresses varied from $10^6$ N m$^{-2}$ to $10^9$ N m$^{-2}$. Thus, for soft zinc crystals there is agreement between the ultrasonic stress and the yield stress, but for all harder materials beginning with aluminum, no such agreement exists, and the experimentally obtained stress-strain curves show that the measured stress drops
Background

Figure 2.3: Stress-strain curves of aluminum crystals; (a) continuous application of 20 kHz ultrasound at various power amplitudes at 18 °C; and (b) without ultrasound at test temperatures ranging from 18 °C to 600 °C [24, 25].

to nearly zero at applied ultrasonic stress levels much lower than the value given by the yield stress. By using specimens of different lengths and diameters and by photoelastic measurements, it was shown that stress reinforcement by standing waves can be discarded as an explanation of the strong effect of relatively low ultrasonic power levels. Langenecker et al. [25] suggested that the cause of the effects observed was due to localised heating, which takes place in regions around dislocations and other imperfections when the ultrasonic waves are scattered.

Baker and Carpenter [26] probed the effect of superimposed mechanical vibrations on the tensile stress-strain curve of polycrystalline copper using a composite resonator technique operating at frequencies near 50 kHz. They found that the stress drop was approximately equal to the maximum vibrational stress. Pohlman and Lehfeldt [27] studied the influence of ultrasonic vibration on the plastic forming of metals in which a polycrystalline copper specimen was subject to tensile deformation while impulses of 20 kHz ultrasound were intermittently superimposed. Their results showed that the force drop due to insonation was only seen in the plastic part of the stress-strain curve and not the elastic part. This finding led them to the conclusion that ultrasound has a direct influence on the internal behaviour of the sample, since an indirect influence through a temperature increase causing a force decrease would be more gradual, and that the force drop was directly proportional to and of the same magnitude as the acoustic stress amplitude.
2.1 Acoustic hardening and softening

In 1966 Langenecker [28] demonstrated that ultrasonic irradiation of low power reduced the apparent static stress necessary for tensile deformation of metal specimens during application of the ultrasound, while work hardening was observed to occur following high power ultrasonic irradiation. Figure 2.4 represents the experimental results he obtained during the tensile elongation of a zinc crystal. Langenecker interpreted this acoustic hardening as a result of a reduction in the mean free path of dislocations caused by the continuous formation of obstacles, such as vacancy rings during acoustically activated dislocation oscillations. Wesmacott and Langenecker [29] in later studies verified the multiplication of dislocations by intense irradiation of ultrasonic stress waves in aluminum.

![Figure 2.4](image_url)

**Figure 2.4:** Acoustic softening by low power ultrasound and acoustic hardening by high power ultrasound in a zinc crystal during tensile deformation. Ultrasound power intensity at point (a) 5 W cm$^{-2}$ (b) 15 W cm$^{-2}$ (c) 25 W cm$^{-2}$ [28].

Puškar [30] probed the change in the dislocation density in mild steel upon exposure to high-intensity ultrasound at 23 kHz. In order to evaluate the effect of high-energy ultrasonic radiation, the central parts of the specimen were subjected to a longitudinal stress amplitude in the range of 350 MPa. He recorded that the original dislocation structure, created by the individual segments of dislocation, did not change for stress amplitudes of up to 70 MPa. The use of higher stress amplitudes shortens the dislocation segments thus non-dense dislocation clusters are created, anchored by the original dislocations. When the stress amplitude is increased still further the density of clusters in a grain increases and the clusters of dislocation are more densely knitted. Following insonation at a stress amplitude of 350 MPa, at a depth of about 0.1 mm, a well formed cellular structure, with cell size of 0.3 µm was observed. At the surface, the arrangement of dislocations was found to be different. Puškar noted that the dislocation clusters at the surface were less dense and it was possible to see some areas, where fatigue slip lines came out onto the surface of the specimens. Figure 2.5 presents his findings on the change in the dislocation density with varying...
stress amplitudes. He also recorded that after insonation by a stress amplitude of 350 MPa, very fine precipitates were present in the areas of high dislocation density, especially with foils, from a depth of about 0.1 mm under the surface of the specimens.

![Graph showing changes in dislocation density](image)

**Figure 2.5:** Changes in the density of dislocations per unit area in mild steel with increasing stress amplitudes of ultrasonic radiation on the surface of specimens 's', at a depth 0.1 mm under the surface 'u' and at a depth of 0.2 mm below the surface of specimens 'd' after insonation at temperatures greater than 20 °C [30].

### 2.1.2 Discussion

Based on the experimental observations it may be concluded that high power ultrasound has three major effects on the mechanical deformation of metal specimens, namely, work softening, work hardening, and reduction of friction between tool and workpiece.

When high-power ultrasonic vibrations possessing sound intensities lower than a certain threshold value are superimposed on a metal specimen during a constant elongation rate tensile test, the tensile stress required to deform the specimen is immediately reduced. The amount of this stress reduction is directly proportional to the intensity of the ultrasound and is independent of frequency over the range from 15 Hz to 1.5 MHz [23, 24]. Some investigators have reported that although there is a slight temperature rise in the test specimen during application of ultrasound, it only amounts to about 1 °C per insonation period, and that $10^7$ to $10^8$ times more thermal energy than ultrasonic energy is required in order to produce the same stress reduction. It is controversial to say how the effect takes place in the elastic range of the tensile stress-strain curve since the behaviour in this region is not consistent with different materials. The effect has been reported to be independent of test temperature in the range 30 °C to 500 °C. The applied tensile stress returns to the value it would
2.2 Surface severe plastic deformation

have attained in the absence of vibrations upon removal of the ultrasonic vibrations. In most experiments this return follows immediately upon removal of the ultrasonic vibrations, while in some experiments it is slightly more gradual. No apparent change in the structure of the metallic test specimens due to the ultrasonic vibrations is observed. The softening effect may be attributed to the preferential absorption of the ultrasonic energy at dislocation cells causing them to move at lower stress. However, when intensities of the ultrasonic vibrations are higher than a certain threshold value, the applied tensile stress reaches a value higher upon removal of the vibrations, than it would have attained in the absence of vibrations [30]. The amount of this work hardening is directly proportional to the intensity of the ultrasound over a considerable range, decreases in amount following repeated applications of ultrasound at a constant intensity, and eventually levels off at a saturation level of ultrasonic intensity. This effect is also reported to be independent of frequency over the range 15 kHz to 1 MHz. The high sound intensity causes considerable heating of the sample. Whether this high heating results in permanent microstructural changes resulting in the hardening effect is controversial although some authors argue that in ultrasonic treatment of alloys, the diffusion processes and the dynamic activation are significantly intensified [31] which leads to phase transformation.

2.2 Surface severe plastic deformation

Surface nano-crystallisation (SNC) [32], which was developed in 1999 by Lu, can transform a coarse grained surface layer of a bulk material into nano-sized grains by surface severe plastic deformation (S\textsuperscript{2}PD). This technique can induce nano-grains and grain size gradients into the surface region of bulk metals and alloys, which significantly alters the properties. The treatment does not change the chemical composition of materials. Many S\textsuperscript{2}PD-based processes have been put forward recently, such as surface mechanical attrition treatment (SMAT) [33, 34], ultrasonic shot peening (USSP) [35, 36], ultrasonic peening (UP) [37], laser shock peening (LSP) [38], ultrasonic surface rolling processing (USRP)[39], ultrasonic surface metallic nano-crystallisation technology (USMNT) and ultrasonic cold forging technology (UCFT) [40]. These technologies have been shown to improve the tensile strength [32, 35], hardness [40, 41], wear resistance [40, 42–44], fatigue strength of materials [40, 41, 45] and alters the residual stress state[32, 40, 41].

Forming a nano-structured surface layer from coarse-grain poly-crystals involves the generation of dislocations, twinning, and development of grain boundaries with high angle misorientation. Plastic deformation behaviour and dislocation activity in metals and alloys depend strongly on the lattice structure and the stacking fault energy (SFE). Using transmission electron microscopy (TEM), the mechanisms of grain refining in several metals and alloys have been investigated, including Fe (body centred cubic (bcc) with a high SFE of about 200 mJ m\textsuperscript{-2}) [34], Al (face centred cubic (fcc)
Background

with a high SFE of about 166 mJ m$^{-2}$) [36], Cu (fcc with a medium SFE of about 78 mJ m$^{-2}$) [46], AISI 304 stainless steel (fcc with a low SFE of about 17 mJ m$^{-2}$) [47], Co (hexagonal close packed (hcp) with a low SFE of about 27 mJ m$^{-2}$) [48], Mg (hcp with a medium SFE of about 50 to 80 mJ m$^{-2}$) [49] and Ti (hcp with a high SFE of about 300 mJ m$^{-2}$) [33].

2.2.1 Experimental observation

Several researchers [50–55] studied the nano-crystalline surface layer of iron and/or steel induced by different S$^{2}$PD-based processes. High resolution transmission electron microscopes (HRTEM) were used to characterise the microstructure. Tao et al. [55] investigated the surface nanocrystallisation mechanism in iron induced by SMAT. Based on the experimental observations, a grain refinement mechanism induced by plastic deformation involves the formation of dense dislocation walls (DDWs) and dislocation tangles (DTs) in original grains and in the refined cells (figure 2.6) as well, transformation of DDWs and DTs into sub-boundaries with small misorientations separating individual cells or sub-grains, and the evolution of sub-boundaries to highly misoriented grain boundaries. Experimental evidence and analysis of the grain refinement mechanism indicated that high strains with a high strain rate are necessary for the formation of nano-crystallites during plastic deformation of metals.

Figure 2.6: A cross-sectional TEM image in the sub-micro-sized section showing equiaxed sub-grains with different dislocation configurations and boundaries (solid triangles for DDWs and DTs, open triangles for sub-boundaries) [55].

Similar microstructural evidence was found in ultrasonically shot peened AISI 304 austenitic stainless steels by Lee et al. [56]. Nano-sized grains, multi-directional mechanical twins and strain-induced martensite were formed on the surfaces, and the volume fraction of strain-induced martensite in the ultrasonically peened specimen
was higher than that of the shot-peened specimen (obtained by treating the specimen using shots with a diameter of 0.8 mm and a work flow of 30 kg min$^{-1}$). The ultrasonically peened specimen (obtained by treating the specimen with ultrasonic equipment with piezoelectric transducer and a tungsten carbide tip), which had a smoother surface and contained more strain-induced martensite, showed superior general and localised corrosion resistance to the as-received and shot-peened specimens. They concluded from TEM observations that the ultrasonically peened specimen had more plastic strain energy than the shot-peened specimen and that the plastic strain caused by both the peening treatments decreases as the depth into the matrix increases.

2.2.2 Discussion

Microstructural investigation by TEM and SEM have revealed that the ultrasonic impact treatment (UIT) produces an ultrafine grain structure in the nano-crystalline regime down to a depth of about 6-10 µm from the top surface layer. The average grain size increases with distance from the surface due to the corresponding decrease in the strain energy. TEM observations suggested that sub-grains and micro-bands can also be formed. Such a surface layer of ultrafine grain size explains the feature, originally named, “white layer structure” which was reported in a previous work [1].

The grain refinement process accompanied by an increase in the strain in the surface layer involves:

- the onset of twins and the intersection of the twin system (depending on crystal structure and SFE)
- the formation of dislocation walls
- the nucleation of microbands associated with the splitting of dislocation walls
- the subdivision of microbands into low angle disoriented blocks and then highly disoriented polygonal sub-micron grains, and
- further breakdown of sub-micron polygonal grains into randomly oriented nano-grains.

In SMA treated iron [55], TEM observations suggest that the sub-grain size gradually increases with increasing distance from the surface, which indicates that the grain size can change within the impact affected zone from the top surface down to the matrix due to strain variations. Such findings acknowledge the fact that the S$^2$PD process parameters, such as impact frequency, amplitude under load and processing speed (mm min$^{-1}$) can control the average grain size and microstructure of aluminium alloys. Further evidence demonstrates that the feed rate mostly controls the surface effects and the amplitude under load is responsible for the penetration depth [57].
2.3 Post weld treatment techniques

Localised heating and melting of a work piece during welding leads to the build up of residual stresses. When distortion is prevented due to constraints in the structures, or due to clamping, stress levels will be high and may exceed the yield strength. In general, the welded areas experience longitudinal and transverse tensile stresses, balanced by areas with compressive stresses.

The microstructure of the weld metal and the heat affected zone (HAZ) is different from the base material due to welding. In combination with construction details and weld geometry, the mechanical properties vary considerably. Furthermore, the residual stresses can have significant influence on the fatigue life of engineering components. The near surface tensile stresses tend to accelerate the initiation and growth stages of fatigue cracks, while compressive residual stresses close to the surface can prolong fatigue life [58]. Post weld treatments are often carried out to mitigate or redistribute the residual stresses, particularly at the component surface. An internal tensile stress zone is not as dangerous as tensile stresses at the surface since cracks are less likely to initiate in the interior [59].

Fatigue design is limited by poor strength details in the majority of cyclically loaded welded structures, such as T-butt welds or fillet welds, which normally fail from the weld toe. The poor fatigue strength of such joints can be explained in terms of the following three factors [60, 61]: First, a geometric stress concentration is introduced by a welded joint, the severity of which depends on the type of weld and its orientation with respect to the loading direction. Of particular significance is the sharp change of section dimensions at the toe of a transversely-loaded weld, which is often exacerbated by the presence of undercut.

Second, crack-like discontinuities in the form of non-metallic inclusions exist at the weld toe. These inclusions, which have been observed by a number of researchers [62–65], are formed as a result of the welding operation and as such are an inherent part of the welded joint. They can be up to 0.4 mm deep, but will be effectively deeper when combined with any undercut. Not surprisingly, they act as sites for fatigue crack initiation, with the result that the crack initiation period is insignificant and the majority of the fatigue life is spent propagating pre-existing flaws.

The poor fatigue strength of transverse fillet welds is in sharp contrast with the behaviour of unwelded components where a significant part of the life may be used simply initiating a fatigue crack. For example, in the case of smooth components, fatigue crack initiation may occupy 95 % of the life in the low stress/long life regime. An obvious consequence is that the fatigue life of the welded joint is much lower than that of the smooth component, especially in the low stress/long life regime, where the fatigue limit can be so low that design for infinite life is not a practical proposition for welded joints.
The third significant factor is the presence of tensile residual stresses produced by welding, which may reach the yield strength magnitude in real structures. The effect of welding residual stress is equivalent to the imposition of a high tensile static stress, with the result that any applied cyclic stress effectively cycles down from tensile yield [60]. This is particularly significant if the applied stress is compressive, since the resultant stress becomes effectively tensile [18] and hence damaging. However, any applied stress cycle is affected and overall it can be concluded that the fatigue lives of welded joints are independent of applied mean stress and dependent only on the applied stress range [60, 66].

There are also other important consequences associated with differences between the fatigue crack initiation and propagation properties in materials. Perhaps the most important is the influence of the material tensile strength. The number of cycles required to initiate a fatigue crack in smooth polished specimens tends to increase with an increase in tensile strength, giving the well-known rule of thumb that the alternating fatigue limit is approximately half the ultimate tensile stress. Thus, the fatigue strength of a machined high strength steel component can be expected to be higher than that of a mild steel component. However, the rate of fatigue crack propagation is not affected in the same way and similar rates have been measured for a whole range of steel strengths. Consequently, the fatigue lives of welded joints do not depend on material strength, and welded high strength steels have the same fatigue lives as welded mild steel [60]. Thus, to take advantage of high strength steels in applications involving welded joints, it will be necessary to take measures to improve their fatigue strengths.

A number of techniques are now available for improving the fatigue strength of welded joints [60, 61, 67]. Since the weld toe is the most common potential site for fatigue cracking, attention has been focused on improvement of this zone in critical regions of a component or structure. All the available techniques rely on addressing one or more of the three factors discussed above. Thus, the required improvements can be achieved by reducing the stress concentration due to the joint geometry, by removing the crack-like weld toe discontinuities and by modifying the residual stress distribution, or more specifically by introducing beneficial compressive residual stress, or by a combination of these principles. Notable examples of the first two are grinding and re-melting of the weld toe, while spot heating and various weld toe peening techniques are examples of methods for introducing compressive residual stress.

Use of weld toe improvement techniques is seen as a way to utilise high-strength steels to advantage since the resulting design S-N curve will be higher than that for the as-welded joint. However, there is also limited evidence to indicate that the beneficial effect of some improvement techniques will be greater in a high-strength steel than in a medium-strength steel weldments [60, 63, 67]. On the one hand this could be expected if the technique extends fatigue crack initiation period, as is the aim of
Background

weld toe grinding or re-melting. On the other hand, the residual stress techniques could be expected to be more beneficial when applied to high-strength steels if the magnitude of the compressive residual stress induced, would increased with material yield strength.

The post weld treatment techniques are employed to improve the fatigue life of the engineering component after welding. A summary of the various improvement techniques is presented in figure 2.7.

2.3.1 Modification of weld toe geometry

The techniques presented in this group are used to achieve two objectives:

- Removal of slag inclusions and
- Achievement of a smooth transition between the weld metal and parent material.

Both the above factors result in introducing a significant fatigue crack initiation period into the fatigue life, thereby increasing both the fatigue life and the fatigue endurance limit. Furthermore, since fatigue crack initiation can be more difficult in high-strength steels, the techniques may prove to be even more beneficial when applied to welded high-strength steels.

In most investigations, this group of improvement techniques has been investigated using test specimens consisting of structural steel plates with transverse or longitudinal fillet welded attachments [2, 51]. In each case, the weld toes or ends lying transverse to the direction of loading were treated. Limited studies have also considered transversely-loaded butt (groove) welds, tubular joints, and fillet welds in aluminum alloys.

2.3.2 Modification of residual stress distribution

As previously stated, the presence of high tensile residual stresses in as-welded structures has an adverse effect on fatigue strength, particularly if the applied loading gives rise to tensile stresses. Thus, relaxation of these tensile residual stresses can be expected to be beneficial from a fatigue point of view. Some of the residual stress improvement techniques aim to achieve such stress relief. However, more important are those techniques that aim to go further and actually introduce beneficial compressive residual stresses. In contrast to stress relief, such techniques can be expected to be beneficial for conditions of applied compressive and tensile stress.
2.3 Post weld treatment techniques

Figure 2.7: Classification of some weld-improvement methods [68]. Ultrasonic peening techniques include ultrasonic impact treatment (UIT) and ultrasonic shot peening (USSP).

An advantage over the techniques considered in section 2.3.1, is that, in principle, some of the residual stress improvement techniques are not confined to the treatment of the weld toe but can be used to treat any stress concentration, including sub-surface discontinuities.
2.3.3 Ultrasonic technologies

The various techniques discussed above use different mechanisms to improve fatigue resistance in engineering components, including the application of heat, mechanical impact, ultrasonic oscillations or a combination of them. The focus of this section is to evaluate the technologies that employ ultrasonic oscillations to generate impact pulses that cause strain hardening of the surface being treated. Ultrasonic Impact Treatment (UIT) [9] and ultrasonic peening (UP) [8] (also called ultrasonic shot peening (USSP) [69]) may be placed among such technologies. In order to affect the material, a freely moving indenter(s) receives kinetic energy from the ultrasonic oscillation system.

The ultrasonic transducer oscillates at a high frequency, typically in the range between 20-30 kHz. The transducer may be based on either piezoelectric or magnetostrictive technology. Both accomplish the same task of converting alternating electrical energy to oscillating mechanical energy, but do it in a different way. In magnetostrictive transducers electrical energy from the ultrasonic generator is first converted into an alternating magnetic field through the use of a wire coil. The alternating magnetic field is then used to induce mechanical vibrations at the ultrasonic frequencies in resonant strips of magnetostrictive material. Piezoelectric transducers convert the alternating electrical energy directly to mechanical energy through the piezoelectric effect. Whichever technology is used, the output end of the transducer will be oscillating, typically at an amplitude of 20-40 µm. During the oscillations, the transducer tip will impact the striker at different stages in the oscillation cycle [8].

![Schematic diagram of the Esonix UIT](image)

**Figure 2.8:** Schematic diagram of the Esonix UIT [6]. 1 magnetostrictive transducer, 2 waveguide, 3 indenter, 4 treated surface, I ultrasonic oscillations, II impact impulses.

Magnetostrictive transducers (as shown in figure 2.8) are generally less efficient than the piezoelectric ones from the energy conversion perspective. This is due primarily to the fact that the magnetostrictive transducer requires a dual energy conversion
2.3 Post weld treatment techniques

from electrical to magnetic and then from magnetic to mechanical. Some efficiency is lost in each conversion. Magnetic hysteresis effects further reduce the efficiency of the magnetostrictive transducer. In addition, the magnetostrictive transducer needs forced water-cooling. The equipment in this case is relatively heavy and expensive [8].

A single striker arrangement is shown in figure 2.8. Different numbers of strikers with different arrangements are used for different industrial application. A few types of working heads with different number of strikers are typically used [70]:

- single-row heads with 3 to 5 strikers is applied, for example, for treatment of weld toe zones,
- heads with one striker is generally applied for treatment of difficult-to-access surfaces such as crossing welds etc.,
- heads with 7 or more strikers are mainly applied for treatment of planar surfaces or surfaces with a large radius of a curve.

Independent research was carried out to study the effectiveness of the indenter geometry in the USSR in the 1950s and 1960s [42, 43, 71], where indenters rigidly attached to the output end of the oscillating system were used for surface strengthening. Ultrasonic treatment variation with a freely moving ball-indenter was also examined [72–74]. It was shown that a fixed and axially freely moving ball-indenter may in principle be used to create plastic deformation; this played a positive role in further developing strain hardening methods. However, there were some drawbacks limiting wide industrial application: the rigidly attached indenter did not provide sufficient depth of plastic deformation due to a low kinetic energy. The freely moving ball did not produce a uniform plastic deformation because of the large number of degrees of freedom, giving rise to random rebounds at various angles from the processed surface during tool movement.

Ultrasonic treatment has been reported to be effective for relieving harmful tensile residual stresses and introducing beneficial compressive residual stresses in the surface layers of parts and welded elements. The mechanism of residual stress redistribution is connected mainly with two factors. At a high-frequency impact loading the oscillation, with complex frequency mode spectrum, propagates in a treated element. The nature of this spectrum depends on the frequency of the ultrasonic transducer, mass, number and form of impact units (spheres, rods etc.), and also on the geometry of the treated element. The second and the more important factor, at least for fatigue improvement, is surface plastic deformation, which leads to introducing of the beneficial compressive residual stresses (figure 2.9).
2.4 Summary

In this chapter, an overview of past research performed in the field of acoustic hardening and softening is presented. Materials behave differently on exposure to ultrasound. Several possible theories are presented to explain such material behaviour. However, the role played by the ultrasound in altering the material microstructure and properties is not clearly understood. Comparison between shot peened and ultrasonically peened techniques to impart severe surface plastic deformation to the material have shown that more plastic strain energy is introduced in the material with ultrasonic treatment. However, the intensities of these impacts have not been reported and the role played by the ultrasonic component cannot therefore be readily determined. A brief report on the various post weld treatment techniques and the mechanisms to alter the mechanical properties in welded structures has been presented. The ultrasonic techniques in post weld treatments is probed in detail. The effect of specific ultrasonic impact treatment, the effect of the ultrasonic component and the mechanical impact, their role in altering the microstructure, grain morphology and residual stress in the weld zone of high strength steel must be understood thoroughly before such techniques can be introduced in the process chain.
Chapter 3

Characterisation of S690 steel base metal

S690 steel belongs to the quenched and tempered group of steels. The steel is rapidly cooled to room temperature after prior heat treatment steps and tempered at a temperature below the eutectoid transformation temperature ($A_{C1}$). Quenching followed by tempering in steels is carried out to obtain an advantageous combination of strength and toughness. Quenching of steel from the fully austenitic state or from the inter-critical region can enhance the formation of martensite at the expense of pearlite. The subsequent tempering treatment results in the formation of tempered martensite, which contributes to enhance ductility. S690 steel is produced in a number of grades, i.e., S690, S690QL and S690QL1. The variants S690QL and S690QL1 are produced to achieve acceptable notch toughness of 50 and 60 J respectively at low temperatures (0 °C) [17]. The current work focuses on the S690QL1 variant. This chapter focuses on the experimental procedures used for the qualitative and quantitative microstructural characterisation and evaluation of the thermal and the mechanical properties of the as-received S690QL1 steel prior to welding.

3.1 Composition of S690QL1

Commercially produced S690QL1 steel follows the EN 10025-6 [17] standard and falls under the group of high yield strength structural steels in the quenched and tempered condition. Table 3.1 gives the chemical composition of S690QL1 according to the standards. The values in the table indicate the maximum amount of each of the alloying elements in the ladle. The carbon equivalent ($CE_{IIW}$) for the alloy varies with the nominal plate thickness. The maximum $CE_{IIW}$ for nominal thickness of up to 50 mm is 0.65, while the maximum $CE_{IIW}$ for nominal thickness within 50-100 mm is 0.77. Depending on the thickness of the product and the manufacturing con-
ditions, the manufacturer may add one or several alloying elements to the steel up to the maximum values given in table 3.1 in order to obtain the specified properties. Grain-refining elements like, Nb, Ti, V, Zr and Al should form at least 0.015 wt. % of the total composition.

Table 3.1: Maximum permissible alloy content of S690QL1 steels according to standards [17] in wt. %.

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Ni</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Mo</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>0.8</td>
<td>1.7</td>
<td>2.0</td>
<td>0.02</td>
<td>0.01</td>
<td>1.5</td>
<td>0.7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>V</th>
<th>N</th>
<th>Nb</th>
<th>Ti</th>
<th>Cu</th>
<th>Zr</th>
<th>B</th>
<th>CE&lt;sub&gt;IIW&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.12</td>
<td>0.015</td>
<td>0.06</td>
<td>0.05</td>
<td>0.5</td>
<td>0.15</td>
<td>0.005</td>
<td>0.83</td>
</tr>
</tbody>
</table>

Table 3.2 shows the chemical compositions from the ladle of the S690QL1 steel used in this work. The total content of alloying elements is about 2 wt. % and the CE<sub>IIW</sub> is around 0.42 wt. %.

Table 3.2: Chemical composition of S690QL1 steels from ladle in wt. %.

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Ni</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.16</td>
<td>0.2</td>
<td>0.87</td>
<td>0.11</td>
<td>0.012</td>
<td>0.001</td>
<td>0.33</td>
<td>0.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mo</th>
<th>N</th>
<th>Nb</th>
<th>Ti</th>
<th>Cu</th>
<th>B</th>
<th>CE&lt;sub&gt;IIW&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.21</td>
<td>0.0035</td>
<td>0.027</td>
<td>0.005</td>
<td>0.02</td>
<td>0.0021</td>
<td>0.42</td>
</tr>
</tbody>
</table>

The chemical composition of the as-received plates was analysed again using the X-Ray Fluorescence (XRF) technique and LECO carbon and sulphur analysis. The XRF analyses were performed on 4 different samples with a Philips PW2400<sup>TM</sup> WD-XRF spectrometer. Data evaluation was done with UniQuant® 5.0 software. The results obtained (see table 3.3) were averaged and found to be within acceptable range of the ladle composition.

The carbon and sulphur content was determined with a LECO CS-225 furnace. The sample was burned in a high-frequency induction furnace and the products of combustion were passed through a moisture trap to the sulphur IR cell where sulphur is measured as sulphur dioxide. The gases exiting the sulphur cell pass through a catalyst where any carbon monoxide is converted to carbon dioxide. SO<sub>3</sub> was removed,
3.2 Microstructural characterisation

then carbon was measured as carbon dioxide in the carbon IR cell. The results were adjusted for weight and calibration factors. The equipment was calibrated with a standard sample of steel rings, (LECO part. no. 501-504) containing 0.358 (± 0.004) wt. % C and 0.0178 (± 0.0008) wt. % S. The analysis was carried out three times and then averaged. The carbon and sulphur content (table 3.3) in the as-received plates were found to be within the range of elemental composition from the ladle.

Table 3.3: Chemical composition of S690QL1 steels obtained by XRF and LECO analysis, in wt. %.

<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>Mn</th>
<th>Ni</th>
<th>P</th>
<th>S</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.16</td>
<td>0.86</td>
<td>0.075</td>
<td>0.04</td>
<td>0.0007</td>
</tr>
<tr>
<td>Cr</td>
<td>0.32</td>
<td>0.18</td>
<td>0.03</td>
<td>0.006</td>
<td>0.015</td>
</tr>
</tbody>
</table>

3.2 Microstructural characterisation

3.2.1 Optical microscopy

The as-received plates measuring 400×1500×16 mm^3 were flame cut into smaller plates measuring 200×300×16 mm^3, which were then cross sectioned transverse to the rolling direction for metallography analysis. Samples were polished and etched for optical and scanning electron microscopy studies. Two etching procedures were followed to delineate the microstructural constituents present. In the first, the polished samples were etched with a 4 % Picral solution for 120 seconds to delineate the grain boundaries. In the second etching method, samples were etched in Le Pera solution for 25 seconds to evaluate the presence of any retained austenite. Etching in Le Pera solution changes the colour of retained austenite/martensite into bright whitish blue, bainite into brown and allotriomorphic ferrite into a tan colour [76, 77]. The samples were subsequently analysed using an Olympus BX60M™ optical microscope with a quantitative analysis software (analySIS™).

3.2.2 Scanning electron microscopy

Two different methods were implemented to prepare samples for scanning electron microscopy (SEM). In the first, the samples were electro-chemically polished using A2 solution; in the second, polished samples were etched in 4 % Picral solution for 120 seconds. Scanning electron microscopy was performed using a JEOL JSM
6500F\textsuperscript{TM}- field emission gun (FEG) scanning electron microscope in secondary and back scattered electron imaging modes. SEM coupled with Energy Dispersive Spectroscopic (EDS) analysis was used for qualitative examination of the alloying elements to identify the precipitates in the matrix.

### 3.2.3 Results

Optical microscopy analysis of S690QL1 steel clearly indicates the presence of tempered martensite in a ferritic matrix (see figure 3.1). The sizes of ferritic grains were found to vary between 10 to 20 μm. The samples were etched with LePera solution to investigate the presence of untempered martensite or retained austenite. No untempered martensite or retained austenite was found in the microstructure.

The microstructure also indicates the presence of carbides along the grain boundaries and precipitates and or inclusions in the matrix as shown in figure 3.2. EDS analysis confirms the inclusions to be rich in copper as shown in figure 3.3. The sizes of the copper inclusions were found to be in the range of 3 to 5 μm.

**Figure 3.1:** Optical micrographs of S690QL1 base metal.
3.3 Mechanical and thermal properties

The as-received plates measuring $400 \times 1500 \times 16$ mm$^3$ were flame cut into smaller plates measuring $220 \times 320 \times 16$ mm$^3$. The edges were then sawed to remove the heat affected region from the plasma cutting process. The final dimensions of the plate measured $200 \times 300 \times 16$ mm$^3$ from which samples for mechanical testing were cut.

Figure 3.2: Scanning electron microscopy images of S690QL1 base metal.
Characterisation of S690 steel base metal

(a) Backscattered electron image of S690QL1 base metal showing copper inclusion in the matrix.
(b) EDS analysis confirms the presence of Cu inclusions in the matrix.

Figure 3.3: EDS of the Cu inclusions in S690QL1 base metal.

3.3.1 Micro-hardness

Vickers micro hardness measurements were performed using a square pyramidal shaped diamond indenter with a load of 500 g in a Buehler® micro-hardness testing (MHT) machine. The indentations were analysed using Buehler® Omnimet® MHT software. The hardness is given by

\[ H_{V500gf} = 0.102 \frac{2F \sin \frac{136^\circ}{2}}{d_m^2} \approx 0.1891 \frac{F}{d_m^2}, \]  

(3.1)

where \( F \) is the force, \( d_m \) is the mean diagonal and \( 136^\circ \) is the angle between opposite faces of the pyramidal indenter at the vertex [78]. A minimum distance of 250 \( \mu m \) was maintained between separate indents to ensure that local deformation did not affect subsequent results.

The average hardness of the S690QL1 base metal is \( 260 \pm 3.7 \) \( H_{V500gf} \).

3.3.2 Room temperature tensile strength

Tensile samples (figure 3.4) were prepared from the as-received S690QL1 plates based on the ASTM-E8/E8M-09 [79] standard by Electro-Discharge Machining (EDM) to avoid any possible stressing in the material during mechanical cutting and grinding. Samples 1, 2 and 3 were prepared from the top, the middle and the bottom section respectively, along the thickness direction of the plate. Tensile testing was carried out in an electro-mechanical Zwick Z100 tensile testing machine with a load cell of 100
3.3 Mechanical and thermal properties

kN. Using an extensometer with a gauge length of 20 mm, elongations were recorded for the applied load using data acquisition software TestXpert® II. Testing was carried out on 3 samples to ascertain the tensile properties of the base metal samples.

Figure 3.4: Schematic view of the tensile testing sample, including dimensions in mm.

Figure 3.5: The tensile stress-strain curves for S690QL1 base metal samples.
The tensile behaviour of S690QL1 base metal samples showed a typical stress-strain curve with continuous yielding and a completely ductile failure (figure 3.5). The results show that the average yield stress of the S690QL1 base metal at 0.2 % offset is 828 MPa. With an average ultimate tensile stress of 868 MPa, the base metal showed a maximum average elongation of 9.81 mm with a total tensile strain of 11.8 %. The average Young’s modulus was 210 GPa. Table 3.4 summarises the mechanical properties of the S690QL1 base metal obtained by tensile testing.

### Table 3.4: Tensile properties of S690QL1 base metal samples (UTS - Ultimate tensile stress and YS - Yield stress, in MPa)

<table>
<thead>
<tr>
<th>Sample</th>
<th>UTS, MPa</th>
<th>Tensile strain at UTS, %</th>
<th>YS at 0.2 % strain, MPa</th>
<th>Extension at break, mm</th>
<th>Tensile strain at break, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>858</td>
<td>6.4</td>
<td>815</td>
<td>9.85</td>
<td>12.2</td>
</tr>
<tr>
<td>2</td>
<td>872</td>
<td>6.1</td>
<td>838</td>
<td>9.72</td>
<td>11.1</td>
</tr>
<tr>
<td>3</td>
<td>873</td>
<td>6.4</td>
<td>831</td>
<td>9.85</td>
<td>12.1</td>
</tr>
<tr>
<td>Mean</td>
<td>868</td>
<td>6.3</td>
<td>828</td>
<td>9.81</td>
<td>11.8</td>
</tr>
</tbody>
</table>

### 3.3.3 Elevated temperature tensile strength

Tensile samples for temperature dependent tests (figure 3.6) were prepared from the as-received S690QL1 plates based on the ASTM-E8/E8M-09 [79] standard. Temperature dependent tensile testing was carried out in a 25 kN MTS servo-hydraulic fatigue machine capable of imposing independent temperature and strain profiles on a test specimen. Solid smooth dog-bone specimens were machined and with a final smooth cylindrical gauge length of 22 ± 0.25 mm and a diameter of 6 ± 0.025 mm. A high-frequency induction generator was used for heating. Cooling was accomplished by blowing compressed air from three sides onto the specimen and by thermal conduction into the water-cooled specimen grips. The temperature was measured and controlled by three K-type thermocouples pressed onto the surface of the specimen at the middle of the gauge length. The maximum temperature deviation along the gauge length of the test specimen was ± 10 °C. The contact pressure of the thermocouples was regulated by means of a spring configuration. This led to a good contact with the specimen surface and consistent temperature measurement. The axial strain was measured via an air-cooled, high temperature, ceramic rod extensometer with 12 mm gauge length.

The MTS thermo-mechanical fatigue machine employs a TestStar controller. To design an arbitrary test, MultiPurpose Testware® (MPT) software is used. This is a graphical test programming environment which allows one to define different test
3.3 Mechanical and thermal properties

Figure 3.6: Schematic view of the elevated temperature tensile testing sample, including dimensions in mm.

Figure 3.7: Experimental set up of the elevated temperature tensile tests. 1. Cooling unit. 2. K type contact thermocouple. 3. Induction coil. 4. Ceramic extensometer.

segments, data acquisition processes and trigger processes. MPT has the ability to synchronise multiple control modes, as is required in temperature dependent tensile testing. Parameters that are typically controlled during a test include mechanical strain wave shape and amplitude, temperature wave shape and amplitude, frequency and phase relation between temperature and mechanical strain. The experimental arrangement for the elevated temperature tensile test is shown in figure 3.7.

Testing was carried out at six temperatures between 300 °C and 1000 °C. Each test was repeated 3 times to ascertain the temperature dependent tensile properties of the base metal samples. For temperatures above 500 °C, tests were performed at two different strain rates: SR1 = 2.5 × 10⁻³ s⁻¹ and SR2 = 2.5 × 10⁻⁴ s⁻¹. These strain rates were kept constant during the whole tensile test. The temperature dependent tensile behaviour of S690QL1 is shown in figure 3.8. Above 400 °C, softening sets in and the material show increased dependency on the applied strain rate.
Using the results of the tensile tests, temperature dependent Young’s modulus, $E$ and yield stress, $\sigma_y$ were calculated (figure 3.9).
3.3 Mechanical and thermal properties

![Graph showing temperature dependent Young's modulus and yield stress of S690QL1.]

Figure 3.9: Temperature dependent Young’s modulus, $E$ and yield stress, $\sigma_y$ of S690QL1.

3.3.4 Dilatation properties

A Böhr thermoanalysis DIL805 dilatometer was employed for temperature dependent thermal expansion measurements. Samples were milled to $4 \times 10 \times 2$ mm$^3$ to ensure that no deformation from shear cutting was present in the sample that could influence the dilatometer measurements. S-type thermocouples were attached on the samples, both for controlling the heating of the sample and to obtain the temperature profile of the sample during the dilatometer test. The heating programs employed are summarised in table 3.5.

<table>
<thead>
<tr>
<th>Table 3.5: Heating programmes for dilatometry.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
</tr>
<tr>
<td>--------</td>
</tr>
<tr>
<td>1</td>
</tr>
<tr>
<td>2</td>
</tr>
</tbody>
</table>

Because the measuring equipment has an unknown influence on the expansion of the samples during heating, due to temperature variation in the measuring probes, a platinum sample was measured with similar heating and cooling programs to those for
S690QL1 samples. By comparing theoretical thermal expansion of platinum with the measured data, the expansion and shrinkage of the measurement probe was obtained and was subtracted from the S690QL1 steel measurements.

The thermal expansion coefficient for the S690QL1 base metal was carried out with the given heating and cooling rates and repeated three times to ascertain the dilatation properties. The different cooling rates were used to promote bainitic (figure 3.10) and martensitic (figure 3.11) transformations in samples 1 and 2 respectively. In figures 3.10 and 3.11, $A_{c1}$ represents austenite nucleation temperature during heating, $A_{c3}$ represents the temperature at which the microstructure is fully austenitic, $\alpha_{25}$ is the coefficient of expansion of the ferritic phases averaged within a temperature range of 25 to 725 $^\circ$C, $\gamma_{600}$ is the coefficient of expansion of the austenitic phases averaged within a temperature range of 600 to 1000 $^\circ$C, $\Delta \epsilon_{25}^{\alpha,\gamma}$ is the difference of thermal strains between $\alpha$ and $\gamma$ phases at the reference temperature of 25 $^\circ$C, $B_s$ is the temperature at which bainite starts to nucleate, $M_s$ is the temperature at which martensite starts to nucleate.
3.3 Mechanical and thermal properties

(a) Temperature dependent thermal expansion coefficient for a heating rate of $10 \, ^\circ C \, s^{-1}$ and a cooling rate of $10 \, ^\circ C \, s^{-1}$ resulting in bainitic transformation.

(b) Microstructure of sample 1 from table 3.5 etched with LePera solution. Bainite appears as brown and ferrite appears as tan colour in the microstructure.

(c) Microstructure of sample 1 from table 3.5 etched with 5 % Nital solution. Bainite appears as brown and ferrite appears as white in the microstructure.

Figure 3.10: Dilatation and final microstructure of sample 1 from table 3.5.
Characterisation of S690 steel base metal

(a) Temperature dependent thermal expansion coefficient for a heating rate of 10 °C s\(^{-1}\) and quenching with a cooling rate of 220 °C s\(^{-1}\) resulting in martensitic transformation.

(b) Microstructure of sample 2 from table 3.5 etched with LePera solution. Martensite appears as bright white and ferrite appears as tan colour in the microstructure.

(c) Microstructure of sample 2 from table 3.5 etched with 5 % Nital solution. Martensite appears as brown and ferrite appears as white in the microstructure.

Figure 3.11: Dilatation and final microstructure of sample 2 from table 3.5.
3.4 Concluding remarks

It is notable that after the material was subjected to heating and cooling, there was a difference of thermal strains of 1 % between \( \alpha \) and \( \gamma \) phases at the reference temperature of 25 °C, which might have a large influence on the residual stress after welding of S690QL1 steel. Table 3.6 summarises the dilatation properties of the S690QL1 base metal obtained by applying the thermal cycles to the base metal.

Table 3.6: *Dilatation and micro hardness properties.*

<table>
<thead>
<tr>
<th>Sample</th>
<th>( A_c_1 ) °C</th>
<th>( A_c_3 ) °C</th>
<th>( \alpha_{\alpha_{25}} \times 10^{-6} ) °C(^{-1} )</th>
<th>( \alpha_{1000,^\circ C} \times 10^{-6} ) °C(^{-1} )</th>
<th>( \Delta \epsilon_{\alpha,\gamma} %)</th>
<th>( B_s ) °C</th>
<th>( M_s ) °C</th>
<th>( H_{V500gf} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>750</td>
<td>840</td>
<td>15.9</td>
<td>24.23</td>
<td>1</td>
<td>595</td>
<td>308 ± 19</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>750</td>
<td>840</td>
<td>15.4</td>
<td>24.19</td>
<td>1</td>
<td>464</td>
<td>407 ± 13</td>
<td></td>
</tr>
</tbody>
</table>

During welding, the heating rate is much higher than the one used here. The actual heating rate during welding lies in a range between 1000 and 2000 °C s\(^{-1} \), but such high rates were not reproducible in a dilatometer. The disadvantage of high heating and cooling rate is that the experiments become less repeatable.

3.4 Concluding remarks

Detailed microstructural, thermal and mechanical characterisation of S690QL1 base metal is presented in this chapter. The base metal microstructure indicates the presence of tempered martensite in a ferritic matrix with carbides along the grain boundaries and copper inclusions in the matrix. The mechanical properties of the base metal are measured both at room temperature and at elevated temperatures and at two different strain rates to calculate temperature dependent Young’s modulus and yield stress. The thermal expansion coefficients of ferrite and austenite were also measured for two separate cooling rates using a dilatometer.
Chapter 4

Welding and characterisation of weld metal

In the previous chapter, a detailed description of the base metal characterisation has been presented. This chapter gives an overview of the welding process, welding equipment and the characterisation of the welding process and the weld metal.

4.1 Gas metal arc welding (GMAW)

In GMAW, an arc is established between the weld pool and the continuous filler metal electrode. The arc and the molten metal in the weld pool are protected from the environment by a shielding gas. Shielding gases are available in a wide range, from the use of a single gas, such as Ar, He or CO$_2$, to mixtures of two or more gases, which can be used to improve the weld quality and the process stability [80, 81]. A schematic diagram of GMAW is shown in figure 4.1.

An important characteristic of the GMAW process can be described by the growth and the subsequent detachment of liquid droplets at the electrode tip. This characteristic is strongly influenced by the welding conditions, such as the welding current, the arc voltage, the electrode diameter the electrode composition, the electrode extension and the shielding gas. In the case of a low current and a short arc, metal transfer takes place in a short-circuiting mode (figure 4.2a). Metal is transferred from the electrode to the work piece only during a period when the electrode is in contact with the weld pool. No metal is transferred across the arc gap. At a low welding current and with a relatively long arc, a globular transfer mode occurs. Globular transfer is characterised by a drop size with a diameter greater than that of the electrode. The large molten drops are formed, which are detached under to the force of gravity (figure 4.2b). With Ar rich shielding gas and high welding current (above a certain
threshold current), it is possible to produce a very stable, spatter free spray metal transfer mode, where very small droplets are formed and detached axially across the arc (figure 4.2c). Because of the high welding current involved in the spray transfer mode, the heat input is high and the weld pool is large. The spray transfer mode results in a highly directed stream of discrete drops that are accelerated by the arc forces to velocities which overcome the effects of gravity. This mode also offers a high deposition rate.

GMAW can also be carried out in the pulsed-current mode during which, the welding current is switched periodically between a low base current and a high peak current. The low base current is used to maintain the arc, whereas the peak current is used to melt and detach the droplet as well as to melt the workpiece. With this process, stable spray transfer can be achieved with a low heat input.
4.1 Gas metal arc welding (GMAW)

4.1.1 Specimen size

The dimensions, weld design and joint design of S690QL1 steel plates used for welding experiments in this work are shown in table 4.1. In order to achieve a full penetration weld, a “V” groove joint was used with the dimensions shown in figure 4.3. Tack welds were made at the starting and the finishing points of the “V” groove to prevent relative plate movements.

<table>
<thead>
<tr>
<th>Material</th>
<th>Dimensions [mm × mm × mm]</th>
<th>Welding process</th>
<th>Weld design</th>
<th>Joint design</th>
</tr>
</thead>
<tbody>
<tr>
<td>S690QL1</td>
<td>260 × 100 × 16</td>
<td>GMAW</td>
<td>Butt weld</td>
<td>V groove</td>
</tr>
</tbody>
</table>

![Figure 4.3: The V groove joint design used during welding of 16 mm thick S690QL1 steel.](image)

4.1.2 Clamping system

A clamping system with five vertical hydraulic clamps (AMF 6958SU-16) on each side of the workpiece was used. Figure 4.4 shows the clamping system. An air operated hydraulic pump (AMF 6904-20), requiring at least 5 bar air pressure, applies pressure. The maximum oil pressure allowed at the clamp is 150 bar which corresponds to an effective clamping force of 4.8 kN (490 kg). The force-pressure ratio, \( \frac{F}{p} \), is 32 N/bar. The oil pressure is controlled by a pressure valve (AMF 6917-1) connected between the pump and the clamps. The clamps are controlled by an electrical valve (AMF 6910-06-04) which permits control of clamping and release times for each clamp. In the present work, the clamps were released when the temperature of the welded plate reached room temperature.
4.1.3 Welding equipment

The welding power source used for GMAW welding was a Cloos Quinto Profi 503 welding machine. The power generator is used in combination with a controller and wire feed unit. The welding rectifiers are fan cooled and fitted with a thermal cut-out to prevent overheating. The GMAW power source is shown in figure 4.5.
4.1 Gas metal arc welding (GMAW)

4.1.4 Filler wire

Böhler UNION NiMoCr filler wire with 1.2 mm diameter was used. According to AWS A5.28 specification [82], the electrode falls under the ER100S-G classification, with chemical composition given in table 4.2.

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Ni</th>
<th>Mo</th>
<th>Cr</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.08</td>
<td>0.6</td>
<td>1.7</td>
<td>1.5</td>
<td>0.5</td>
<td>0.2</td>
<td>bal.</td>
</tr>
</tbody>
</table>

4.1.5 Welding condition

The range of preheating temperature depends on the plate thickness. For 16 mm thick plates, the preheating temperature and the interpass temperatures range from 25 °C to 225 °C and 100 °C to 150 °C [83] respectively. Preheating was not implemented in the current case. Welding was carried out at room temperature, i.e., 25 °C. The interpass temperatures were measured in accordance with ISO 13916 [84]. The interpass temperature was measured on the weld metal by a contact thermocouple before the passage of the arc for the next pass and was maintained between 100 and 150 °C.

The shielding gas used for the GMAW process was 85 % Ar-15 % CO₂. This mixture of shielding gas gives a stable arc, low spatter and improves the weld bead profile. The shielding gas flow rate was 15 l min⁻¹ for the first pass and 16 l min⁻¹ for the subsequent passes. Ceramic strips with rounded groove were used for backing and are shown schematically in figure 4.6a. The electrode was tilted at 100 ° from the welding direction as shown in figure 4.6b.

The welding conditions for the respective passes are listed in table 4.3. For each pass the welding torch was re-positioned (see figure 4.7 and table 4.4). Figure 4.7 shows a schematic bead sequence as applied in the experiments.
Welding and characterisation of weld metal

Figure 4.6: a.) Ceramic backing used in GMAW process with rounded groove; b.) Orientation of the filler wire with respect to the welding direction.

Table 4.3: Welding conditions for the respective passes.

<table>
<thead>
<tr>
<th></th>
<th>Pass 1</th>
<th>Pass 2</th>
<th>Pass 3</th>
<th>Pass 4</th>
<th>Pass 5</th>
<th>Pass 6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal transfer</td>
<td>Short-circuit</td>
<td>Spray</td>
<td>Spray</td>
<td>Spray</td>
<td>Spray</td>
<td>Spray</td>
</tr>
<tr>
<td>Electrode angle [°]</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Contact tube workpiece distance [mm]</td>
<td>9</td>
<td>9</td>
<td>9</td>
<td>9</td>
<td>9</td>
<td>9</td>
</tr>
<tr>
<td>Wire feed speed [m min⁻¹]</td>
<td>6.2</td>
<td>11</td>
<td>11</td>
<td>11</td>
<td>11</td>
<td>11</td>
</tr>
<tr>
<td>Average welding current [A]</td>
<td>220</td>
<td>305</td>
<td>305</td>
<td>305</td>
<td>305</td>
<td>305</td>
</tr>
<tr>
<td>Average welding voltage [V]</td>
<td>23.5</td>
<td>31</td>
<td>31</td>
<td>31</td>
<td>31</td>
<td>31</td>
</tr>
<tr>
<td>Welding speed [mm s⁻¹]</td>
<td>4.5</td>
<td>8.5</td>
<td>8.5</td>
<td>8.5</td>
<td>8.5</td>
<td>8.5</td>
</tr>
<tr>
<td>Energy/length [kJ mm⁻¹]</td>
<td>1.15</td>
<td>1.11</td>
<td>1.11</td>
<td>1.11</td>
<td>1.11</td>
<td>1.11</td>
</tr>
<tr>
<td>Shielding gas flow rate [l min⁻¹]</td>
<td>15</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td>16</td>
</tr>
<tr>
<td>Shielding cup diameter [mm]</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
</tbody>
</table>
4.2 Welding thermal cycle

Figure 4.7: The red dots represent the positioning of the welding torch for each pass. The number in red next to the red dots represents the pass number. A schematic view of the beads is superimposed over the torch positions.

Table 4.4: Torch positions for the respective passes.

<table>
<thead>
<tr>
<th>Pass number</th>
<th>Distance from the weld centre line Y, mm</th>
<th>Distance from the rear surface of the work piece Z, mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>9</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>12</td>
</tr>
<tr>
<td>3</td>
<td>-3</td>
<td>14</td>
</tr>
<tr>
<td>4</td>
<td>2.5</td>
<td>16</td>
</tr>
<tr>
<td>5</td>
<td>-5</td>
<td>18</td>
</tr>
<tr>
<td>6</td>
<td>4</td>
<td>18</td>
</tr>
</tbody>
</table>

4.2 Welding thermal cycle

The thermal profile of the welding process dictates the microstructural evolution in the heat affected (HAZ) and fusion zone (FZ). During the welding experiments, the temperature of the workpiece was measured by means of thermocouples. Standard $\phi$ 0.25 mm glass insulated k-type thermocouples (Chromel-Alumel) were used to measure temperature within a range of -270 to 1372 °C. The measurement accuracy is $\pm 2.2$ °C [85]. The thermocouple wires were welded to the cleaned surface of the workpiece at different positions using a Labfacility L60+ electric discharge thermocouple welder.
The temperature measurements were carried out at the rear surface of the workpieces (see figure 4.8). The thermocouples were installed perpendicular to the welding line. In order to make sure that two wires of thermocouple cannot have contact with each other, ceramic tubes were used. The exact positions of thermocouples were measured after welding.

![Figure 4.8: Thermal cycle on the rear surface during GMA welding of S690QL1 steel at different distances from the weld centre line.](image)

The thermocouples were spot welded on the rear surface of the positive “Y” direction of figure 4.7. The effect of the torch re-positioning is clearly visible on the peak temperatures measured on the rear surface (see figure 4.8). Due to the thermocouple placement in the positive “Y” direction, the peak temperatures in passes 4 and 6 are higher than in passes 3 and 5.

### 4.3 Microstructural characterisation of the welded S690QL1 steel plates

A cross-section of the weld is shown in figure 4.9a. Different regions can be distinguished in the cross section of the welds: the unaffected base metal region, a tempered zone, a partially transformed zone, a re-crystallised zone, a grain growth region and
4.3 Microstructural characterisation of the welded S690QL1 steel plates

finally the weld region (fusion zone). Figure 4.9b shows a schematic representation of these regions. The fusion zone (FZ) consists primarily of acicular ferrite and occasional martensite and bainite (figure 4.10a). The grain growth region \( i.e. \); the coarse grained heat affected zone (CGHAZ) consists of Widmanstätten ferrite and bainite (figure 4.10b and 4.11). The fine grained heat affected zone (FGHAZ) is the region consisting of an equiaxed fine grained structure containing dislocation substructures as shown in figure 4.10c. The inter-critical heat affected zone (ICHAZ) consists of pearlite and ferrite as shown in figure 4.10d. The fusion zone shows the presence of inclusions (figure 4.12), mainly with a random distribution. The average size of the inclusions in the fusion zone is approximately 1 µm. The presence of hard inter-metallic inclusions in the fusion zone surrounded by soft ferritic grains is detrimental to the elongation, strength and toughness of the welds. Inclusions can act as cleavage crack initiation sites. It is reported that oxide inclusions tend to affect fracture toughness and fatigue properties of the welds [86–88].

![Figure 4.9: a.) Macroscopic image of the weld cross-section; b.) Schematic representation of the different regions in a weld.](image)

4.3.1 Inclusion formation in fusion zones

The fusion zones in S690QL1 steels contain complex inclusions. These inclusions are generally found at the grain boundaries and occasionally in the grain interiors. Figure 4.12 gives an overview of the inclusion distribution in the fusion zone of a GMA welded S690QL1 steels. The average size of the inclusions in the fusion zone is approximately 1 µm.

Morphological analysis of the inclusions in the fusion zones of GMA welded S690QL1
Welding and characterisation of weld metal

Figure 4.10: Microstructure of a.) fusion zone consisting of acicular ferrite and traces of martensite and bainite b.) coarse grained heat affected zone consisting of Widmanstätten ferrite and bainite c.) fine grained heat affected zone consisting of equiaxed fine grained structure containing dislocation substructures and d.) inter-critical heat affected zone consisting of pearlite and ferrite.

Steels using electron microscopy indicates that the inclusions consists of a primary core (figure 4.13). The presence of strong deoxidisers such as silicon and aluminium, as in the case of S690QL1 steels under investigation, and copper from the coatings of the welding filler wires, leads to the formation of oxide and sulphide inclusions during welding [89, 90]. The reaction between the dissolved alloying elements in the weld pool with the available oxygen, nitrogen, carbon and sulphur forms non-metallic inclusions. Due to the strong affinity for oxygen and sulphur, Al and Cu readily form oxides and sulphides, respectively during welding. This can be seen from figures 4.14
4.3 Microstructural characterisation of the welded S690QL1 steel plates

Figure 4.11: Widmanstätten ferrite in the coarse grained heat affected zone.

Figure 4.12: S690QL1 GMA welded; the black circle indicates inclusions in the fusion zone (FZ).

and 4.15, where the energy dispersive elemental analysis of the inclusions present shows a higher concentration of aluminium (figure 4.14b) and copper (figure 4.15b). The aluminium oxide inclusions are primarily found in the fusion boundary, while the copper sulphide inclusions are found in the fusion zone.

The observation of inclusions in the fusion zone of low alloyed steel welds is an extensively researched topic with studies on the formation mechanism of inclusions, their effects on the subsequent phase transformations and final mechanical properties of the welds [89–93]. The non-metallic inclusions formed in the weld pool favour the formation of intergranular acicular ferrite by acting as potential nucleation sites. Increased volume fractions of acicular ferrite optimise the combination of strength and toughness. The thermal cycle of the welding process, inclusion size distribution, num-
Welding and characterisation of weld metal

Figure 4.13: Secondary electron images indicating the presence of inclusions in GMA welded S690QL1 steels.

Figure 4.14: An inclusion in a GMA welded S690QL1 steel fusion boundary showing an aluminium rich core.

ber density, volume fraction and composition of the inclusions strongly influences the formation of acicular ferrite [90, 94]. The formation of acicular ferrite in the GMA welded S690QL1 steels is confirmed by microstructural analysis.

There are four sources of oxygen for the formation of oxide inclusions in the fusion zone of steels during welding [95]:

- Oxygen present in the shielding gas or in the welding atmosphere leading to oxidation at the weld pool surface resulting in oxide inclusions.

- Oxide inclusions arising from an oxidised plate surface prior to welding. The oxide scales present at the plate surface tends to float, transported with the motion of the weld pool and be retained during subsequent solidification.
4.4 Micro-hardness variation in welded S690QL1 steels

Figure 4.15: An inclusion in a GMA welded S690QL1 steel fusion zone showing copper and sulphur enrichment.

- Inclusions forming from the deoxidisers in the filler metal while welding is carried out using filler metal additions and
- The interaction of oxygen dissolved in the parent material combining with alloying elements to form oxide inclusions.

In the present work, the oxide scales on the surface of the plate close to the groove were removed by grinding which eliminates the second possibility for the formation of non-metallic inclusions. In GMA welding, commercial purity shielding gas was used which generally contain residual oxygen. The interaction between the aluminium and the available oxygen during welding forms oxide inclusions in the weld pool of the S690QL1 steels. The filler wires used in the present work were copper coated which were added into the weld pool during welding. This results in the formation of the copper sulphide inclusions in the fusion zone.

4.4 Micro-hardness variation in welded S690QL1 steels

Micro-hardness measurements provide first hand information about the variation in mechanical properties of welded steels across weld zones. Measurements were carried out transverse to the weld centre line over a surface of $15 \times 29 \text{ mm}^2$ in steps of 1 mm in the horizontal direction and 0.5 mm in the vertical direction. Figure 4.16 shows the micro-hardness variation over the weld surface.
The overall hardness in the fusion zone ranges between 240 and 280 $H_V_{500gf}$, which primarily consists of acicular ferrite. The hardness rises to between 280 and 320 $H_V_{500gf}$ in the weld metal close to the fusion boundary. This could be due to the formation of martensitic structures in this region because of the faster cooling rates. In the fusion zone, the heterogeneity in the grain sizes and the presence of martensite and acicular products dictates the variation of hardness in the solidified microstructure. The hardness values drop again to a range between 200 to 240 $H_V_{500gf}$ close to the fusion boundaries due to grain coarsening in the heat affected zone combined with the effects of the formation of polygonal ferrite in the inter-critical heat affected zone.

4.5 Summary

The welding parameters and conditions, the thermal cycle of the welding process and the detailed microstructural characterisation of S690QL1 weld metal is presented in this chapter. The weld metal shows a complex microstructure along with the formation of inclusions.
Chapter 5

*In-situ* synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1*†

During welding, significant stresses are produced by differential thermal expansion of material under a non-uniform thermal field. As it cools, material close to the source of heating is constrained by material further away, generally resulting in high tensile residual stresses in an area around the weld seam. These tensile stresses have a negative impact on mechanical properties of the joint, aiding both fatigue cracking [61] and brittle fracture [96]. They also cause distortion of the component, a problem which is especially evident in the welding of thin plates, which are prone to buckling. The presence of a thermal gradient gives rise to Type I and Type II residual stresses [97] resulting in macroscopic distortion. Several *in-situ* and post weld thermo-mechanical straining methods may be used to mitigate the residual stresses [11, 98–101]. To enable the best use of such techniques, it is necessary to characterise the transient thermal-mechanical stress and strain fields in the HAZ due to the weld thermal cycles. This will allow the design of processes which can more precisely counter act the thermal-mechanical stress fields.


The primary objective of this chapter is to determine the thermal and mechanical properties of high strength (830 MPa yield stress) quenched and tempered S690QL1 structural steel. The specific aims are:

- To determine the diffraction elastic moduli by means of *in-situ* tensile testing of S690QL1 at elevated temperatures.

- To determine the thermal anisotropy during phase transformation by continuous cooling.

### 5.1 Experimental procedure

#### 5.1.1 Sample preparation

Small cuboidal blocks were cut from the as-received S690 steel plate which were homogenised at 950 °C for 48 hours in a vacuum chamber. The blocks were then annealed in a salt bath at 890 °C for 15 minutes in order to obtain a completely austenitic microstructure and quenched in cold water. This was followed by tempering in another salt bath at 635 °C for 20 minutes followed by air cooling. This heat treatment was given to achieve a tempered martensite microstructure for the tensile specimens, similar to the base metal microstructure of S690QL1 steels. The block and the sample orientation with respect to the rolling direction of the plate is shown in figure 5.1. Tensile specimens (figure 5.2) having a 1×1 mm² square cross section and a gauge length of 16 mm were electro-discharge machined from heat treated blocks. The loading axis was chosen parallel to the transverse direction (TD).

![Figure 5.1: Block and the sample orientation with respect to the rolling direction of the as-received plate.](image-url)
5.1 Experimental procedure

![Diagram of experimental arrangement]

Figure 5.2: Sketch of the experimental arrangement of in-situ thermo-mechanical studies by synchrotron based high energy X-ray diffraction with sample dimensions. TD and ND correspond to the transverse and normal direction of the rolled sheet.

5.1.2 In-situ synchrotron diffraction

In-situ synchrotron diffraction experiments were carried out at the ID11 beam line of the European Synchrotron Radiation Facility (ESRF) in Grenoble, France. To evaluate the various properties such as temperature dependent diffraction elastic constants (DECs), thermal expansion coefficients, phase fraction evolution and temperature dependent strain evolution, several tests were performed using an Instron/NPL electro-thermal mechanical testing (ETMT, [102]) machine. This comprised a 3 kN screw driven mechanical testing stage, with thermal cycles being achieved by Ohmic heating due to a direct current passing through the sample. Control was achieved by means of feedback from an R-type thermocouple, discharge welded to the centre of the gauge length.

The experimental arrangement is shown in figure 5.2. To limit the effects of temperature gradients due to the parabolic temperature distribution generated along the gauge length of the samples, slits positioned in the incident beam path were used to define a 677 (vertical) × 761 (horizontal) × 1000 (normal) µm³ diffraction gauge volume at the centre of the sample. To ensure correspondence between the temperature measured by the thermocouple and that of the gauge volume, the beam centre was aligned in the centre of the sample directly above the centre of the thermocouple bead. With this configuration, a temperature difference of no more than 1.5 °C is expected at temperatures of 600 °C across the 761 µm horizontal gauge length. To prevent decarburisation/oxidation during the thermal cycles, the ETMT chamber was continuously flushed with helium gas at a flow rate of 1 l min⁻¹.
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

Figure 5.3: Photograph of the ETMT machine on the ID11 beam line at the ESRF facility in Grenoble, France. The sample environmental chamber is photon transparent, so that X-rays can pass through the back of the chamber (from behind the machine on the left), and then pass through the sample gauge volume before reaching the detector which lies on the other side of the ETMT. (1) Load cell (2) Sample clamps (3) Thermocouple (4) Sample (5) Motor (6) Clamping system for the photon transparent lid.

The instrument parameters (sample-detector distance, position of the straight-through beam and the tilt of the detector) of the 3-D X-ray diffraction microscope were determined using a CeO$_2$ calibrant placed on each sample prior to the tests. The photon energy was 78.7 keV ($\lambda = 0.157649$ Å), which enabled the transmission geometry shown in figure 5.2 to be used. Diffraction data was acquired with a FReLoN 2-D CCD detector [103] mounted $\sim$273 mm behind the sample. The detector size is 2048 $\times$ 2048 pixels, each corresponding to 46.8 $\times$ 48.0 µm$^2$. The 2-D diffraction patterns were continuously recorded using an exposure time of 0.08 s. Including data recording time, a time resolution of $\sim$0.3 s was achieved and powder diffraction rings were therefore recorded at $\sim$0.3 s intervals.
5.1 Experimental procedure

5.1.3 Characterisation of diffraction peaks

The 1-D diffraction images acquired after integrating over the azimuthal angles usually consist of a large number of pixels of background intensity, interspersed with a considerably smaller number of pixels of signal intensity (i.e. intensity higher than the background) on diffraction rings from the diffracting grains. For diffraction data with peak overlap, the determination of peak characteristics by fitting a diffraction image as a whole with multiple diffraction peaks can be computationally very intensive because (i) a significant number of pixels (background) are not of interest and thus need not be processed; and (ii) depending on the number of peaks in the diffraction image, the number of parameters to be fitted can be very large. In the methodology presented here, these issues are avoided as follows: (i) background pixels in the diffraction image are removed by applying a threshold to the diffraction image; (ii) The 1-D diffraction images are then grouped into ‘regions’ (a smaller set than all the peaks present in the diffraction image); and (iii) estimates of the parameters to be fitted are obtained using information about the peaks in the ‘regions’. The procedure for the determination of the characteristics of the peaks is then divided into two parts:

- A peak-searching algorithm to carry out the thresholding operation, to determine the number of peaks in each set of 1-D diffraction pattern using the number of maxima and to determine the approximate characteristics of the peaks (integrated intensity and centre-of-mass position).
- A peak-fitting routine to refine the peak characteristics by fitting the peak shape with a symmetric two-dimensional function.

5.1.4 Peak searching

The diffraction images are first corrected for background (a combination of the dark current of the detector and the background from the sample), the flood of the detector (defined as the heterogeneity in the pixel response to photons) and the beam current. A linear background is subtracted from the whole profile. The peaks are then fitted separately (see section 5.1.5). For this the 2θ range of the peak is determined by finding the local minima on either side of the theoretical peak position, which changes with temperature.

5.1.5 Peak fitting

The program for fitting requires an input from peak searching. The algorithm works by fitting peaks in the 1-D diffraction image. The output of the program contains the refined parameters of the peaks along with the time, temperature and load information corresponding to each 1-D diffraction image. In the present framework, the results of peak searching are used only as initial values for fitting, and thus the accuracy of the values of the peak characteristics obtained from peak searching is not...
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

paramount. A first estimate of the peak height is determined by finding the maximum in this 2θ range. A first estimate of the peak position is calculated from the centre of mass of the peak.

Minimisation: The algorithm is designed to fit peaks using a one-dimensional pseudo-Voigt function, although a Gaussian or a Lorentzian function can also be used. The minimised function in the fitting program is

\[
\sum \left( I - \left\{ I_0 + \sum_n A_n \left[ \mu_n L_n + (1 - \mu_n) G_n \right] \right\} \right)
\]

\[ (5.1) \]

i.e. the sum of the squared differences between the recorded intensity in the diffraction image and the intensity according to the parameters of the peaks obtained using the one dimensional pseudo-Voigt profile. I is the recorded intensity of the \( x_i^{th} \) pixel on the detector. \( L_n \) and \( G_n \) are the Lorentzian and Gaussian contributions per peak, respectively, given by

\[
L_n = \frac{1}{1 + \left( \frac{x_i - x_{nc}^n}{w^n} \right)^2}
\]

\[ (5.2) \]

and

\[
G_n = \exp \left[ -0.5 \left( \frac{x_i - x_{nc}^n}{w^n} \right)^2 \right]
\]

\[ (5.3) \]

The parameters that are refined are the background intensity \( I_0 \), the maximum intensity for each peak \( A_n \), the Lorentz fraction \( \mu \), the coordinate position of the centre \( x_{nc}^n \) and the width of the \( n^{th} \) peak \( w^n \) in each search region.

5.1.6 Implementation and workflow

The algorithm was implemented in MATLAB®. The minimisation of equation 5.1 is carried out using the function \texttt{lsqnonlin}\(^\dagger\) inbuilt in MATLAB®. Although \texttt{lsqnonlin} is a local minimisation routine, it has been observed that initial guesses of the parameters obtained from peak searching are a good starting point.

For each 1-D diffraction image, the algorithm first inputs the list of search regions in that diffraction image and the parameters of the search regions. Then for each search region a linear background is subtracted. The fitting routine is then called to

\[^\dagger\text{http://www.weizmann.ac.il/matlab/toolbox/optim/lsqnonlin.html}\]
minimise the function in equation 5.1 and refine the five parameters per peak and the background intensity using the initial guesses obtained from peak searching (section 5.1.4). The integrated intensity of a peak is calculated in two ways: (i) the area under the pseudo-Voigt peak with the fit parameters for the peak and (ii) integrating the raw intensity data of each pixel from the 1-D diffraction image.

5.2 Temperature dependent plane specific elastic constants in S690QL1 steel

5.2.1 Introduction

The mechanical behaviour of a wide variety of materials and in particular the fatigue performance, is influenced to a great extent by the presence of residual stresses [18–20]. Hence, it is important to quantify the residual stress state within a component with accuracy. There are various techniques to quantify the residual stress in a material; e.g., destructive methods like bulk sectioning [104] and hole drilling [105], and non-destructive methods, which are often based upon diffraction theory [106–109].

The application of such methods almost always necessitates estimation of the residual stresses from some measure of the local elastic strain field. For destructive methods the bulk elastic properties of the material under consideration must be accurately known. The degree of complexity increases when using diffraction methods and monochromatic sources of radiation. Estimation of the residual stress state from lattice strains associated with a specific \{hkl\} reflection demands the use of the corresponding plane specific diffraction elastic constants (DECs), which in general differ from the bulk elastic constants. This is particularly important for materials that are elastically anisotropic. In section 5.2, the temperature dependent plane specific diffraction elastic constants (DECs) of a high strength quenched and tempered structural steel, S690QL1, have been determined with a considerable degree of precision. For this purpose, in-situ tensile tests have been carried out at different temperatures in a high energy X-ray diffractometer. The data reported are of considerable significance, since they allow an accurate determination of the residual stress states from the measured local d-spacings between lattice planes.

5.2.2 Thermal mechanical cycles

All the specimens were heated at a rate of 10 °C s\(^{-1}\) to 25 °C, 100 °C, 200 °C, 300 °C, 400 °C, 500 °C and 600 °C and maintained for 10 s. During each heating cycle and isothermal dwell, the ETMT was run under load control with a set point of zero load to permit free thermal expansion of the sample. After the isothermal dwell, the machine was switched to displacement control and one end of the sample was pulled with a displacement speed of 0.1 mm min\(^{-1}\). The samples were pulled for 0.96 mm (6 % macro strain) after which the samples were quenched to room temperature.

5.2.3 Data analysis

The raw diffraction images were processed using Fit2D image processing software, including corrections for the spatial distortion and detector efficiency along with subtraction of the background signal [110]. No obvious evidence of textural effects were observed from the 2-D diffraction patterns. The Debye Scherrer rings until the \{220\} plane of ferrite were fully captured in the field of view of the detector. The plane specific DEC was calculated from the change in \(d\)-spacing of the constituent phase oriented parallel and perpendicular to the loading direction. The evolution of these two families of grains has been determined by studying a specific angular section of 15° (352.5° to 7.5° and 172.5° to 187.5° for the parallel and 82.5° to 97.5° and 262.5° to 277.5° for the perpendicular direction - see figure 5.4) on the detector [111]. This angular range was chosen to sample a sufficient number of grains to obtain a statistically relevant signal. A 1-D pattern was obtained for these zones of the 2-D patterns. A fit of individual reflections to a pseudo-Voigt profile function (described in section 5.1.3) was used in order to study the stress partitioning between subsets of grains having specific \{hkl\} plane-normals oriented parallel and perpendicular to the loading direction [112].
5.2 Temperature dependent plane specific elastic constants in S690QL1 steel

Figure 5.4: (a) 2-D X-ray diffraction pattern. (b) Schematic of the masks created on the 2-D X-ray diffraction pattern to study specific angular section of 15° on the detector. The \{110\}, \{200\}, \{211\} and \{220\} ferrite reflections are indicated in the figure. LD indicates the loading direction.

5.2.4 Results

The lattice strain \( \varepsilon_{hkl} \) is defined as:

\[
\varepsilon_{hkl} = \frac{d_{hkl} - d^0_{hkl}}{d^0_{hkl}} \quad (5.4)
\]

where \( d_{hkl} \) is the \( d \)-spacing of the diffracting \( \{hkl\} \) planes and \( d^0_{hkl} \) is the unstrained value at the corresponding temperature after subtracting the contribution of the thermal expansion. The lattice strains were calculated at 25 °C, 100 °C, 200 °C, 300 °C, 400 °C, 500 °C and 600 °C. Significant variations in the stiffness of all the \( \{hkl\} \) planes are observed for the parallel and perpendicular orientations. The lattice strains at different temperatures are presented in figure 5.5.
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

Figure 5.5: Lattice strain of the ferritic matrix as a function of the macroscopic stress for different \{hkl\} reflections oriented parallel (dotted lines) and perpendicular (solid lines) to the tensile load at (a) 25 °C, (b) 100 °C, (c) 200 °C, (d) 300 °C, (e) 400 °C, (f) 500 °C and (g) 600 °C.
5.2 Temperature dependent plane specific elastic constants in S690QL1 steel

The plane specific DEC at different temperatures for the ferritic matrix are presented in figure 5.6 and summarised in table 5.1. The errors shown in figure 5.6 correspond to the standard errors from the regression analyses and the fitting of the individual reflections to pseudo-Voigt profile functions.

![Graphs showing temperature dependent plane specific elastic constants](image)

**Figure 5.6:** Plane specific (a) Young’s modulus, $E$ and (b) Poisson ratio $\nu$ at different temperatures.
*In-situ* synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

Table 5.1: *Experimentally measured plane specific Young’s modulus, E and Poisson ratio ν at different temperatures.*

<table>
<thead>
<tr>
<th>Temperature</th>
<th>E, GPa</th>
<th>ν</th>
</tr>
</thead>
<tbody>
<tr>
<td>25 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>213.3±0.6</td>
<td>0.244±0.003</td>
</tr>
<tr>
<td>{200}</td>
<td>169.3±0.6</td>
<td>0.295±0.003</td>
</tr>
<tr>
<td>{211}</td>
<td>221.5±0.6</td>
<td>0.227±0.003</td>
</tr>
<tr>
<td>{220}</td>
<td>218.2±0.6</td>
<td>0.239±0.003</td>
</tr>
<tr>
<td>100 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>202.8±0.9</td>
<td>0.252±0.002</td>
</tr>
<tr>
<td>{200}</td>
<td>145.8±0.5</td>
<td>0.237±0.006</td>
</tr>
<tr>
<td>{211}</td>
<td>214.5±1.0</td>
<td>0.224±0.003</td>
</tr>
<tr>
<td>{220}</td>
<td>216.4±0.9</td>
<td>0.150±0.016</td>
</tr>
<tr>
<td>200 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>200.2±0.5</td>
<td>0.246±0.005</td>
</tr>
<tr>
<td>{200}</td>
<td>147.8±0.4</td>
<td>0.275±0.002</td>
</tr>
<tr>
<td>{211}</td>
<td>203.9±0.5</td>
<td>0.220±0.002</td>
</tr>
<tr>
<td>{220}</td>
<td>202±0.7</td>
<td>0.235±0.004</td>
</tr>
<tr>
<td>300 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>200.2±0.4</td>
<td>0.253±0.002</td>
</tr>
<tr>
<td>{200}</td>
<td>143.7±0.4</td>
<td>0.304±0.002</td>
</tr>
<tr>
<td>{211}</td>
<td>196.5±0.5</td>
<td>0.251±0.001</td>
</tr>
<tr>
<td>{220}</td>
<td>199.3±0.6</td>
<td>0.250±0.002</td>
</tr>
<tr>
<td>400 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>191.9±0.5</td>
<td>0.304±0.003</td>
</tr>
<tr>
<td>{200}</td>
<td>136.8±0.4</td>
<td>0.327±0.002</td>
</tr>
<tr>
<td>{211}</td>
<td>188.6±0.6</td>
<td>0.261±0.002</td>
</tr>
<tr>
<td>{220}</td>
<td>195.1±0.6</td>
<td>0.313±0.004</td>
</tr>
<tr>
<td>500 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>180.8±0.7</td>
<td>0.300±0.003</td>
</tr>
<tr>
<td>{200}</td>
<td>115.6±0.8</td>
<td>0.322±0.003</td>
</tr>
<tr>
<td>{211}</td>
<td>179.6±0.4</td>
<td>0.274±0.001</td>
</tr>
<tr>
<td>{220}</td>
<td>185.2±0.5</td>
<td>0.292±0.002</td>
</tr>
<tr>
<td>600 °C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>{110}</td>
<td>171.8±0.8</td>
<td>0.335±0.004</td>
</tr>
<tr>
<td>{200}</td>
<td>89.7±0.6</td>
<td>0.340±0.004</td>
</tr>
<tr>
<td>{211}</td>
<td>167.3±0.7</td>
<td>0.290±0.004</td>
</tr>
<tr>
<td>{220}</td>
<td>176±1.2</td>
<td>0.372±0.010</td>
</tr>
</tbody>
</table>
5.3 Anisotropy in thermal expansion of bainitic ferrite

For a cubic single crystal, the orientation-dependent variation in elastic strain caused by a tensile stress is characterised by the cubic elastic anisotropy factor $A_{hkl}$ [113]:

$$A_{hkl} = \frac{h^2 k^2 + k^2 l^2 + l^2 h^2}{(h^2 + k^2 + l^2)^2}$$  \hspace{1cm} (5.5)

where $h$, $k$ and $l$ are Miller indices of the diffracting plane. The elastic strain for a family of grains with a plane normal $\langle hkl \rangle$ aligned parallel ($\parallel$) or perpendicular ($\perp$) to the loading direction is given by [112, 114, 115]:

$$\varepsilon_{hkl}^{\parallel} = \frac{S_{11} - 2 \left[ S_{11} - S_{12} - \frac{1}{2} S_{44} \right]}{\sigma} A_{hkl}$$ \hspace{1cm} (5.6)

$$\varepsilon_{hkl}^{\perp} = S_{12} + 2 \left[ S_{11} - S_{12} - \frac{1}{2} S_{44} \right] A_{hkl}$$ \hspace{1cm} (5.7)

where $S_{ij}$ are the elastic compliances. For the planes aligned with the plane-normal, parallel to the loading axis, a greater value of $A_{hkl}$ implies a greater stiffness $E_{hkl} = \sigma/\varepsilon_{hkl}^{\parallel}$. Hence for ferrite, the $\langle 200 \rangle$ direction is the most compliant crystallographic direction in the axial loading direction with $A_{200} < A_{110} = A_{211} = A_{220}$ and therefore $E_{200}^\alpha < E_{110}^\alpha = E_{211}^\alpha = E_{220}^\alpha$.

Figure 5.6 confirm these predictions. Along the loading direction, the $\{200\}$ ferrite plane is most strained by the applied stress at all temperatures. A comparable strain development for the $\{110\}$, $\{211\}$ and $\{220\}$ ferrite planes is observed up to the yield point. The qualitative elastic response observed experimentally is well predicted by the cubic elastic anisotropy factor.

5.3 Anisotropy in thermal expansion of bainitic ferrite

5.3.1 Introduction

Estimation and modelling of residual stresses in welded components from lattice strains associated with a specific reflection $\{hkl\}$ demands the use of the corresponding plane specific diffraction elastic constants (see section 5.2) and thermal expansion coefficients, which in general differ form the bulk properties. While the elastic and

---

thermal anisotropy of materials has long been known, experimental evidence of tetragonality of bainitic ferrite has only recently been verified [116]. In the present paper, the anisotropic thermal expansion behaviour of bainitic ferrite has been investigated. For this purpose, in-situ thermal tests have been carried out during continuous cooling in a high energy X-ray diffractometer. The data reported are of considerable significance, since they allow an accurate determination of the thermal stress states from the measured local $d$-spacings between lattice planes.

5.3.2 Thermal mechanical cycles

All the specimens were heated at a rate of $10 \, ^\circ \text{C s}^{-1}$, maintained at a temperature of $900 \, ^\circ \text{C}$ for 10 s, and then cooled to room temperature at a constant rate of $10 \, ^\circ \text{C s}^{-1}$. Including data recording time, a time resolution of $\sim 0.3$ s was achieved between successive images with data smearing over a temperature range of 0.8 $^\circ \text{C}$. The applied thermal cycle led to austenite formation during heating and the transformation of austenite into bainite (figure 5.7) during cooling. The sample was allowed to undergo free thermal expansion and contraction throughout the tests.

![Figure 5.7: The bainitic microstructure after thermal cycling.](image)

5.3.3 Data analysis

The raw diffraction images were processed using Fit2D image processing software, including corrections for the spatial distortion and detector efficiency along with subtraction of the background signal [110]. No obvious evidence of textural effects were observed from the 2-D diffraction patterns. The DeBye Scherrer rings up to the $\{220\}$ plane of ferrite were fully captured in the field of view of the detector. Integration over the azimuthal angles $\eta$ at a constant scattering angle was performed to obtain
5.3 Anisotropy in thermal expansion of bainitic ferrite

the corresponding one-dimensional (1-D) diffraction patterns [117, 118]. A fit of individual reflections to a pseudo-Voigt profile function over the integrated azimuthal angles was used to find the peak positions and intensities from the 1-D diffraction pattern [117]. The volume fractions of the bainitic ferrite (see figure 5.8) were calculated from the integrated intensities of three bainitic ferrite \( \alpha(200), \alpha(211) \) and \( \alpha(220) \) rings using the procedure described by van Dijk et al. [119].

![Figure 5.8: Evolution of bainitic ferrite fraction as a function of temperature.](image)

The planar thermal expansion coefficients as a function of the azimuthal angle were calculated from the change in the \( d \)-spacing of the constituent phase. This was performed by integrating over specific angular sections of 5° on the detector. A fit of individual reflections to a pseudo-Voigt profile function over the integrated azimuthal angular sections was used to find the peak positions and hence \( d \)-spacings. The intensity as a function of the scattering angle for the minimum and maximum temperature is presented in figure 5.9.
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

Figure 5.9: The intensity as a function of the scattering angle for the minimum and maximum temperature between which the thermal expansion coefficients were determined.
5.3 Anisotropy in thermal expansion of bainitic ferrite

5.3.4 Results

The temperature dependent interplanar spacings of the diffracting \{hkl\} planes of bainitic ferrite as a function of the azimuthal angle are presented in figure 5.10.

Figure 5.10: Variation of the interplanar spacing $d$ (Å) of the diffracting (a) \{200\}, (b) \{211\} and (c) \{220\} planes of $\alpha_b$ as a function of azimuthal angle $\eta$ and temperature.
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

In general, the temperature dependence of the interplanar spacings $d_{hkl}$ of a phase can be described by a polynomial of the form:

$$d_{hkl} = d_{hkl}^0 (1 + \alpha_{hkl} T + \beta_{hkl} T^2)$$

(5.8)

in which the coefficients $d_{hkl}^0$, $\alpha_{hkl}$ and $\beta_{hkl}$ can be determined by fitting the experimental data. The evolution of $\alpha_{hkl}$ as a function of azimuthal angle $\eta$ for the steel under investigation is presented in figure 5.11 and is valid for the temperature range 30 °C to 340 °C (when $f_{\alpha b} > 0.9$) during the cooling cycle.

![Figure 5.11](image)

**Figure 5.11:** Variation of the coefficient of thermal expansion of the diffracting (a) $\{200\}$, (b) $\{211\}$ and (c) $\{220\}$ planes of $\alpha^b$ as a function of azimuthal angle $\eta$. The results are shown for two different thermal cycles (see inset in figure 5.11a).
5.4 Discussion

The coefficient of thermal expansion (CTE) of bainitic ferrite showed anisotropy arising from the $d$-spacings (see $\alpha_{hkl}$ in figure 5.11). CTE is isotropic in cubic materials. However anisotropy in CTE was observed in the present case. This anisotropic CTE could arise due to the tetragonality in bainitic ferrite.

5.4 Discussion

5.4.1 Discussion: Diffraction elastic constant

The DECs for all the reflections measured at 100 °C (see figure 5.6) show a deviation from the trend. This discrepancy is reflected in the lattice strain perpendicular to the loading direction measured at 100 °C (figure 5.5b). The tensile test at 100 °C was carried out with a smaller beam size of 200 (vertical) × 200 (horizontal) $\mu$m$^2$ instead of 677 (vertical) × 761 (horizontal) $\mu$m$^2$. A smaller beam size resulted in a smaller gauge volume thereby capturing an insufficient number of grains to obtain a statistically relevant signal.

The errors in the calculation of the plane specific DECs could come from a number of sources. The instrument parameters (sample-detector distance, position of the straight-through beam and the tilt of the detector) during every measurement were determined using a CeO$_2$ calibrant placed on each sample prior to the tests. The errors arising from these global instrumental parameters were therefore eliminated. Errors could also be related to detector characteristics but they are systematic and are much smaller in magnitude than indicated by the error bars. Errors arising from the fitting of the individual reflections to a pseudo-Voigt profile functions and regression analyses are indicated in figure 5.6.

Smaller changes in the total strain are observed in the perpendicular direction than in the parallel direction (figure 5.5). This resulted in a proportionally larger scatter in the strains and hence in the slopes in the perpendicular direction. Nonetheless, the response of the strains from the individual peaks is approximately linear until the start of yielding. The larger scatter in the slopes for the perpendicular directions at elevated temperatures resulted in a drop in the Poisson’s ratio for the \{110\} and \{220\} peaks at around 500 °C (figure 5.6b).

5.4.2 Discussion: Anisotropy in thermal expansion of bainitic ferrite

Diffusionless transformation of austenite containing carbon necessarily leads to a body-centred tetragonal lattice [120]. The mechanism of the bainite reaction is diffusionless in the first instance, but carbon subsequently precipitates as cementite [121], or partitions into the residual austenite if the silicon or aluminium concentration
In-situ synchrotron diffraction studies on the strain development during thermal mechanical cycles in S690QL1

sufficiently high [122]. The maximum solubility of carbon in ferrite that is in equilibrium with austenite is \( \sim 0.02 \) wt. % at a temperature of about 600 °C [123, 124] based on the assumption of equilibrium between cubic ferrite and austenite; i.e. the equilibrium Fe-C phase diagram. The presence of excess carbon in bainitic ferrite has been shown from direct atom probe measurements [125, 126]. If the bainite inherits the composition of the austenite, then it will be body-centred tetragonal. Jang et al. [127] have shown using ab initio calculations that the maximum solubility of carbon in body-centred tetragonal ferrite that is in equilibrium with austenite is \( \sim 0.5 \) wt. % at a temperature of about 400 °C. This value is much higher than that for cubic ferrite and explains why excess carbon present in bainitic ferrite does not partition into the residual austenite despite prolonged heat treatment [125, 128–132]. Recently a tetragonal or slightly orthorhombic unit cell of bainitic ferrite was observed [116]. The tetragonality in bainitic ferrite could be the reason for the observed anisotropic thermal expansion behaviour. The latter should be taken into account when modelling thermal and thermal-mechanical strains and stresses in bainitic ferrite unit cells.

5.5 Conclusion

The temperature dependent plane specific diffraction elastic constants (DECs) were determined with a considerable degree of precision. For this purpose, in-situ tensile tests have been carried out at different temperatures in a high energy X-ray diffractometer. For the planes aligned with the plane-normal, parallel to the loading axis, a greater value of \( A_{hkl} \) implies a greater stiffness \( E_{hkl} = \sigma/\varepsilon^\parallel_{hkl} \). Hence for ferrite, the \( \langle 200 \rangle \) direction is the most compliant crystallographic direction in the axial loading direction with \( A_{200} < A_{110} = A_{211} = A_{220} \) and therefore \( E_{200}^\alpha < E_{110}^\alpha = E_{211}^\alpha = E_{220}^\alpha \). Figure 5.6 confirms these predictions. Along the loading direction, the \{200\} ferrite plane is most strained by the applied stress at all temperatures. A comparable strain development for the \{110\}, \{211\} and \{220\} ferrite planes is observed up to the yield point. The qualitative elastic response observed experimentally is well predicted by the cubic elastic anisotropy factor.

The evolution of local \( d \)-spacings between lattice planes during continuous cooling from austenite to bainitic ferrite in S690QL1 steel were determined to calculate the thermal expansion behaviour. For this purpose, in-situ continuous cooling tests carried out in a high energy synchrotron X-ray diffractometer. The results indicate anisotropic thermal expansion coefficients in the bainitic ferrite planes. The anisotropic CTE could arise due to the tetragonality in bainitic ferrite. Recently tetragonal or slightly orthorhombic unit cell of bainitic ferrite was observed [116]. The tetragonality in bainitic ferrite could arise from the excess carbon that persists in the ferrite, which is in contact with austenite. This is a consequence of an increased solubility due to the change in symmetry from the conventional cubic unit cell.
Chapter 6

The effect of tensile deformation with \textit{in-situ} ultrasonic treatment on the microstructure of low carbon steel*

6.1 Introduction

Steel industries develop steels with higher and higher tensile strength. It is commonly considered that the fatigue limit of the steel will increase as the ultimate tensile strength increases [133]. In the case of welded components of high strength steels with high stress concentration at the weld toe, the fatigue limit does not depend on the steel grade. In order to reduce the probability of weld cracking and to increase the fatigue resistance of welded components, it is possible to influence the following three parameters: the weld quality, the local geometry and the residual stresses. Post-weld treatments, such as heat treatments, thermo-mechanical treatments or volume and surface treatments, change one or more of these parameters. The traditional fatigue life enhancing techniques applied to welds are grinding, shot peening, hammer peening and tungsten inert gas (TIG) dressing. In the class of mechanical post weld treatments, most recent developments have occurred in the field of relatively novel high frequency peening, and in particular, ultrasonic methods, such as ultrasonic peening and ultrasonic impact treatment. Although reported results on fatigue life are very

promising [1–5, 7, 8], the detailed changes induced in the treated material and the mechanisms by which such changes occur are poorly understood. Ultrasonic impact treatment consists of an ultrasonic component and a mechanical impact component. Whether the ultrasonic component has a significant influence on material properties and microstructure in addition to the high frequency impacts during ultrasonic peening and ultrasonic impact treatments is not yet clear. However, it has been known for some time that ultrasound absorption can lead to movement and creation of dislocations.

The softening effect (reduction of the quasi-static stress) of superimposed ultrasonic vibrations on metals and alloys undergoing deformation is a well known effect [21–28, 134–136]. Many different machining, shaping and post weld treatment processes use ultrasonic vibrations; these include drilling [137], wire drawing [138, 139], lapping and turning of brittle materials [140], impact peening to modify surface properties [53], and welding of different metals [141] or metallic glasses [142]. Many of these applications are based on the reduction of quasi-static stress required for deformation when ultrasonic vibrations are applied; this phenomenon is called the Blaha [21] or acoustostatic effect.

Different types of deformation test have been carried out to show the effect of ultrasonic excitation. Nevill et al. [22] superimposed mechanical vibrations on low carbon steel wire during tensile deformation and found that the decrease in stress was proportional to the amplitude of the vibration. Izumi et al. [143, 144] reported similar results with superimposed ultrasonic vibration on compressive deformation of several metals and alloys. Similar tests have also been conducted [23–25, 27, 28, 134–136, 145–147] which show a reduction of the quasi-static applied stress during the superimposition of ultrasonic vibrations. More recent research by Huang et al. was conducted on plasticine with superimposed ultrasonic vibration during upsetting [148] and wedge indentation [149]. Using finite element simulation the authors initially concluded that the load reduction effect was due to stress superposition and reduction of friction. Further research by these authors [150, 151] and Daud et al. [152, 153] led to the conclusion that the effect of ultrasonic vibration cannot be entirely explained simply by these mechanisms. A change in material properties during ultrasonic excitation was suggested by Siu et al. [154]. They recently showed that the ultrasonic treatment enhances sub-grain formation underneath the hardness indents during deformation in polycrystalline aluminum.

There are different hypotheses for the mechanism behind the change in the material properties during ultrasonic excitation. Langenecker [28] proposed that under ultrasonic action, a change in the character of dislocation distribution and activation of new dislocation sources occurs as a result of preferential absorption of the energy of ultrasonic oscillations at defects of the crystalline lattice. An increase in dislocation mobility allows the metal to deform at a lower load. Other hypotheses, also touched upon above, include (i) the superposition of stresses [139, 155–158] (ii) thermal soften-
6.2 Experimental

In this chapter, a tensile deformation was applied using an electro-mechanical Instron tensile testing machine. DC04 steel specimens were examined with and without ultrasonic vibration applied in a direction normal to the tensile axis. Factors including the duration of the ultrasonic excitation and heating up of the sample during tensile deformation were investigated. The cross-sectional deformation microstructure was analysed using optical microscopy, scanning electron microscopy (SEM), electron backscatter diffraction (EBSD) and X-ray diffraction (XRD), in order to elucidate the effects of the ultrasound vibration.
The effect of tensile deformation with *in-situ* ultrasonic treatment on the microstructure of low carbon steel

### 6.2.1 Sample preparation

Commercial sheets of 2 mm thick low carbon DC04 steel with chemical composition presented in table 6.1, were cut into 4 tensile samples (see figure 6.1a) according to the ASTM-E8/E8M-09 [79] standard. They were cut in the rolling direction of the steel sheet. The tensile samples were then heat treated by austenitising at 890 °C for 1 hour in a salt bath furnace followed by air cooling to room temperature to minimise the stresses in the sample due to machining. The heat treated samples were electro-polished in a solution of 700 ml ethanol (absolute), 120 ml distilled water, 100 ml glycerol and 80 ml perchloric acid [159] to avoid the formation of dislocations at the surface due to grinding and polishing.

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Al</th>
<th>P</th>
<th>S</th>
<th>V</th>
<th>Ti</th>
<th>N</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.051</td>
<td>0.002</td>
<td>0.224</td>
<td>0.045</td>
<td>0.008</td>
<td>0.011</td>
<td>0.001</td>
<td>0.001</td>
<td>0.0037</td>
<td>bal.</td>
</tr>
</tbody>
</table>

### 6.2.2 Tensile deformation with *in-situ* ultrasonic treatment

The ultrasonic treatment of the heat treated and polished samples was carried out by excitation *via* a waveguide. The oscillation amplitude and frequency were 26 µm and 27 kHz, respectively. The contact force between the waveguide and tensile sample was maintained at approximately 50 N *via* a spring loaded cell to ensure good contact but at the same time to avoid sample bending. The ultrasonic treatment was applied *in-situ* during tensile deformation between strain levels of 8 and 20 %. The cross-head displacement velocity was 2.7 mm min\(^{-1}\). Using an extensometer with a gauge length of 20 mm, elongation was recorded for the applied load. Four cases are considered: 1.) 0 % strain, 2.) 8 % strain, 3.) 20 % strain and 4.) 20 % strain with *in-situ* ultrasonic treatment during tensile deformation between strain levels of 8 and 20 % (samples 1, 2, 3 and 4 respectively, see figure 6.1b).
6.2 Experimental

Figure 6.1: Schematic view of a.) sample orientation. RD, TD and ND correspond to the rolling, transverse and normal direction of the rolled sheet and b.) test conditions. The red curve shows the tensile curve superimposed with ultrasonic vibrations.
The effect of tensile deformation with \textit{in-situ} ultrasonic treatment on the microstructure of low carbon steel

6.2.3 Microstructure characterisation

To prepare samples for optical microscopy, EBSD and X-ray diffraction, the tensile specimens were sectioned along the normal direction (ND) either transverse (TD) or parallel to the rolling direction (RD) for all the cases (see figure 6.1a). The sectioned specimens were prepared for optical microscopy and etched with 5 % nital solution to reveal the microstructure. The sectioned specimens for EBSD and X-ray diffraction were first plane ground with SiC paper to a 2400 grit finish, followed by fine polishing with diamond paste to a 1 micron finish. Finally the surface was electro-polished using a solution of 700 ml ethanol, 120 ml distilled water, 100 ml butoxyethanol and 80 ml perchloric acid [159].

Optical microscopy was performed using an Olympus BX60M$^{TM}$ optical microscope. Vickers micro hardness measurements were performed with a load of 300 g in a Buehler$^{\textregistered}$ microhardness testing (MHT) machine. The hardness measurements were performed 10 times and the results were averaged to improve the measurement statistics.

The EBSD scans were carried out on a NOVA 600 focused ion beam (FIB) scanning electron microscope (SEM) equipped with a field emission gun (FEG) operating at an accelerating voltage of 15 kV, a beam current of 0.59 nA and a working distance of 7 mm. The sample was tilted at 70 $^\circ$ to the horizontal axis for the EBSD scans and a step size of 0.25 $\mu$m in a square scan grid measuring 175 $\times$ 175 $\mu$m$^2$ was used for all the scans. By choosing an optimum image resolution for pattern processing and by optimising the Hough transform parameters, an angular resolution of better than 0.5 $^\circ$ was obtained. The lateral resolution of the system is around 30 nm parallel to the tilt axis and around 90 nm perpendicular to the tilt axis, determined on iron at 15 kV [160]. The scans were made in the centre of the sample at a depth of around 1 mm. The EBSD data were post-processed by means of TSL$^{\textregistered}$ - Orientation Imaging Microscopy (OIM$^{TM}$) data analysis software. Post-processing was done in order to eliminate the points with confidence index (CI) lower than 0.12 from the EBSD maps.

Quantitative X-ray diffraction (XRD) measurements were performed with a Bruker D8 Advance diffractometer equipped with a Vantec position sensitive detector and a graphite monochromator in the diffracted beam [161]. X-ray diffraction (XRD) patterns were recorded in a Bragg-Brentano geometry at room temperature using monochromatic Co-K$\alpha_1$ radiation ($\lambda = 0.179026$ nm). Scans from $2\theta = 35^\circ$ to $135^\circ$ were performed with a step size of 0.041 $^\circ$ and a time step of 2 s. The sample was placed on a Si $\langle 510 \rangle$ substrate and was not rotated during measurement. The irradiated volume was of the order of $12 \times 2 \times 0.01$ mm$^3$. Data evaluation was performed with the Bruker program EVA to obtain the peak positions and the full-width at half maximum (FWHM). In the present work (110), (200), (211) and (220) reflections of ferrite were measured.
6.3 Methodology of calculation

6.3.1 Calculating geometrically necessary dislocation (GND) densities from EBSD data

By means of high-resolution electron backscatter diffraction (HR-EBSD) it is possible to obtain information about the grain orientation, dislocation density and sub-grain structure in a representative area [162–164]. Individual crystallographic orientations as well as polarised arrays of dislocations with the same sign can be studied. Using automated orientation imaging microscopy (OIM), the electron beam scans the inspected area. The crystallographic orientation and a value for the quality of the Kikuchi pattern, viz. the image quality (IQ) for each point is determined. The latter can be linked qualitatively to lattice imperfections. Local changes in the lattice orientation reflect lattice curvature and can be used to calculate GND densities. One of the methods to retrieve the GND densities from the HR-EBSD data follows Takayama and Szpunar [165, 166]. As a first order approach, the kernel average misorientation (KAM), which is calculated from EBSD data, was chosen as a measure for the local misorientations. The KAM is defined for a given point as the average misorientation of that point with all of its neighbours [163], which is calculated with the proviso that misorientations exceeding a tolerance value are excluded from averaging calculations. The tolerance value for the judgement of the grain interiors used in this work is 5° and the first neighbour is used in the calculation. From the KAM value, the dislocation density can be derived [165]. However, it is likely to be underestimated because of the existence of dislocations that do not contribute to build up of the misorientation. According to Humphreys et al. [167], a low angle tilt boundary consists of edge dislocations, while a low angle twist boundary consists of screw dislocations [168]. The GND density $\rho_{gnd}$ was calculated from the measured average misorientation angle and the spacing between dislocation boundaries using the following equation [169]:

$$\rho_{gnd} = \frac{\gamma \vartheta}{ub}$$  \hspace{1cm} (6.1)

where $\vartheta$ is the average misorientation angle across dislocation boundaries, $u$ is the distance between the misoriented points (namely the ‘step size’ of the EBSD map) and $b$ is the magnitude of the Burgers vector. The constant $\gamma$ is dependent on the geometry of the boundaries having values of 2 and 4 for pure tilt and pure twist boundaries, respectively [35]).

The rank of the neighbour or the distance between two measurement points considered for the misorientation calculation is critical for a physically meaningful GND density determination. The following points have to be considered when choosing the distance between two measurement points: (1) the distance has to be small enough to allow detailed information to be obtained; (2) the distance has to be large enough to average out the scatter due to EBSD spatial resolution limits; and (3) the calculations
must be performed with misorientations above the angular EBSD resolution limits.

As the simple KAM values are not normalised by spacing, they increase with increasing neighbour rank. This is not the case for the calculated GND densities, as these values are distance normalised. Comparing the respective GND densities obtained for the 1st and 2nd neighbour sets revealed that the contrast between high and low GND density areas increases with increasing neighbour rank. Since the EBSD scan step used in the present research was 0.25 µm, the first nearest neighbours were selected for defining the kernel.

In the present chapter, differences of grain orientation greater than 15° were defined as high-angle grain boundaries, while those lower than 15° were defined as low-angle grain boundaries. The low-angle grain boundaries were again discretised between 2-5° and 5-15°.

6.3.2 Evaluation of dislocation density from X-ray diffraction profiles

The density of dislocations can be evaluated from the Williamson-Hall procedure [170] using the full width at half maximum, FWHM (Δ2θ) values for the undeformed, deformed and reference specimens as:

\[
(D2\theta)^2 = (D2\theta_{(un)deformed})^2 - (D2\theta_r)^2
\]

where Δ2θ_{(un)deformed} and Δ2θ_r are the measured values of FWHM for the (un)deformed and reference specimens (NIST SMR66a LaB₆), respectively. This is carried out to eliminate the instrumental broadening.

In the classical Williamson-Hall procedure [170], the full width at half maximum, FWHM (Δ2θ) of profiles are plotted as a function of \(K = 2\sin\theta/\lambda\), where \(\theta\) is the diffraction angle and \(\lambda\) is the wavelength of X-rays. The intercepts and the slopes of the regressions through the measured data provide the crystallite size parameters and the values of the mean square strain, respectively [170]. Due to strain anisotropy however, the data points do not follow smooth curves, making reliable regression impossible. It can be shown that the anisotropic contrast of the dislocations enables the rationalisation of strain anisotropy in terms of the modified Williamson-Hall plot [171, 172] for a texture free specimen that contains one type of dislocation with a random orientation distribution:

\[
\Delta K \cong \frac{\alpha}{D} + \left(\frac{\pi \kappa b^2}{2}\right) D^{\frac{1}{2}} K^2 C + O(K^4 C^2)
\]
where $\Delta K$ is $(2\Delta 2\cos \theta / \lambda)$, $D$ characterises the size of the coherently diffracting domains, $b$ is the Burgers vector, equal to $\frac{a\sqrt{3}}{2}$ for a bcc Fe alloy [173], $\alpha$ is 0.9 for the FWHM, $\kappa$ is a constant depending on the effective outer cut-off radius of the dislocations, $a$ is the lattice parameter and $\overline{C}$, given by equation 6.4, is the average contrast factor of dislocations depending on the relative positions of the diffraction vector, the line vectors of the dislocations and the elastic constants of the crystal [171, 172, 174–176]. $O$ stands for higher order terms in $K^2\overline{C}$. In the present work, the modified Williamson-Hall plot (equation 6.3) was used to evaluate the density of dislocation from the X-ray diffraction profiles. The fourth order term $O(K^4\overline{C}^2)$ is considered negligible.

In a texture free cubic polycrystal or if the Burgers vector population on the different slip systems is random, the contrast factors of dislocations can be averaged over the permutations of the $hkl$ indices [174, 177]:

$$\overline{C} = \overline{C}_{h00}(1 - qH^2)$$  \hspace{1cm} (6.4)

where $\overline{C}_{h00}$ is the average dislocation contrast factor for the $h00$ reflections, $H^2 = \frac{h^2k^2+k^2l^2+l^2h^2}{(h^2+k^2+l^2)^2}$ and $q$ is a parameter depending on the elastic constants of the crystal and on the edge or screw character of the dislocations [174]. $\overline{C}_{h00}$ and $q$ for pure edge and pure screw dislocations for Fe were evaluated from the elastic anisotropy factor, $A\left(\frac{2C_{14}}{C_{11}-C_{12}}\right)$ and $\frac{C_{12}}{C_{44}}$ [174], where $C_{11}$, $C_{12}$ and $C_{44}$ are the second order elastic constants, which for Fe are 230.1 GPa, 134.6 GPa and 116.6 GPa respectively [178]. For pure edge dislocations in Fe, $\overline{C}_{h00}$ and $q$ were calculated to be 0.2648 and 1.31, respectively and for pure screw dislocations, $\overline{C}_{h00}$ and $q$ were calculated to be 0.3055 and 2.64, respectively.

### 6.4 Results

#### 6.4.1 Microstructure and mechanical properties

The microstructure of the undeformed DC04 steel after the initial heat treatment (sample 1) consists of ferritic grains (see figure 6.2a) with small colonies of pearlite (see figure 6.2b) at the grain corners. The average grain diameter calculated using the TSL®-OIM data analysis software for sample 1 was $(22 \pm 0.9) \mu m$.

The average microhardness of the undeformed steel after the initial heat treatment (sample 1) was $94.6 \pm 1.4 \text{ H}_{V_{0.005}}$. The tensile behaviour of DC04 steel samples at room temperature showed a typical stress-strain curve with continuous yielding and a completely ductile failure (figure 6.3). The results show that the average yield stress at 0.2% offset was $240 \pm 13$ MPa, with an average ultimate tensile stress of
The effect of tensile deformation with *in-situ* ultrasonic treatment on the microstructure of low carbon steel

Figure 6.2: Microstructure of DC04 steel after initial heat treatment and before deformation (sample 1) etched with 5 % nital. a) Optical micrograph showing recrystallised and equiaxed grains. b.) Scanning electron micrograph showing small pearlite colonies at grain corners.

314 ± 10 MPa and a tensile strain at fracture of 43.8 ± 0.3 %. The Young’s modulus was 210 ± 6 GPa.

Surface analysis of the 20 % strained DC04 steel with superimposed ultrasonic vibrations (sample 4) revealed heavy damage at the contact surface of the tensile sample as shown in figure 6.4a. This damage was caused by the contact between the waveguide and the tensile sample resulting in an increased temperature. The depth of the damaged layer was around 60 µm (see figure 6.4b). Transient temperature measurements were made on the tensile sample, at a depth of 1 mm from the contact surface between the tensile sample and the waveguide, with a 0.25 mm diameter K-type thermocouple and the transient distribution is shown in figure 6.5. During the initial stages of *in-situ* ultrasonic treatment, the temperature rose rapidly and then stabilised. The maximum temperature reached at the given depth in the tensile sample during the entire treatment was around 140 °C.
6.4 Results

Figure 6.3: The tensile stress-strain curves for DC04 steel samples at room temperature.

Figure 6.4: Scanning electron micrograph of sample 4 showing a) heavy surface damage due to friction between the waveguide and the sample at the contact surface b) depth of damage.
The effect of tensile deformation with in-situ ultrasonic treatment on the microstructure of low carbon steel

6.4.2 Orientation gradients and GNDs

The effect of ultrasound on the distribution of the dislocation density within the grain and along the grain boundary can be visualised by using a HR-EBSD analysis placing emphasis on local orientation gradients at interfaces. Figure 6.6 shows the EBSD maps of undeformed DC04 steel (sample 1, figures 6.6a and 6.6d), 20 % strained DC04 steel (sample 3, figures 6.6b and 6.6e) and 20 % strained DC04 steel with superimposed ultrasonic vibrations (sample 4, figures 6.6c and 6.6f). In these maps, figures 6.6a, 6.6b and 6.6c correspond to the image quality (IQ), the darker the colour, the lower the IQ and the higher the lattice distortion. The IQ value allows one to distinguish the distributions of dislocation density within the grains and along the grain boundaries as it exhibits higher lattice distortion. It follows that the specimen without deformation (sample 1, figure 6.6a) shows a small number of sub-grains (see the red and blue lines indicating 2-5° and 5-15° angle of rotation, respectively), whereas the 20 % strained DC04 steel specimen (sample 3, figure 6.6b) shows a significantly larger number of sub-grains. The occurrence of a large number of sub-grains upon plastic deformation in sample 3 can be understood as follows: at 20 % total strain, the specimen experiences local, inhomogeneous plastic deformation because the sum of the internal (misfit) stress generated by the dislocations and the externally applied tensile stress locally exceeds the yield stress of ferrite. The stored strain energy due to plastic
6.4 Results

deformation can be (partly) released by regrouping dislocations into dislocation walls; *i.e.* low-angle boundaries; resulting in the formation of sub-grains. The formation of the sub-grains is not observed in the specimen subjected to ultrasound (sample 4, figure 6.6c) even though both the specimens (samples 3 and 4) have the same amount of plastic deformation. The fraction of the high and low angle grain boundaries for the different samples are presented in table 6.2. With increment in strain, the fraction of low-angle grain boundaries increases remarkably in sample 3 compared to 1. This phenomenon is attributed to the occurrence of a dislocation sub-structure induced by deformation [179]. The fraction of low-angle grain boundaries decreases significantly with *in-situ* ultrasonic treatment (sample 4 compared to sample 3).
The effect of tensile deformation with \textit{in-situ} ultrasonic treatment on the microstructure of low carbon steel

![Image](image.png)

**Figure 6.6:** Image quality (IQ) maps with high and low angle grain boundaries, kernel average misorientation (KAM) map (in $^\circ$) and geometrically necessary dislocation density ($\rho_{gnd}$) maps (in m$^{-2}$) calculated from kernel data of sample 1, sample 3 and sample 4.
6.4 Results

Table 6.2: Fraction of high and low angle grain boundaries for the different samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Rotation angle 2-5°</th>
<th>Rotation angle 5-15°</th>
<th>Rotation angle 15-180°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>0.07</td>
<td>0.07</td>
<td>0.860</td>
</tr>
<tr>
<td>Sample 3</td>
<td>0.56</td>
<td>0.32</td>
<td>0.12</td>
</tr>
<tr>
<td>Sample 4</td>
<td>0.36</td>
<td>0.06</td>
<td>0.57</td>
</tr>
</tbody>
</table>

Figures 6.6d, 6.6e and 6.6f show the kernel average misorientation (KAM) for samples 1, 3 and 4, respectively. Here, the average misorientation of an EBSD point is calculated with respect to all the first nearest neighbours at 0.25 µm distance (values above 5° are excluded). As expected, the undeformed DC04 steel (sample 1) shows almost no gradient in misorientation, while the 20% strained DC04 steel (sample 3) shows the largest orientation gradients. More importantly, the KAM maps reveal considerable orientation gradients spreading along the grain boundaries in the 20% strained DC04 steel with superimposed ultrasonic vibrations (sample 4, figure 6.6f). More detailed analysis reveal that these are oriented along the direction of propagation of ultrasound (figure 6.6f). On the basis of the HR-EBSD scans sized 175 µm × 175 µm the overall geometrically necessary dislocation density in the ferrite was calculated to be $3.03 \times 10^{14}$ m$^{-2}$, $7.31 \times 10^{14}$ m$^{-2}$ and $3.87 \times 10^{14}$ m$^{-2}$ for samples 1, 3 and 4, respectively. As expected, the overall dislocation density increases with increasing deformation (samples 1 and 3). However, the dislocation density decreases with in-situ ultrasonic treatment (samples 3 and 4) for the same amount of deformation.

From the KAM values, the GND density was calculated using equation 6.1 with $\gamma = 3$ for boundaries of mixed character (see figures 6.6g, 6.6h and 6.6i). The dislocation density maps show that while the dislocations are scattered throughout the matrix in the 20% strained DC04 steel (sample 3), they are concentrated primarily along the grain boundaries in the case of the 20% strained DC04 steel with superimposed ultrasonic vibrations (sample 4). The GND density values vary from about $5.2 \times 10^{14}$ m$^{-2}$ to $4.2 \times 10^{15}$ m$^{-2}$ in both samples 3 and 4.

The image quality map of the undeformed sample (figure 6.6a) shows a recrystallised structure with a large variation in grain size, most likely as a result of the processing of the sample. No lattice curvature was apparent in the undeformed material. The image quality map of the 20% strained DC04 steel (sample 3, figure 6.6b) shows heavily deformed grains, as expected. Strikingly, undeformed grains were found in the 20% strained DC04 steel with superimposed ultrasonic vibrations (sample 4, figure 6.6c) with the same amount of tensile deformation. With in-situ ultrasonic treatment, the extent of grain deformation and in-grain orientation gradient is reduced significantly.
The effect of tensile deformation with *in-situ* ultrasonic treatment on the microstructure of low carbon steel

The distributions of kernel average misorientation of the samples are plotted in figure 6.7. For the undeformed DC04 steel (sample 1), a lognormal KAM distribution and a small average value of KAM indicate a small and randomly distributed local strain. The distribution of KAM shifts to higher values and the mean KAM increases with increasing deformation as seen in the 20 % strained DC04 steel (sample 3). However, in sample 4, with *in-situ* ultrasonic treatment, the peak fractions of KAM shifts to lower values and the mean KAM decreases, indicating a lower local strain, and hence lower GND density.

![Figure 6.7: Experimentally determined KAM distributions of samples 1, 3 and 4.](image)

### 6.4.3 Determination of dislocation contrast factors and density of dislocation by modified Williamson-Hall plot

A strong strain anisotropy caused by dislocations was observed in the classical Williamson Hall plot of the FWHM which justified the use of the modified Williamson-Hall plot. Substituting \( C \) from equation 6.4 into equation 6.3, the latter was solved for \( D, \frac{(\pi \kappa b^2)}{2} \rho \bar{\rho}^2 \) and \( q \) by the method of least squares. For a wide range of dislocation characteristics, the value of \( \kappa \) can be taken as 0.1 [180, 181]. The dislocation density was then calculated from \( \frac{(\pi \kappa b^2)}{2} \rho \bar{\rho}^2 \). Table 6.3 shows the density of dislocations \( \rho \) and the screw-edge character of dislocations \( q \) under different conditions. The FWHM for DC04 steel samples under different conditions are shown in a modified Williamson-Hall plot in figure 6.8.
6.5 Discussion

The perpendicular vibration of the sample with respect to the tensile axis caused significant surface damage on the contact surface of the tensile specimen. The damage was caused by the contact between the waveguide and the tensile sample resulting in increased temperature. However, the increased temperature is far below the temperature at which self diffusion of iron occurs.

The perpendicular vibration of the sample with respect to the tensile axis is likely to cause an oscillatory shear stress wave in the contact region due to the axial tension [182]. This shear stress wave will reinforce the stress field due to the axially loaded

---

Table 6.3: Microstructural parameters calculated from the X-ray diffraction profiles.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\rho_{m^{-2} \times 10^{-15}}$</th>
<th>$q$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.7±0.25</td>
<td>2.64</td>
</tr>
<tr>
<td>2</td>
<td>11.3±1.2</td>
<td>1.68</td>
</tr>
<tr>
<td>3</td>
<td>24.5±4.3</td>
<td>1.61</td>
</tr>
<tr>
<td>4</td>
<td>20.6±1.8</td>
<td>1.32</td>
</tr>
</tbody>
</table>

Figure 6.8: The modified Williamson-Hall plot of the FWHM for DC04 steel samples under different conditions.

---

6.5 Discussion

The perpendicular vibration of the sample with respect to the tensile axis caused significant surface damage on the contact surface of the tensile specimen. The damage was caused by the contact between the waveguide and the tensile sample resulting in increased temperature. However, the increased temperature is far below the temperature at which self diffusion of iron occurs.

The perpendicular vibration of the sample with respect to the tensile axis is likely to cause an oscillatory shear stress wave in the contact region due to the axial tension [182]. This shear stress wave will reinforce the stress field due to the axially loaded
The effect of tensile deformation with \textit{in-situ} ultrasonic treatment on the microstructure of low carbon steel

sample, to produce a more intensive total stress field. If the intrinsic resistance to deformation is the same, a smaller axial force is required to deform the metal with the ultrasound vibration and this is manifested by the decrease in the tensile stress with superimposed ultrasonic vibrations.

Both the X-ray diffraction (XRD) and the EBSD measurements show that the dislocation density decreases with \textit{in-situ} ultrasonic treatment during tensile testing. The XRD profile analysis yields dislocation densities which are higher than those calculated from the KAM. The reason is that statistically stored dislocations are additionally counted when evaluating the XRD profile. The dislocations accumulated in the material can be divided into statistically stored dislocations and geometrically necessary dislocations [183]. The former accumulate under homogeneous deformation conditions and evolve from random trapping processes [183], whilst the latter are the result of heterogeneous deformation and are required to maintain lattice continuity [183–185]. The geometrically necessary dislocations involve an excess of dislocations which have the same Burgers vector and lead to lattice rotations responsible for the lattice curvature [186, 187]. The geometrically necessary dislocations can be stored uniformly or lead to the formation of dislocation walls. In the latter case the lattice rotation concentrates on the formed boundary and the misorientation angle across it increases with strain [187, 188]. Another reason for the difference in the dislocation densities is probably due to the difference in volume of material investigated and different methods of measurements. The decrease in the dislocation density due to ultrasonic treatment could be due to the increased mobility of the screw dislocations. Atomistic simulations have found that a screw dislocation in BCC metals lies along an energetically favorable direction and experiences very high lattice resistance [189–193]. The motion of a screw dislocation is typically carried out by thermally-assisted kink-pair nucleation. Once the kinks are nucleated, they quickly move away over long distances before annihilating with other kinks. As a result, the screw dislocation moves forward by one lattice distance. The ultrasonic energy could be preferentially absorbed by dislocations [28], thereby helping them to overcome the lattice resistance.

It is seen from the image quality maps (figures 6.6b and 6.6c) and table 6.2 that with increment in the strain level, the fraction of small-angle grain boundary increases remarkably. This phenomenon is attributed to the occurrence of dislocations and sub-structures induced by deformation [179]. The fraction of low-angle grain boundaries decreases significantly with \textit{in-situ} ultrasonic treatment (sample 4 compared to sample 3). This may be attributed to enhanced dipole annihilation caused by an oscillatory stress wave that is superimposed on a uni-directional tensile stress [154]. Another reason for the decrease in the low-angle grain boundaries could be an increased dislocation mobility due to the oscillatory stress wave.

Figures 6.6a, 6.6b and 6.6c show that ultrasonic vibrations reduce the formation of sub-grains. Both, samples 3 and 4 have the same amount of tensile deformation, but the sub-grain formation is much more pronounced in sample 3 (see figure 6.6b)
6.6 Summary

compared to sample 4 (see figure 6.6c). However, more detailed analysis of figure 6.6f for the 20 % strained DC04 steel with superimposed ultrasonic vibrations (sample 4) reveals the formation of shear bands within the grains. The shear bands are oriented at an angle of approximately 45° to the tensile direction; i.e., the direction of propagation of ultrasound. The significant reduction in sub-grain formation with the application of ultrasound to the DC04 steel is contrary to previous observations made for aluminum [154, 194]. This could be due to the fundamental differences that exist in the deformation mechanisms between fcc and bcc metals. In fcc metals, glide occurs on the \{111\} \langle 110 \rangle slip systems whereas in bcc systems glide may occur on \{110\} \langle 111 \rangle or on \{211\} \langle 111 \rangle systems. In a few cases glide on \{321\} \langle 111 \rangle systems is also observed in bcc materials. Another important difference is that in bcc metals screw dislocations have a complex atomic structure with the core spreading onto different lattice planes [195]. As a result, the mobility of screw dislocations is lower than that of edge dislocations at temperatures below the so called athermal temperature $T_a$ ($T_a \sim 0.15T_m$ [196–198] where $T_m$ is the melting point). Below $T_a$, screw dislocations move by thermally activated processes which leads to a strong dependence of the yield strength on the strain rate [199]. This can also be seen with the parameter $q$ from table 6.3, which varies between 2.64 and 1.32 for samples 1–4. The higher value of $q$ for the undeformed DC04 steel (2.64) indicates that screw dislocations are dominant in the annealed state, and with further deformation, there is a rapid increase in the fraction of edge type dislocations. It is observed that at higher strain, the dislocation structure becomes edge-dominated.

6.6 Summary

Tensile deformation with in-situ application of ultrasonic vibration was performed on low carbon DC04 steel samples. The ultrasonic vibration was found to produce a significant and permanent change in the microstructure which was analysed using optical microscopy, scanning electron microscopy (SEM), electron backscatter diffraction (EBSD) and X-ray diffraction (XRD). The observations can be summarised as follows:

- Significant surface damage was found on the contact surface of the tensile specimen caused by the contact between the waveguide and the tensile sample resulting in increased temperature. However, the increased temperature is far below the temperature at which self diffusion of iron occurs.
- The dislocation density decreases by in-situ ultrasonic treatment.
- Sub-grain formation is substantially reduced by in-situ ultrasonic treatment.
- The fraction of low-angle grain boundaries decreases significantly by in-situ ultrasonic treatment.
The effect of tensile deformation with *in-situ* ultrasonic treatment on the microstructure of low carbon steel

- Dislocations are concentrated in the vicinity of the grain boundaries after *in-situ* ultrasonic treatment.
- The extent of grain deformation and in-grain orientation gradient is reduced significantly by *in-situ* ultrasonic treatment.
- The orientation gradients spread along the grain boundaries in the direction of propagation of ultrasound.

These observations are interpreted *via* enhanced dipole annihilation and/or increased dislocation mobility caused when an oscillatory stress wave is superimposed on a uni-directional tensile stress. The role played by ultrasound in modifying the microstructure will help to understand how an ultrasonic component may influence material properties and microstructures when ultrasonic peening and ultrasonic impact treatments are employed to mitigate welding induced residual stresses.
Chapter 7

Accommodation of plastic deformation by ultrasound induced grain rotation*

7.1 Introduction

A substantial reduction of sub-grains and low-angle grain boundaries with *in-situ* ultrasonic treatment during tensile deformation of low carbon steel has been shown in chapter 6 and in [200], indicating that plastic deformation is accommodated by ultrasound induced grain boundary rotation. In this chapter the possibility of the reduction in low angle grain boundaries by grain rotation is further explored. To this end, investigations on the role of *in-situ* ultrasonic treatment on the microstructure and sub-grain structure of low carbon steel during tensile deformation have been performed using optical microscopy and electron backscatter diffraction (EBSD).

7.2 Experimental

The details of material used, sample preparation and methodology of microstructural characterisation has been discussed in section 6.2 of chapter 6.

---

7.3 Results and discussion

7.3.1 Microstructure evolution

Cross-sections of the low carbon steel without and with in-situ ultrasonic treatment during tensile deformation are shown in figure 7.1. It is evident that the in-situ ultrasonic treatment significantly reduces the number of sub-grains. The histogram of grain size distribution of the low carbon steel without and with in-situ ultrasonic treatment is shown in figure 7.2. The deformation is more homogeneous without in-situ ultrasonic treatment as is evident from the grain size distributions (figures 7.2a and 7.2b). With in-situ ultrasonic treatment, the deformation is clearly inhomogeneous, as shown by the mixture of equiaxed grains (shown by black arrows in figure 7.1b) and very elongated grains (bimodal grain size distributions in figures 7.2c and 7.2d). These relatively small equiaxed grains measuring between 5 and 15 µm in diameter are observed at the grain boundaries of large elongated grains. The EBSD observations imply that the formation of relatively small equiaxed grains is attributed to dynamic recrystallisation during the process of deformation, rather than static recrystallisation during a heating and cooling process, because a typical feature of grains formed by static recrystallisation is that they are nearly strain/defect free [201]. The occurrence of dynamic recrystallisation is supported in a more quantitative manner by the distributions of misorientations between neighbouring crystallites (figure 7.1c). Without in-situ ultrasonic treatment, a large fraction of misorientations is below 15°, which is due to the continuous generation of sub-grain boundaries. The fraction of misorientation between 15° and 63°, corresponding to high angle boundaries, increases significantly in the in-situ ultrasonic treated sample, which may be attributed to the progressive transformation of sub-grain boundaries into grain boundaries (shown by red arrows in figure 7.1b).

The grain orientation spread (GOS) parameter corresponds, for one grain, to the average deviation between the orientation of each point in the grain and the average orientation of the grain [202]. Figure 7.3a shows the grain orientation spread map of the ultrasonically treated sample. Figure 7.3b shows the image quality map of the low carbon steel with in-situ ultrasonic treatment where the grains with average grain diameter <15 µm are highlighted in blue. The grain orientation spread (GOS) distributions were plotted for the grains with average grain diameter <15 µm and >15 µm. The grains with average grain diameter <15 µm show a lower average spread compared to the grains with average grain diameter >15 µm indicating that the larger grains have undergone greater plastic deformation. Local lattice rotations can be quantitatively analysed using the Grain Reference Orientation Deviation (GROD) function [202]. This tool reveals the angular deviation of each point relative to a given reference orientation, e.g. a point representing the stable grain interior far away from the grain boundary interface. In the present case, the grain average orientation is used as the reference. Figure 7.4a shows such GROD maps in which the colour code given in the index reveals those areas that are highly misoriented
7.3 Results and discussion

Figure 7.1: EBSD characterisation of the cross-section of the low carbon steel. Image quality map with low angle boundaries and high angle boundaries for (a) 20% deformation without in-situ ultrasonic treatment and (b) in-situ ultrasonic treatment during tensile deformation between strain levels of 8 and 20%. $\omega_1 = 16.6^\circ$, $\omega_2 = 13.1^\circ$ and $\omega_3 = 13.1^\circ$ represent the boundary misorientation between the adjacent grains. (c) Grain boundary misorientation angle distribution for the deformed steel with and without in-situ ultrasonic treatment.
Accommodation of plastic deformation by ultrasound induced grain rotation

Figure 7.2: Grain size distribution of the steel without ultrasonic treatment: (a) diameter as a function of number fraction and (b) diameter as a function of area fraction. Grain size distribution of the steel with in-situ ultrasonic treatment: (c) diameter as a function of number fraction and (d) diameter as a function of area fraction.

with respect to the grain average orientation. Figure 7.4b shows the kernel average misorientation (KAM) map of the same area. KAM in combination with GOS can be used to estimate plastic deformation [202]. In fact, Kamaya [203] has shown that the GOS is an even better tool than KAM, as it is less affected by the measurement conditions. The KAM criterion can also be used to determine geometrically necessary dislocation concentrations in grains of different orientations. Kamaya [204] has shown that comparing KAM values for near grain boundary regions with the average KAM value of the neighbouring grains may give information about concentration or localisation of geometrically necessary dislocations. If the two values are similar, this
7.3 Results and discussion

means that dislocations are uniformly distributed in the grain, whereas a KAM value for near grain boundary regions higher than the average KAM of the grain means that dislocations are mainly located in grain boundary regions. This effect can be easily visualised with the help of the GROD maps (figure 7.4a) as it is normalised to the grain average orientation. It is seen that in the smaller grains (with grain diameter less than 15 µm), the grain reference orientation deviation is higher closer to the grain boundaries than in the grain interior (see for example figure 7.5 for the case of grain 2 from figure 7.1b).

Figure 7.3: (a) Grain orientation spread map of the ultrasonically treated sample. (b) Image quality map of the low carbon steel with in-situ ultrasonic treatment during tensile deformation between strain levels of 8 and 20 %. The grains with average grain diameter <15 µm are highlighted in blue. (c) Experimentally determined grain orientation spread distribution for the grains with average grain diameter <15 µm and >15 µm.
Accommodation of plastic deformation by ultrasound induced grain rotation

Figure 7.4: Low carbon steel with in-situ ultrasonic treatment during tensile deformation between strain levels of 8 and 20 %. (a) Grain reference orientation deviation map with grain average orientation as the reference. (b) Kernel average misorientation map.

There are two possible mechanisms for dynamic recrystallisation: rotational and migrational types [205]. One way to determine which of these is operating is to consider the change in misorientation moving from the original grains to the recrystallised region [206, 207]. Figure 7.6a is the combined image quality (IQ) and colour coded inverse pole figure (IPF) map of grains 1 and 2 from figure 7.1b. The point-to-point and the point-to-origin misorientations are plotted as a function of distance along path ABCD from the elongated grain (ABC) to the centre (D) of the equiaxed grain, as shown in figure 7.6b. It is seen that there are three apparently different regions divided by points A, B, C and D in figures 7.6a–c, showing the distinct microstructures and the corresponding misorientations. Moving from A to B, the image quality decreases and there is a gradual accumulation of \(\sim 6.5^\circ\) of misorientation over \(\sim 12.5\, \mu\text{m}\).
7.3 Results and discussion

Figure 7.5: *GROD map of grain 2 from figure 7.1b.*

indicating long range lattice rotations within this grain (figure 7.6b). The misorientation gradient in this region is relatively low indicating low lattice strain and defect density. Moving from B to C, there is a sharp peak in the point-to-point misorientation at \( \sim 19 \, \mu \text{m} \) indicating the presence of a low angle or sub-grain boundary. The misorientation gradient across the sub-grain boundary reaches an average value of \( 30^\circ \, \mu \text{m}^{-1} \). Crossing into the equiaxed grain, (moving from C to D), the misorientation reaches the highest value (\( \sim 58^\circ \) at \( \sim 30 \, \mu \text{m} \)) indicating the presence of a high angle grain boundary.

To further reveal the deformation induced lattice rotation in one grain (grain 1 from figure 7.1b), the tolerance angle map (figure 7.6c) is illustrated using the colour gradient from blue to red. Moving from point A to D, it can be seen that the lattice is gradually rotated. The tolerance angle gradually increases across the sub-grain boundary (between point B and C) reaching maximum close to the high angle boundary (between point C and D). The lattice rotations along the line ABCD are also illustrated using the inverse pole figures (figure 7.6d and 7.6e) and table 7.1. These results show that the lattice in the elongated grain is progressively rotated until high angle boundaries appear at the equiaxed grain. Similar observations were made in other pairs of elongated and equiaxed grains (see for example figure 7.3b and 7.3c).

Dynamic recrystallisation can be classified into either continuous or discontinuous recrystallisation [208]. In general, during continuous recrystallisation, dislocations will remain in the recrystallised grains whereas discontinuous recrystallisation removes dislocations through the sweeping action of high angle boundaries. Continuous recrystallisation is also considered as a recovery dominated process where there will be a progressive increase in boundary misorientation and conversion of low angle boundaries into high angle boundaries.
In the present work, dislocations are observed in the smaller equiaxed recrystallised grains, primarily along the grain boundaries. Well defined sub-grain boundaries were rarely observed in the recrystallised grains interior, inferring that sub-grain boundary misorientation increased during deformation with *in-situ* ultrasonic treatment and
low angle grain boundaries transformed into high angle boundaries. Evidence of this transformation can be found in figure 7.1b (shown by red arrows). The boundary misorientation $\omega$ between the adjacent grains increases from $\omega_3 = 13.1^\circ$ to $\omega_1 = 16.6^\circ$ indicating progressive transformation of sub-grain boundaries into grain boundaries.

### 7.3.2 Conceptual modelling of microstructure evolution

Based on the present observations, it is reasonable to propose a model describing a mechanism for dynamic recrystallisation occurring with *in-situ* ultrasonic treatment. The dislocation distribution is homogeneous without ultrasonic treatment (figure 7.1a). As a consequence of *in-situ* ultrasonic treatment, the original grains are elongated into substructures. Dislocations accumulate at substructure boundaries leading to the break up of the elongated substructures. As the sub-boundaries reorient, there is an increase in orientation difference at the boundaries followed by rotation of sub-boundaries and the formation of recrystallised grains with high angle boundaries. This conceptual model is, in fact, in good agreement with those proposed by Xu *et al.* [209] and Li *et al.* [210]. This sequence of events, which is well known for severe plastic deformation, has been given different names in the literature: (1) Rotational dynamic recrystallisation (*e.g.* Derby [205]), which needs concurrent plastic deformation, is well documented for geological materials. This was the interpretation given in adiabatic shear bands by Meyers *et al.* [211, 212] for titanium, Andrade *et al.* [213] for copper and Nesterenko *et al.* [214] for tantalum. (2) Formation of geometrically necessary boundaries [215–221]. (3) Continuous recrystallisation [222, 223]. Once this equiaxed fine grain structure is achieved, it can undergo additional plastic deformation under the imposed conditions.

### 7.3.3 Recrystallised grain diameter

Experiments on dynamic recrystallisation [222, 224] suggest that the recrystallised grain size ($D_R$) may be estimated using a simple relationship: 
\[
\sigma \left( \frac{b}{G} \right)^n = K,
\]
where $\sigma$ is applied stress, $n$ and $K$ are constants (being 0.8 and 15, respectively), $b$ is the Burgers vector (0.248 nm for iron) and $G$ is the shear modulus (80 GPa for steel). The applied stress $\sigma$ during *in-situ* ultrasonic treatment at 20 % strain was about 255 MPa and the calculated dynamic recrystallised grain size ($D_R$) is about 10 $\mu$m, which is in good agreement with the observed grain size by EBSD. Empirical equations (equation 7.1) to estimate the average steady state grain sizes during recrystallisation [225–227] yielded $\overline{D} = 7$ $\mu$m.

\[
\overline{D} = 38.26 \left( \frac{Z}{A} \right)^{-0.08} \mu m
\]

where $Z = \dot{\varepsilon} \exp \frac{Q_{\text{deformation}}}{RT}$ is the Zener–Hollomon parameter and $Q_{\text{deformation}}$ is the
Accommodation of plastic deformation by ultrasound induced grain rotation

activation energy of deformation. $Q_{\text{deformation}}$ and $A$ for the present steel were calculated, based on the chemical composition [225], to be 269 kJ mol$^{-1}$ and $2.8 \times 10^9$ s$^{-1}$. These results further confirm that dynamic recrystallisation is a possible mechanism for the formation of the small equiaxed grains (average grain diameter of the grains with size $<15 \, \mu$m is 6.5 $\mu$m) at the grain boundaries of elongated grains during deformation with in-situ ultrasonic treatment.

7.4 Summary

Tensile deformation with in-situ application of ultrasonic vibration was performed on low carbon DC04 steel samples. The ultrasonic vibration was found to produce a significant and permanent change in the microstructure which was analysed using optical microscopy, scanning electron microscopy (SEM) and electron backscatter diffraction (EBSD). Sub-grain formation was substantially reduced and the fraction of low angle grain boundaries decreased by in-situ ultrasonic treatment.

The deformation is clearly inhomogeneous, as shown by the mixture of equiaxed grains and very elongated grains giving rise to a bimodal grain size distribution. It was shown through EBSD that significant grain rotation can take place during the process of deformation with in-situ ultrasonic treatment. The average grain boundary misorientations increase with the in-situ treatment. The formation of the small equiaxed grains at the grain boundaries of elongated grains is attributed to rotational dynamic recrystallisation. The lattice in the elongated grain is progressively rotated until high angle boundaries appear at the equiaxed grain. GROD maps show that the dislocations are primarily concentrated along the grain boundaries for the smaller grains (with average grain diameter $<15 \, \mu$m), whereas for larger grains (with average grain diameter $>15 \, \mu$m) they are more homogeneously distributed.

In conclusion, although the exact mechanisms of the development of the bimodal grain size distribution during deformation with in-situ ultrasonic treatment is not completely understood, it is becoming obvious that grain boundary rotation plays an important role.
Chapter 8

Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

8.1 Introduction

Surface severe plastic deformation (S\textsuperscript{2}PD) processes have been developed over the past decade to introduce nano grains and grain size gradients into the surface region of bulk materials [228]. The formation of a nanocrystalline microstructure can result in a considerable improvement in mechanical properties, such as the low and high temperature strength, fatigue resistance and tribological properties [54, 229–235]. The key feature of these processes, such as hammer peening [236], surface rolling [237], machining [228] and surface mechanical attrition treatment (SMAT) [55], is severe plastic deformation at the surface region to induce nanocrystallisation at the surface while leaving the interior untouched and creating a gradual transition both in properties and microstructure between the two extremes. This kind of surface modification, referred as surface nanocrystallisation (SNC), will greatly enhance the surface properties without changing the chemical composition [55, 228]. Optimisation of the surface structure of materials is of great importance since most defects initiate from the sur-
Formation of nanostructures in severely deformed high strength steel
induced by high frequency ultrasonic impact treatment

face. Another advantage of surface nanocrystallisation is that it greatly enhances the diffusion kinetics of atoms. It has been found that the nitriding temperature of iron produced using SMAT can be reduced to 300 °C, which is at least 200 °C below the conventional nitriding temperature [238].

The evolution of severely strained microstructures has been the subject of a number of studies focused on cubic and hexagonal materials [55, 239–248]. These studies reveal that surface nanocrystallisation mechanisms depend strongly on the crystal structure and stacking fault energy (SFE) of the material. For materials with high stacking fault energies, grain refinement is dominated by dislocation activities, entailing generation of high dislocation densities, the formation of subgrains and the evolution of subgrain boundaries to highly misoriented grain boundaries [245]. Also the microstructure refinement during large deformation is associated with the creation of deformation-induced boundaries [241, 244], including (i) geometrically necessary boundaries (GNBs), which separate regions that deform by different combinations of slip systems, and (ii) incidental dislocation boundaries, consisting of ordinary cell boundaries that form by the trapping of glide dislocations. The formation of geometrically necessary boundaries with moderate-to-high misorientation requires a relatively high level of strain [244].

Recently, such surface nanocrystallisation techniques have been used to improve the fatigue limit in welded components of high strength steels. The effective life of welded components with high stress concentration at the weld toe, is determined by its fatigue limit. In order to reduce the probability of weld cracking and to increase the fatigue resistance of welded components, it is possible to influence the weld quality, the local geometry and the residual stresses. Post-weld treatments, such as heat treatments, thermo-mechanical treatments or volume and surface treatments, change one or more of these parameters. The traditional fatigue life enhancing techniques applied to welds are grinding, shot peening, hammer peening and tungsten inert gas (TIG) dressing. In the class of mechanical post weld treatments, most recent developments have occurred in the field of relatively novel high frequency peening, and in particular, ultrasonic methods, such as ultrasonic peening (UP) and ultrasonic impact treatment (UIT). Such ultrasonic peening techniques are known to greatly enhance the fatigue life of welded components [1, 3, 4] via surface modification by the generation of a nanostructured surface layer. Ultrasonic impact treatment consists of an ultrasonic and a mechanical impact component. The effect of tensile deformation with in-situ ultrasonic treatment on the microstructure of low carbon steel was studied previously by Dutta et al. [200]. Characterising surface nanocrystallisation caused by the combined effect of the ultrasonic component in addition to the high frequency impacts during ultrasonic peening and ultrasonic impact treatment is thus a necessary step in understanding the reason behind the improvement in fatigue life of welded components.
In order to characterise this heavily deformed surface layer, a combination of crystalllographic data and imaging capabilities is needed. However, it is difficult to index microstructures that are at most a few tens of nanometres thick using the conventional electron backscattered diffraction (EBSD) technique based on scanning electron microscopy (SEM) \[249-251\]. Conventional transmission electron microscopy (TEM) is not convenient for the analyses of orientation relationships. Automated crystal orientation mapping in TEM (ASTAR) couples the advantages of the TEM and EBSD techniques to investigate the orientation mapping with the high spatial resolution of TEM \[252-256\]. This technique presents many advantages, in a complementary way to the classical EBSD technique based on SEM. First of all, it presents a high spatial resolution of $\sim$2 nm. Since information of every pixel can be retrieved based on the corresponding diffraction pattern, it is thus possible to recreate virtual bright/dark field images by selecting specific reflections from the diffraction pattern. However, the area/volume that can be considered is much smaller than with classical EBSD. The present work aims at quantitative nanoscale characterisation of the severely deformed top layers induced by post weld high frequency ultrasonic impact treatment.

### 8.2 Experimental

The investigated steel has the general composition Fe-0.16C-0.2Si-0.87Mn-0.11Ni-0.33Cr-0.21Mo-0.005Ti-0.02Cu (wt. %) \[257\]. Details of the welding procedure can be found in chapter 4 and in \[258\]. The microstructure of the welded zone prior to the ultrasonic impact treatment is shown in figure 8.1. A martensitic microstructure with an average grain diameter of 1.84 $\mu$m was present in the heat-affected zone prior to the ultrasonic impact treatment. The UIT tool was positioned at the weld toe of the sixth pass at an angle of 30\(^\circ\) to the normal direction (ND) (see figure 8.2a). The treatment parameters are listed in table 8.1.
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

Figure 8.1: Micrograph of the welded zone prior to the ultrasonic impact treatment. Combined image quality (IQ) and colour coded inverse pole figure (IPF) map of the heat affected zone from where the TEM foil were made after nano-crystallisation via the ultrasonic impact treatment.

Table 8.1: UIT process parameters.

<table>
<thead>
<tr>
<th>Process parameters</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Waveguide frequency, kHz</td>
<td>27</td>
</tr>
<tr>
<td>Waveguide Amplitude, µm</td>
<td>28</td>
</tr>
<tr>
<td>Pin dimensions, mm</td>
<td>Ø3 × 26, R3</td>
</tr>
<tr>
<td>Tool weight, kg</td>
<td>3</td>
</tr>
<tr>
<td>Impact frequency, Hz</td>
<td>120</td>
</tr>
<tr>
<td>Feed rate, mm s⁻¹</td>
<td>5</td>
</tr>
<tr>
<td>Number of treatment passes</td>
<td>1</td>
</tr>
</tbody>
</table>
8.2 Experimental

Figure 8.2: (a) Scheme of the UIT tool orientation with respect to the welded sample; (b) Optical micrograph of the region from where the FIB foil was extracted. The white dotted line shows the boundary between the surface nano-crystallised zone and the weld metal. The black dotted line shows the fusion line; (c) SEM micrograph of the region from where the FIB foil was extracted; and (d) SEM micrograph of a cross-sectional TEM foil prepared by FIB.

8.2.1 Microstructure characterisation

To prepare samples for optical microscopy, the welded and ultrasonically impact treated plates were sectioned along the normal direction either transverse or parallel to the rolling direction for all the cases. The sectioned specimens were prepared for optical microscopy and etched with 5% nital solution to reveal the microstructure.

TEM foils were extracted from different depths under the ultrasonic impact treated surface as indicated in figure 8.2b, 8.2c and 8.2d using a focused ion beam (FIB). The black and the white dotted lines in figure 8.2b show the fusion line and the boundary between the surface nano-crystallised zone and the weld metal respectively. For the
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

preparation of the TEM sample, a FEI Quanta 3D was employed. Firstly, a protective platinum layer was deposited prior to FIB milling. This prevents the surface of the steel being damaged by the incident Ga$^+$ ions. An ion beam of 30 kV/5 nA and 30 kV/0.3 nA was used for sample cutting and early stage milling. For the final step, an ion beam of 30 kV/0.1 nA was employed to achieve the final milling and to minimise any amorphous layers generated during high voltage FIB milling on both sides of the samples. A Philips CM20 transmission electron microscope equipped with a LaB$_6$ gun operating at 200 kV was used for the observations. Diffraction patterns were recorded with a precession angle of 1° using an external CCD camera. Off-line, every diffraction pattern was compared to the pre-calculated templates of selected phases and the best match selected [252, 253]. Orientation mapping characteristics for the TEM foils at different depths are listed in table 8.2. The automated crystal orientation mapping in TEM (ASTAR) data was post-processed by means of TSL® - Orientation Imaging Microscopy (OIM$^\text{TM}$) data analysis software. Post-processing was done in order to eliminate the points with reliability lower than 0.1 from the TEM maps. For any given TEM foil, at least two field scans were made and analysed individually for statistical representation; however, for brevity, data from only one of the scans is presented here. In the present study, different maps were reconstructed using inverse pole figure colour coding combined with reliability contrast or virtual dark field contrast.

<table>
<thead>
<tr>
<th>Table 8.2: Orientation mapping characteristics for the TEM foils.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Depth from the surface</td>
</tr>
<tr>
<td>Step size (nm)</td>
</tr>
<tr>
<td>electron beam size (nm)</td>
</tr>
</tbody>
</table>

8.2.2 Criterion for grain boundary characterisation

Special boundary relationships were identified using the coincidence-site-lattice (CSL) theory via the Brandon (or proximity) criterion [259]:

$$ \Delta \theta_m \leq 15^\circ \Sigma^{-1/2} $$

(8.1)

where $\Delta \theta_m$ is the maximum permissible deviation from the exact coincidence. In the present work, the boundaries were classified as following:

- Low angle boundaries (LABs), characterised by $\Sigma 1$ if the misorientation $\theta < 15^\circ$;
- High angle grain boundaries (HAGBs) if the misorientation $\theta \geq 15^\circ$. The HAGBs are classified as:
8.2 Experimental

- Special grain boundaries (SGBs) or low-\(\Sigma\) CSL boundaries, with a \(\theta\) deviation within \(\Delta\theta_m\) characterised by \(1 < \Sigma \leq 29\);
- Random grain boundaries (RGBs) when:
  * \(\theta\) deviation within \(\Delta\theta_m\) characterised by \(\Sigma > 29\);
  * \(\Delta\theta_m > 15^\circ \Sigma^{-1/2}\).

8.2.3 Post processing methodology by TSL\textsuperscript{®} - OIM\textsuperscript{TM}

Previous investigations on IF-steel [260] have shown anomalies arising due to the unusual incidence of \(\Sigma_{13b}\) and \(\Sigma_3\) CSL boundaries due to misindexing by the TSL\textsuperscript{®} - OIM\textsuperscript{TM} software. In the case of severely deformed bcc iron this phenomenon arises because 27.8\(^\circ\) \(\langle 111\rangle\) and 60\(^\circ\) \(\langle 111\rangle\) orientations are more prone to pseudo-symmetry than others [261, 262]. In order to minimise such errors and false interpretations associated with such errors, all maps were cleaned by disregarding the effect of the \(\Sigma_{13b}\)-boundaries. In order to avoid ambiguities for the measurement of the grain size distribution, \(\Sigma_3 \langle 111\rangle\) were also excluded from the measurements.
8.3 Results

8.3.1 TEM investigations of post weld ultrasonic impact treatment

To investigate the grain refinement mechanism and the evolution of grain boundary character distribution, several TEM foils were extracted from different depths under the ultrasonic impact treated surface. The gradient structure resulting from a gradual decrease in the applied strain and strain rate as the depth from the treated surface layer increases, from very high (top surface) to zero (substrate), represents the complete range of structural changes which occur during treatment. A colour coded inverse pole figure (IPF) map is shown to represent the microstructure as a function of the depth from the treated surface. The grain boundary statistics are presented as misorientation angle and axis distributions and also in terms of the grain boundary character distribution (GBCD) represented by the reciprocal density of coincidence sites in table 8.3. The relatively small number of low angle boundaries in the misorientation angle distribution is due to the 5° tolerance, i.e. boundaries with misorientation angles less than 5° are not displayed.

Table 8.3: Grain boundary character distribution as a function of depth from the surface as per Brandons criterion.

<table>
<thead>
<tr>
<th>Depth from the surface</th>
<th>0 µm (%)</th>
<th>~27 µm (%)</th>
<th>~52 µm (%)</th>
<th>~98 µm (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Σ1</td>
<td>51.3</td>
<td>42.6</td>
<td>42.4</td>
<td>58.0</td>
</tr>
<tr>
<td>Σ3 - 29b</td>
<td>23.5</td>
<td>32.2</td>
<td>25.6</td>
<td>13.5</td>
</tr>
<tr>
<td>Σ &gt; 29</td>
<td>2.2</td>
<td>1.7</td>
<td>1.5</td>
<td>1.8</td>
</tr>
<tr>
<td>Δθ_m &gt; 15°Σ^{-1/2}</td>
<td>23.0</td>
<td>23.5</td>
<td>30.5</td>
<td>26.7</td>
</tr>
</tbody>
</table>

98 µm below the treated surface

Figure 8.3a shows the microstructure at a depth of ~98 µm below the treated surface. Very fine parallel lamellae or microbands (200 - 1000 nm in thickness) are observed, some of which are divided into a series of blocks. In some places, the fine lamellar structure has been converted into submicronic polygonal grains (indicated by grey arrows in figure 8.3a). The near-uniform colours of the lamellae indicate that these lamellae correspond to an individual single zone axis although gradations in shading signify the presence of a substructure with boundaries of misorientation below 15°. It is reasonable to believe that the increase in strain is the cause of the subdivision of lamellae into blocks and the formation of polygonal grains.
8.3 Results

Figures 8.3b and 8.3c show the misorientation angle and axis distributions respectively and the grain boundary character distribution (GBCD) is presented in table 8.3. The misorientation angles show a high fraction of low misorientation angles (5° – 10°). The grains at a depth of ∼98 µm below the treated surface exhibit the lowest fraction of low-Σ CSL boundaries (∼14%).

![Figure 8.3](image1)

(a)

![Figure 8.3](image2)

(b)

![Figure 8.3](image3)

(c)

**Figure 8.3:** Micrograph of the welded zone after the ultrasonic impact treatment at a depth of 98 µm from the surface. (a) Combined image quality (IQ) and colour coded inverse pole figure (IPF) map; (b) Grain boundary misorientation by misorientation angles; (c) Misorientation axis distributions in misorientation angle sections of 5° (except for the first two sections, which are 0°-10°).

52 µm below the treated surface

Figure 8.4a shows the microstructure at a depth of ∼52 µm below the treated surface. The near-uniform colours of the grains in the microstructure indicate that these grains corresponds to an individual single zone axis although gradations in shading signify the presence of a substructure with boundaries of misorientation below 15°. Observation of the TEM foil shows scattered areas of residual lamellae or polygonal grains and subdivision of microbands into low angle misoriented blocks.

Figures 8.4b and 8.4c show the misorientation angle and axis distributions respect-
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

ively and the grain boundary character distribution (GBCD) is presented in table 8.3. The misorientation angles show a high fraction of misorientation angles around 35°. This is also evident from the misorientation axis distributions in figure 8.4c showing high intensity of grain boundaries oriented along ⟨001⟩ directions. This arises due to the Σ5 boundaries (36.9° ⟨100⟩). The fraction of Σ5 boundaries is ~16%. The grains at a depth of ~52 µm below the treated surface exhibit the highest fraction of high angle grain boundaries (~31%).

Figure 8.4: Micrograph of the welded zone after the ultrasonic impact treatment at a depth of 52 µm from the surface. (a) Combined image quality (IQ) and colour coded inverse pole figure (IPF) map; (b) Grain boundary misorientation by misorientation angles; (c) Misorientation axis distributions in misorientation angle sections of 5° (except for the first two sections, which are 0°-10°).

27 µm below the treated surface

Figure 8.5a shows the microstructure at a depth of ~27 µm below the treated surface. An increasing fraction of polygonal grains, traces of heavily deformed residual lamellae and the presence of substructures is observed as a result of increasing applied strain and strain rate. It is suggested that the microbands divide into highly misoriented polygonal submicronic grains.

Figures 8.5b and 8.5c show the misorientation angle and axis distributions respect-
8.3 Results

ively and the grain boundary character distribution (GBCD) is presented in table 8.3. The misorientation angles show a high fraction of misorientation angles around 35°. This is also evident from the misorientation axis distributions in figure 8.5c showing a high intensity of grain boundaries oriented along (001) directions. This arises due to the Σ5 boundaries. The fraction of Σ5 boundaries is ~22%. The grains at a depth of ~27 µm below the treated surface exhibit the highest fraction of low-Σ CSL boundaries (~32%).

Figure 8.5: Micrograph of the welded zone after the ultrasonic impact treatment at a depth of 27 µm from the surface. (a) Combined image quality (IQ) and colour coded inverse pole figure (IPF) map; (b) Grain boundary misorientation by misorientation angles; (c) Misorientation axis distributions in misorientation angle sections of 5° (except for the first two sections, which are 0°-10°).

Top treated surface layer

The morphology of the microstructure of the nanocrystalline grains present in the surface layer (figure 8.2d) after surface nanocrystallisation by ultrasonic impact treatment was studied by ASTAR-TEM (as described earlier). Figure 8.6a shows a colour coded phase map in which blue corresponds to a bcc lattice and red corresponds to a face centred cubic (fcc) lattice (austenite). The pixels with reliability lower than 0.1 were removed from analysis and are marked in black in the same map. Phase analysis showed 96.3% bcc and the remaining 3.7% retained austenite. The retained austenite is dispersed in the microstructure. The average grain size of the retained
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

austenite is 16.7 nm. In the present chapter only the bcc phase is discussed in detail.

Figure 8.6b shows an inverse pole figure (IPF) map of the bcc phase (ferrite/martensite). The histogram of grain size distribution of the ferrite/martensite phase is shown in figures 8.6c and 8.6d. The average grain size of the nanocrystalline bcc phase is 34.5 nm. It should also be noted that, in order to avoid ambiguities for the measurement of the grain size distribution, $\Sigma 3 \{111\}$ twin boundaries were excluded in the measurements shown in figures 8.6c and 8.6d.

![Image](a)

![Image](b)

![Image](c)

![Image](d)

**Figure 8.6:** (a) Colour coded phase map in which the blue corresponds to bcc lattice (ferrite/martensite) and red corresponds to fcc lattice (austenite). The black corresponds to the pixels with confidence index < 0.1; (b) IPF map of the bcc lattice; (c) Grain size distribution of the bcc lattice, diameter vs. area fraction; and (d) Grain size distribution of the bcc lattice, diameter vs. number fraction.

As shown in figures 8.6c and 8.6d, although the average grain size of ferrite/martensite is 34.5 nm, there is a large size distribution of grains in the matrix. For this purpose, three separate grains of different sizes were selected for misorientation calculation.
8.3 Results

Figure 8.7a shows two adjacent bcc grains each with a diameter measuring \(\sim 490 \text{ nm}\) and figure 8.7b shows another bcc grain with a diameter measuring \(\sim 185 \text{ nm}\). The grains are superimposed with linear traverses for calculation of the point-to-point and point-to-origin lattice misorientations. For the adjacent martensite grains in figure 8.7a, four linear traverses numbered 1 through 4 were drawn and for the individual bcc grain in figure 8.7b, one linear traverse numbered 1 was drawn. A and B in the traverse numbers represent the start and end positions. The near-uniform colours of each of the three grains indicate that each of these grains corresponds to an individual single zone axis although gradations in shading signify the presence of a substructure with boundaries of misorientation below 15°. Comparing traverse 1 in figure 8.7a with traverse 1 in figure 8.7b, it is evident the larger grains are more prone to long range lattice rotations involving a gradual accumulation of 12.5° misorientation (black dotted line in figure 8.7c) over \(\sim 250 \text{ nm}\) compared to less than 5° misorientation (black dotted line in figure 8.7d) over \(\sim 300 \text{ nm}\). Traverse 2 in figure 8.7a also shows a sharp peak in the point-to-point misorientation profile (red solid line in figure 8.7c) at \(\sim 48 \text{ nm}\) from the start position of the traverse, indicating the presence of sub-boundaries. The gradual accumulation of 8° of misorientation (red dotted line in figure 8.7c) over \(\sim 118 \text{ nm}\) indicated long range lattice rotations within this grain. The same traverse shows another sharp peak of 37.8° misorientation in the point-to-point misorientation profile (red solid line in figure 8.7c) at \(\sim 122 \text{ nm}\) from the start position of the traverse, as it crosses into the neighbouring grain. This peak indicates the presence of \(\Sigma 9\) (38.9° (110)) CSL boundaries. Traverse 3 in figure 8.7a also shows the presence of long range lattice rotations within this grain (blue dotted line in figure 8.7c). Traverse 4 in figure 8.7a shows a sharp peak of 59.8° misorientation in the point-to-point misorientation profile (green solid line in figure 8.7c) at \(\sim 39 \text{ nm}\) from the start position of the traverse as it crosses into the neighbouring grain. This peak indicates the presence of a \(\Sigma 3\) (twin) CSL boundary between the two neighbouring grains. Since there is a wide distribution of grain sizes and the deformation process is grain size dependent, the deformation process, the grain boundary distribution of misorientation angles and the grain boundary character distribution is classified for (1) overall distribution; (2) grains with average diameter less than 50 nm (D<50 nm); (3) grains with average diameter between 50 nm and 100 nm (50 nm\(\leq D<100 \text{ nm}\)) and (4) grains with average diameter greater than 100 nm (D\(\geq 100 \text{ nm}\)).
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

Figure 8.7: (a) Large single grain showing long range lattice rotation; (b) Point-to-point and point-to-origin misorientation for the lines in figure 8.7a. A and B represents the start and end positions of the lines; (c) Smaller grain showing lack of long range lattice rotation; and (d) Point-to-point and point-to-origin misorientation for the line in figure 8.7b.
8.3 Results

Figures 8.8 shows the kernel average misorientation (KAM) map, the kernel average misorientation distribution and the image quality distribution. Here, the average misorientation of a pixel point is calculated with respect to all the second nearest neighbours (values above 5° are excluded). The KAM map (figure 8.8a) reveals considerable orientation gradients spreading within as well as along the grain boundaries. In the absence of local strain, the KAM shows a lognormal distribution with a small average value (typically less than 0.4° [200]). In the present case, with severe surface deformation, the KAM distribution shifts to higher values. The average KAM for the overall distribution in the present case is 1.0012°. On the basis of the ASTAR-TEM scans of 1.836 μm × 1.808 μm, the overall average geometrically necessary dislocation (GND) density was calculated from the mean KAM [200]. The average GND was calculated to be 2.7×10^{16} m^{-2}. It is observed from figure 8.8b that the distribution of KAM shifts to lower values and the mean KAM decreases with increasing grain size, indicating a lower local strain, and hence lower GND density. High KAM and low IQ (figure 8.8c) qualitatively indicates that the disordered regions are primarily seen in the smaller grains.

The grain boundary statistics were measured from 1722 grains and presented as misorientation angle distributions in figure 8.9, as misorientation axis distributions in figure 8.10 and also in terms of the grain boundary character distribution (GBCD) represented by the reciprocal density of coincidence sites in table 8.4. The relatively small number of low angle boundaries (figure 8.9) is due to the 5° tolerance, i.e. boundaries with misorientation angles less than 5° are not displayed. In all cases the correlated (i.e. grain boundaries in the microstructure) distribution is included alongside the theoretical curve for a random misorientation distribution (indicated by the solid lines in figure 8.9). The overall GB distributions of misorientation angles (figure 8.9a) show a bimodal character with peaks at low and high angles. The fraction of low angle boundaries (Σ1) is ~51%. The distribution of misorientation angles show a peak at ~60°. The overall misorientation axis distribution is presented in figure 8.10a. Misorientations are plotted in standard unit triangles of the stereogram at misorientation angle intervals of 5°. There are several very distinct maxima, most of which correspond to one of the three lowest-index axes, ⟨001⟩, ⟨101⟩ or ⟨111⟩. Misorientation peaks occur in the overall distribution for axes near ⟨111⟩ at 55° and 60° and near ⟨101⟩ and ⟨111⟩ at 65°. The distribution of misorientation angles showing a peak at ~60° corresponds to twin (Σ3) misorientation. The fraction of Σ3 boundaries is ~11%.

A similar trend is observed for the grains with average diameter less than 50 nm (D<50 nm). The fraction of Σ3 boundaries is ~8%. This class of grain sizes also exhibit the highest fraction of low-Σ CSL boundaries (19%).

For the grains with an average diameter between 50 nm and 100 nm (50 nm≤D<100 nm), the misorientation angles show peaks at low and high angles. However, there is a high fraction of low misorientation angles (5° – 10°) and a low fraction of high
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

Figure 8.8: (a) Kernel average misorientation (KAM) map (in $^\circ$); (b) Experimentally determined KAM distributions for the different grain size distributions and (c) Experimentally determined IQ distributions for the different grain size distributions.

Table 8.4: Grain boundary character distribution of the surface layer as per Brandons criterion.

<table>
<thead>
<tr>
<th></th>
<th>Overall (%)</th>
<th>D&lt;50 nm (%)</th>
<th>50 nm≤D&lt;100 nm (%)</th>
<th>D≥100 nm (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Sigma 1$</td>
<td>51.3</td>
<td>64.9</td>
<td>85.2</td>
<td>75.3</td>
</tr>
<tr>
<td>$\Sigma 3$ - 29b</td>
<td>23.5</td>
<td>19.0</td>
<td>5.9</td>
<td>9.1</td>
</tr>
<tr>
<td>$\Sigma &gt;29$</td>
<td>2.2</td>
<td>2.0</td>
<td>0.6</td>
<td>1.4</td>
</tr>
<tr>
<td>$\Delta \theta_m &gt; 15^\circ \Sigma^{-1/2}$</td>
<td>23.0</td>
<td>14.1</td>
<td>8.3</td>
<td>14.2</td>
</tr>
</tbody>
</table>

misorientation angles (55$^\circ$ – 65$^\circ$) for this group of grains. This class of grains exhibit the lowest fraction of low-$\Sigma$ CSL boundaries (~6%) and also the lowest fraction of high angle grain boundaries (~8%). However, the fraction of $\Sigma 1$ boundaries increases to ~85%. For these grains, there is also a peak arising in misorientation angles close
8.3 Results

to 30° (figure 8.9c). This is also evident from the misorientation axis distributions (figure 8.10c). This arises due to the Σ17a boundaries (28.1° ⟨100⟩). The fraction of Σ3 and Σ17a boundaries is ∼1.5% and ∼1.1% respectively.

For the grains with an average diameter greater than 100 nm (D ≥ 100 nm), the misorientation angles show peaks at low and high angles. However, there is a higher fraction of low misorientation angles (5° – 10°) compared to the overall distribution and the grains with average diameter less than 50 nm. The fraction of Σ1 and Σ3-29b boundaries is ∼75% and ∼9% respectively. The fraction of high misorientation angles (55° – 65°) is however comparable to the overall distribution and for grains with average diameter less than 50 nm. This class of grains exhibit ∼9% low-Σ CSL boundaries. The fraction of Σ3 boundaries is ∼5%. For these grains, there is also a peak arising in misorientation angles close to ∼18° (figure 8.9d). This arises due to the Σ31a boundaries (17.9° ⟨111⟩). The fraction of Σ31a boundaries is ∼0.15%.

Figure 8.9: Grain boundary misorientation by misorientation angles for (a) overall distribution; (b) grains with average diameter less than 50 nm (D < 50 nm); (c) grains with average diameter between 50 nm and 100 nm (50 nm ≤ D < 100 nm) and (d) grains with average diameter greater than 100 nm (D ≥ 100 nm).
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment.

Figure 8.10: Misorientation axis distributions in misorientation angle sections of $\phi$ (except for the first two sections, which is $\phi=10^\circ$) for (a) the overall distribution; (b) grains with average diameter greater than 100 nm; (c) grains with average diameter between 50 nm and 100 nm ($50 \text{ nm} < D < 100 \text{ nm}$); (d) grains with average diameter less than 50 nm ($D < 50 \text{ nm}$).
Table 8.5 shows the average grain size as a function of the grain rotation angle. It is observed that the average grain size decreases with increasing grain rotation angle.

Table 8.5: *Average grain size of the surface layer as a function of grain rotation angle.*

<table>
<thead>
<tr>
<th>Grain rotation angle (GRA)</th>
<th>$D_{avg}$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{GRA}&lt;5^\circ$</td>
<td>145.7</td>
</tr>
<tr>
<td>$5^\circ \leq \text{GRA}&lt;15^\circ$</td>
<td>41.2</td>
</tr>
<tr>
<td>$15^\circ \leq \text{GRA}&lt;30^\circ$</td>
<td>30.6</td>
</tr>
<tr>
<td>$30^\circ \leq \text{GRA}&lt;45^\circ$</td>
<td>27.9</td>
</tr>
<tr>
<td>$45^\circ \leq \text{GRA}&lt;70^\circ$</td>
<td>20.5</td>
</tr>
</tbody>
</table>

8.4 Discussion

The ultrasonic impact treatment cause surface nanocrystallisation resulting in the formation of a heavily deformed surface layer down to a depth of $\sim 75 \, \mu m$.

Based on the microstructural features observed at various depths and different levels of strain in the deformed surface layer, it can be concluded that, as the strain and strain rate increase, the following changes occur in the microstructure of the heat affected zone of the steel during ultrasonic impact treatment, the details of which are discussed below:

- The formation of low angle misoriented lamellae displaying a high density of dislocations.
- The subdivision of microbands into blocks and the resulting formation of polygonal submicronic grains.
- The further breakdown of submicronic polygonal grains into nano grains.

At larger depths from the treated top surface ($\sim 98 \, \mu m$), the microstructure consists of narrow lamellae. The lamellae show evidence of subdivision into dislocation cells or low angle misoriented blocks. These narrow lamellae look very much like microbands developed in deformed polycrystalline metals which form by splitting of dense dislocations walls (DDWs) with increasing strain [263–266]. At a certain deformation, dislocation annihilation and rearrangement occur in order to minimise the total energy of the system resulting in the formation of individual cells. Such individual cells referred to as dislocation cells are observed inside microbands (indicated by the grey arrows in figure 8.3a). These dislocation cells are at the origin of the formation of subgrain boundaries which will subsequently be converted into low angle misoriented blocks before becoming submicronic polygonal grains. Point-to-point misorientation
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

across such a cell boundary ranges between $2^\circ$ and $3^\circ$. The microstructural observations indicate that additional strain accommodation is achieved by successive grain subdivision. As the strain and strain rate increases, the microstructure of the top treated surface shows the formation of equiaxed nano grains with well delineated grain boundaries. The smaller the nano grains on the top treated surface, the more homogeneous the distribution in the KAM, revealing the absence of large in-grain deformation.

Phase analysis of the top treated surface layer showed that even after severe surface plastic deformation, $\sim$3.7 % austenite was present in the matrix. The average grain size of the retained austenite was $\sim$17 nm. The stabilisation of the retained austenite is attributed to the significant plastic deformation of that phase during the ultrasonic impact treatment. Plastic deformation creates a high density of dislocations, which increases the magnitude of the internal stress of the retained austenite. The geometrically necessary dislocation density $\rho_{GND}$ in the retained austenite is calculated to be $1.68 \times 10^{15}$ m$^{-2}$ using:

$$\rho_{GND} = \frac{\theta_{KAM}}{|\vec{b}| \cdot u \cdot n}$$

where $\theta_{KAM}$ is the kernel average misorientation in radians, $|\vec{b}|$ is the magnitude of burgers vector (taken to be 2.567 nm for austenite), $u$ is the step size and $n$ is the nearest neighbour shell which was taken. The average $\theta_{KAM}$ in retained austenite was $0.988826^\circ$. The bcc transformation requires long-range movement of the interface under the stress $\tau_T$, which originates from the change in chemical free-energy $\Delta G$ of the transformation [267] as:

$$\tau_T = \phi \Delta G$$

where $\phi$ is a constant. The bcc transformation does not occur when the magnitude of the internal stress exceeds $\tau_T$ during ultrasonic impact treatment. The bcc present in the matrix had an average grain size of $\sim$35 nm. However, larger grains of martensite with average diameter measuring $\sim$490 nm were also present in the matrix. Because of the wide distribution of grain sizes, the grains were classified in several size ranges as described previously. The grains with average diameter less than 50 nm (D<50 nm) were equiaxed polygons with an aspect ratio of 0.47. However, with increasing size (diameter), the grains became slightly elongated (aspect ratio of 0.37 for D$\geq$100 nm).

Analysis showed that larger grains are more prone to long range in-grain lattice rotations while smaller grains are prone to grain rotation. In the larger grains, a point-to-origin misorientation traverse reveals the presence of substructures with boundaries...
8.4 Discussion

of misorientation below 15°. Small grains (D<50 nm) were present primarily at the grain boundaries of the larger grains. It is suggested that dislocation cells were the origin of the formation of subgrain boundaries. In order to accommodate plastic deformation, the dislocations rearrange to minimise the total energy of the system. The dislocation cells convert into low angle misoriented blocks before becoming nano-sized polygonal grains. The dislocations rearrange in order to minimise the total system energy.

The in-grain deformation mechanism was grain size dependent. It is observed (figure 8.8) that the distribution of KAM shifts to lower values (the mean KAM decreases) and the mean index increases with increasing grain size, indicating a lower local strain, and hence a lower GND density. High KAM and low index qualitatively indicates that the disordered regions are primarily seen in the smaller grains. Also the smaller nano grains exhibit a homogeneous contrast in the index map revealing the absence of a long range deformation. Table 8.5 also reveals a gradual decrease in the average grain size with increasing grain rotation angle.

While lattice rotation effects cannot be neglected [268], the presence of high angle boundaries are also important. The high angled boundaries are produced due to increased magnitudes of dislocation storage at the cell/subgrain boundaries [269]. As stated by Embury [270], dislocation density at the boundaries for highly strained materials is characterised by an upper limit and shares an inverse relationship with the square of the dislocation spacing. Correspondingly, increasing the levels of imparted strain enhances the likelihood of activation of mechanical annihilation processes (involving in-situ local recovery effects [270]) and produces even larger boundary misorientation angles [271, 272]. The end result of such dislocation accumulation and recovery is the formation of dense dislocation substructures with ever higher angles of misorientation between them.

Owing to the structural characteristics of nano grains described above, it is assumed that a recrystallisation process may play a role in their formation. Such recrystallisation can be considered as dynamic since it occurs during the UIT process. Derby [205] has carried out a systematic analysis of dynamic recrystallisation in relation to two mechanisms: nucleation and the growth of recrystallised grains in a deformed material (classical recrystallisation) and the formation of recrystallisation by the gradual rotation of subgrains (rotation recrystallisation). Both mechanisms lead to the break-up of the original grain structure. In classical or migration recrystallisation, new grains are nucleated in areas of high plastic strain and grow into the deformed material. In rotation recrystallisation, rotation of the cells and subgrains occurs gradually, until all the dislocations are absorbed by the grain boundaries. A very high strain and strain rate is also produced during the UIT process and a certain amount of adiabatic heating may occur but the exact temperature on the top treated surface is difficult to evaluate. The increase in temperature due to adiabatic heating was estimated to be ~42 °C based on similar high frequency peening techniques [239, 273]. Also, TEM
Formation of nanostructures in severely deformed high strength steel induced by high frequency ultrasonic impact treatment

examination does not show any evidence of a nucleation and growth mechanism for nano grains but clearly indicates the importance of grain boundary rotation in micro-structural development. It is therefore suggested that rotation recrystallisation may play a major role in the final grain refinement mechanism during UIT.

Despite a fundamental simplification in considering only three degrees of freedom, the concept of the coincidence site lattice (CSL) is generally accepted for grain boundary classification because it allows an indication of the boundaries with potentially low energy. In comparison with non-coincidence boundaries, a high number of CSL boundaries can improve many materials properties, such as cracking and corrosion resistance [274–277]. Previous studies have shown that cracking occurs preferentially at random boundaries and that low angle and low-Σ CSL boundaries can offer resistance to the propagation of cracks. It was suggested that an optimum grain boundary character distribution described as a high frequency of low angle or low-Σ CSL boundaries and a more discontinuous random boundary network in the microstructure can offer the potential for decreasing the ductile-to-brittle transition temperature [276]. Experimental studies have shown that CSL boundaries of high CSL density/low-Σ do not crack during low cycle fatigue of Ni [278], stress corrosion of a Ni-based superalloy [279], and cavitation of Cu [280]. This is partly attributed to the low-Σ GBs containing a lower distribution of fatigue detrimental intergranular oxides [281]. While intergranular fatigue cracking was non-existent for GBs within 3° of a Σ3 coherent twin, the ratio of failure by this mechanism increased for deviations from 3° to 5° and dominated for GBs with deviations greater than 9° [282]. It has also been observed that GBs dissociate into Σ3n (Σ1, Σ3, Σ9, Σ81, Σ243) type to introduce lower energy segments of boundaries [283, 284]. It has been shown that boundary structure and not just the crystallographic orientation on either side of the boundary is important with regard to the early stages of damage [285]. Cerreta et al. [285] found that boundary type and not only orientation difference across the boundary is a deterministic feature of damage evolution. A reason for this may be related to the way in which line defects interact with grain boundaries [286]. Voids are likely to open at boundaries where stress concentrations develop. During dynamic loading, high densities of line defects are generated [287]. If these defects pile up against a boundary, a stress concentration is likely to develop. If, however, significant fractions of line defects are able to transmit across the boundary or activate secondary slip on the other side of the boundary, these stress concentrations can be diminished [288]. This observation is also supported by the work of Wayne et al. [289] who found that grain boundaries with certain misorientations, in polycrystalline Cu, serve as preferred locations for intergranular damage. These insights regarding damage nucleation at high strain-rates apply equally to damage at lower strain-rates. Work by Mikhailovskij [290] on bcc tungsten shows that under uniaxial stress conditions, special boundaries such as Σ1, Σ3, and Σ9 possess higher resistance to failure in comparison with other types of CSL and non-CSL boundaries. Experiments by Lim [278] on low-cycle fatigue of polycrystalline fcc nickel samples showed that low-order CSL boundaries such as Σ3 and Σ5 did not fail during the deformation process. Evrard [291] and McMurtrey [292] made
similar observations through both experiment and simulations showing that special boundaries were more resistant to crack nucleation in pre-irradiated austentic stainless steels. These results collectively suggest that all grain boundaries, regardless of materials or loading condition, are not equal in terms of their propensity for serving as void nucleation sites. Wayne et al. [289] observed that Σ3 boundaries are less susceptible to damage when compared to grain boundaries with misorientation angles in the 25°-50° range, implying that the latter boundaries are microstructurally weaker. They attributed this to a combination of the high boundary energies associated with these high misorientation angles, leading to lower interfacial strength [288], combined with high property mismatch, which can lead to local stress concentrations [293]. Fensin et al. [294] studied the stresses required for void nucleation and the extent of plasticity at grain boundaries. They found that the Σ3 symmetric tilt boundary was the most resistant (among the boundaries studied) to void nucleation due to its ability to efficiently dissipate stresses associated with mechanical loading via maximum dislocation emission. In the present case, surface nanocrystallisation led to the increased fraction of low angle and low energy CSL boundaries especially in the smaller grains (D<50 nm). The increased fatigue lifetime of welded components via surface modification by the generation of a nanostructured surface layer could be attributed to the presence of a high fraction of low angle and low energy CSL boundaries.

The heavy deformation resulted in recrystallisation at the top treated surface, thereby minimising the total energy of the system. This was supported by grain rotation and by minimising the grain boundary energy. Sangid et al. [295] calculated the grain boundary energies of the various CSL boundaries in a nickel based supper alloy and found that the grain boundary energy of Σ3 boundary (∼110 mJ m⁻²) is an order of magnitude lower than that of Σ5 boundary (∼1270 mJ m⁻²). The top treated surface has the highest fraction of Σ3 boundaries. Σ3 boundaries having the lowest grain boundary energy attributed to minimising the total energy of the system. With increasing depth from the surface, there is a gradual decrease in the applied strain and strain rate, and hence in the amount of deformation. The plastic energy at the subsurface was not enough to recrystallise the microstructure, and therefore the total energy was not minimised in this zone. Hence an increased fraction of Σ5 boundaries with high grain boundary energy is observed at the subsurface region (27 µm and 52 µm below the treated surface).
8.5 Summary

Surface modification by the generation of a nanostructured surface layer induced via UIT was performed at the weld toe. Such high frequency peening techniques are applied to improve the fatigue life of welded components. The nanocrystallised structure at the surface layer was characterised via a recently developed technique for automated crystal orientation mapping in a transmission electron microscope. The observations can be summarised as follows:

- UIT at the weld toe produced a severely deformed nanocrystalline structure. The submicronic structures appear on the surface and down to a depth of \(~75\ \mu\text{m}\) below the top surface.
- The newly developed ASTAR-TEM technique was used to characterise the deformed microstructure. The high spatial resolution of this technique (4 nm) enables effective orientation mapping of nanocrystalline samples with mean grain sizes below 10 nm. The high spatial resolution also allows routine characterisation of highly deformed materials, including those that have undergone room temperature severe plastic deformation and have extremely high dislocation densities.
- TEM investigations show the following changes in the initial coarse grain structure as the strain increases: (1) Formation of dislocation walls, (2) nucleation of microbands associated with the splitting of dislocation walls, (3) subdivision of microbands into low angle misoriented blocks and highly misoriented polygonal submicronic grains, and (4) further breakdown of submicronic polygonal grains into randomly oriented nano grains.
- Phase analysis showed that even after severe surface plastic deformation, \(~3.7\ %\) austenite was present in the matrix on the treated top surface. The average grain size of the retained austenite was \(~17\ \text{nm}\). The bcc present in the matrix had an average grain size of \(~35\ \text{nm}\).
- Larger grains were more prone to long range in-grain lattice rotations while smaller grains are prone to grain rotation. Also the smaller nano grains exhibit a homogeneous contrast in the index map revealing the absence of a long range deformation but presence of short range disorder.
- Surface nanocrystallisation led to an increased fraction of low angle and low energy CSL boundaries especially in the smaller grains (D<50 nm). The increased fatigue lifetime of welded components via surface modification by the generation of a nanostructured surface layer could be attributed to the presence of a high fraction of low angle and low energy CSL boundaries.

Further study needs to be performed on the deformation and grain refinement mechanism due to surface nanocrystallisation via UIT. The quantification of grain boundary
character distribution and the evolution of low angle and low energy CSL boundaries as a function of depth from the top treated surface is necessary to completely understand the reasons behind the improvement of fatigue life of welded components via such peening techniques.
Chapter 9

Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

9.1 Introduction

In order to characterise the heavily deformed surface layer in the TD × ND cross section of the weld, a combination of crystallographic data and imaging capabilities is needed. The present chapter aims at quantitative characterisation of the severely deformed top layers induced by post-weld high-frequency ultrasonic impact treatment using high resolution EBSD and understanding the enhancement in mechanical properties and textural evolution by the generation of nano-sized grains.

9.2 Experimental

The investigated high strength quenched and tempered S690QL1 steel examined has the general composition Fe-0.16C-0.2Si-0.87Mn-0.11Ni-0.33Cr-0.21Mo-0.005Ti-0.02Cu (wt. %) [257]. Details of the welding procedure can be found in chapter 4 and in [258]. The microstructure of the welded zone prior to the ultrasonic impact treatment is shown in figure 9.1. A martensitic microstructure with an average equivalent grain diameter of 1.84 µm was present in the heat-affected zone prior to the

Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

ultrasonic impact treatment. The UIT tool was positioned at the weld toe of the sixth pass at an angle of $30^\circ$ to the normal direction (ND) (see figure 9.2a). The treatment parameters are listed in table 8.1.

![Figure 9.1: Representative EBSD micrograph of the microstructure in the heat affected zone (HAZ) prior to the ultrasonic impact treatment. Overlay of colour-coded inverse pole figure (IPF) and image quality (IQ) map.](image)

Table 9.1: UIT process parameters.

<table>
<thead>
<tr>
<th>Process parameters</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Waveguide frequency, kHz</td>
<td>27</td>
</tr>
<tr>
<td>Waveguide Amplitude, µm</td>
<td>28</td>
</tr>
<tr>
<td>Pin dimensions, mm</td>
<td>Ø3 × 26, R3</td>
</tr>
<tr>
<td>Tool weight, kg</td>
<td>3</td>
</tr>
<tr>
<td>Impact frequency, Hz</td>
<td>120</td>
</tr>
<tr>
<td>Feed rate, mm s$^{-1}$</td>
<td>5</td>
</tr>
<tr>
<td>Number of treatment passes</td>
<td>1</td>
</tr>
</tbody>
</table>
9.2 Experimental

Figure 9.2: (a) Scheme of the experimental arrangement during peening. (b) Optical micrograph of the zone marked in black box in (a). The red dotted line shows the fusion line between the weld and the heat affected zone (HAZ). The black dotted line delineates the surface nanocrystallised zone (SNZ) from the undeformed zone. Regions where the EBSD scans were performed are marked by rectangles.

9.2.1 Microstructure characterisation

To prepare samples for optical microscopy and electron backscatter diffraction (EBSD), the welded and ultrasonically impact treated plates were sectioned along the normal / transverse direction plane. The sectioned specimens were ground and polished for optical microscopy and etched with 5% nital solution to reveal the microstructure. For EBSD the specimens were prepared by standard mechanical grinding and polishing procedures with a 50 nm colloid suspension of SiO$_2$ as a final polishing step. Orientation imaging microscopy was performed using a JEOL JSM 6500F Schottky field emission gun scanning electron microscope (FEG-SEM). A high-speed CCD camera (DigiView) for pattern acquisition was used. The high resolution EBSD scans were carried out at 20 kV with step sizes of 30–100 nm at a working distance of 16 mm. The lateral resolution as determined on iron at 15 kV is around 30 nm parallel to the tilt axis and around 90 nm perpendicular to the tilt axis of the 70° inclined surface [160]. The EBSD maps were processed (confidence index filtering of 0.12) and analysed using TSL® OIM™ analysis software. Figure 9.2b gives an overview of the regions where EBSD scans were performed; details on the scans are given in table 9.2. For calculating parameters such as grain boundary fraction, grain size distribution and kernel average misorientation distribution, the scanned area was sub-divided into layers of 5 µm depth. Textures were calculated over the entire scanned area (as mentioned in table 9.2) to obtain statistically relevant information at each depth.
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

Table 9.2: Orientation mapping characteristics for the EBSD sections.

<table>
<thead>
<tr>
<th>Box colour in figure 9.2b</th>
<th>Depth from the surface</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0–20 $\mu$m</td>
</tr>
<tr>
<td>Step size (nm)</td>
<td>30</td>
</tr>
<tr>
<td>Scanned area ($\mu$m$^2$)</td>
<td>110×20</td>
</tr>
</tbody>
</table>

Electron Channelling Contrast Imaging (ECCI) [251, 296–301] was used to image dislocations in the EBSD scanning regions in a Zeiss Crossbeam SEM equipped with a Gemini field emission gun (FEG) column at an acceleration voltage of 30 kV and a working distance of 6 mm.

9.2.2 Mechanical properties

The effect of the high frequency ultrasonic impact treatment on the mechanical properties of the welded high-strength steel were quantified by means of micro-hardness and residual stress measurements. Vickers micro-hardness measurements were performed using a square pyramidal shaped diamond indenter with a load of 50 g in a Struers DuraScan70 micro-hardness testing machine. The indentations were analysed using workflow-based software (ecos Workflow). Based on the average mean diagonal of the indents, a distance of 60 $\mu$m was maintained between separate indents to ensure that local deformation did not affect subsequent results.

To measure closer to the surface, nanohardness tests were performed using a Hysitron nanoindenter system (TriboIndenter). The instrument included an XYZ sample stage and an arrangement combining a piezo-scanner as known from conventional atomic force microscopy (AFM) with a transducer and a Berkovich indenter tip. The nanohardness tests were conducted in displacement controlled mode. A trapezoid loading-unloading pattern was used with a maximum displacement of 100 nm and a holding time of 5 s at that displacement. The displacement of the indenter was measured simultaneously to give a force-displacement curve. To calculate both hardness and Young’s modulus accurately from the force-displacement data, it was necessary to know the exact geometry of the indenter tip. The indenter cross-sectional area function was determined by performing indentation tests at different depths on quartz. 600 indentation tests were carried out in the form of a 200 (normal direction) × 3 (transverse direction) matrix in the same area where the EBSD scans were performed (figure 9.2). A spacing of 2 $\mu$m along the depth in the thickness (or normal) direction and 4 $\mu$m in the lateral (or transverse) direction between the indents was maintained. The hardness and elastic modulus values were calculated for each indentation from the elastic unloading curves according to the equivalent indenter method [302, 303].
9.2 Experimental

The surface profiles of the indented and surrounding area were determined using AFM.

Through thickness strain measurements in two orthogonal directions were carried out at beamline ID15 of the European Synchrotron Radiation Facility by energy dispersive synchrotron X-ray diffraction. The details of the measurement strategy are discussed by Gao et al. [304]. The planar residual stresses were calculated from the biaxial Hooke’s law,

\[
\sigma_{i,hkl} = \frac{E_{hkl}}{1 - \nu_{hkl}^2} \left( \varepsilon_{i,hkl} + \nu_{hkl} \varepsilon_{j,hkl} \right)
\]

using the diffraction elastic constants of the individual planes [305].
9.3 Results

9.3.1 Microstructure evolution due to post weld ultrasonic impact treatment

The microstructure after the impact treatment as a function of depth from the ultrasonic impact treated surface is illustrated in figure 9.3. A gradual change from ultrafine grains at the surface (figure 9.3b) to a pancake structure (figure 9.3c) at a distance of around 30 µm below the surface is visible. Figure 9.3d shows a lath martensite microstructure in a region 290 µm below the deformed zone.

Figure 9.3: ECCI micrographs of the deformation microstructure at the weld toe after UIT. The dashed white line marks the outline of the weld fusion line. (b-d) show magnified regions of (a). (b) depicts the surface region, (c) the region around 30 µm depth and (d) ~290 µm depth.
9.3 Results

The EBSD scans were performed in four regions (for an overview see figure 9.2b):

- 150–400 µm below the surface, referred to as the undeformed zone (figure 9.4)
- 50–150 µm below the surface (figure 9.5)
- 29.5–52.5 µm below the surface (figure 9.6)
- 0–20 µm below the surface (figure 9.7)

For figures 9.4, 9.5, 9.6 and 9.7 an overlay of image quality (IQ) map and grain boundary network and inverse pole figure (IPF) maps are shown. The grain size distributions for the different zones are presented in figure 9.8. It is observed that the average grain size increases with increasing distance from the surface.
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

Figure 9.4: Microstructure of the undeformed zone (150–400 µm): (a) Overlay of image quality (IQ) map and grain boundary network. High angle grain boundaries (HAGBs) are shown in blue, low angle grain boundaries (LAGBs) below 15° misorientation in red; (b) the corresponding IPF map.
Figure 9.5: Microstructure of the transition zone (50–150 µm): (a) Overlay of image quality (IQ) map and grain boundary network. High angle grain boundaries (HAGBs) are shown in blue, low angle grain boundaries (LAGBs) below 15° misorientation in red; (b) the corresponding IPF map.
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

Figure 9.6: Microstructure of the deformed zone 29.5 µm below the top surface (29.5–52.5 µm): (a) Overlay of image quality (IQ) map and grain boundary network. High angle grain boundaries (HAGBs) are shown in blue, low angle grain boundaries (LAGBs) below 15° misorientation in red; (b) the corresponding IPF map.

Figure 9.7: Microstructure of the deformed zone on the surface (0–20 µm): (a) Overlay of image quality (IQ) map and grain boundary network. High angle grain boundaries (HAGBs) are shown in blue, low angle grain boundaries (LAGBs) below 15° misorientation in red; (b) the corresponding IPF map.
9.3 Results

Figure 9.8: Grain size (diameter) distribution at different depths below the treated surface.

The kernel average misorientation (KAM) was calculated from the second neighbour shell of measurement points, taking a maximum misorientation angle of $5^\circ$ into account. Figure 9.9 shows the gradient of the geometrically necessary dislocation (GND) density $\rho_{GND}$, the high angle grain boundary (HAGB) fraction, the aspect ratio (AR) and the average grain size as a function of depth from the surface. $\rho_{GND}$ was calculated from the average KAM using:

$$\rho_{GND} = \frac{\theta_{KAM}}{|\vec{b}| \cdot u \cdot n} \quad (9.2)$$

where $\theta_{KAM}$ is the kernel average misorientation in radians, $|\vec{b}|$ is the magnitude of the burgers vector (taken to be 0.248 nm for bcc steel [306]), $u$ is the step size and $n$ is the nearest neighbour shell on which the KAM calculation was performed (here 2). The GND density starts with intermediate values close to the surface, reaches a
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

maximum at a depth of about 32 µm before it levels off to lower values in the base material. The fraction of HAGBs is around 80 % in the region close to the surface and decreases at around 32 µm depth after which it remains more or less constant at about 40 % to 50 %.

Figure 9.9: Geometrically necessary dislocation ($\rho_{\text{GND}}$) density, grain boundary (GB) fraction, grain aspect ratio (AR) and average grain sizes ($d_{\text{avg}}$) as a function of depth from the surface. The dynamically recrystallised zone, the transition zone and the bulk material are marked in pink, yellow and grey respectively.

9.3.2 Deformation microtexture

Pole figures and orientation distribution functions (ODFs) were calculated for the undeformed and the deformed zones. ID and IPN refer to the impact direction and impact plane normal respectively. The pole figures in figure 9.10 show the body centred cubic fibers, $\{011\} \parallel$ IPN and $\{111\} \parallel$ ID, developing as the distance to the surface decreases. While the loading configuration represents complex stress states, each of the pole figures contains features expected for body centred cubic shear textures, however none are ideal. Texture analysis of the deformed regions, based on the distance from the surface and intensities, reveals that well-defined simple shear textures develop as the distance to the surface decreases. Pole figures show a gradual lattice rotation around the $\alpha$ fiber to accommodate the imposed severe plastic deformation. This lattice rotation can be observed in terms of the reorientation around the transverse direction (TD) in relation to the ideal orientation $\{011\} \parallel$ ND ($\alpha$ fiber).
9.3 Results

Figure 9.10: Pole figures representing (a) 150–400 µm below the treated surface (undeformed zone), (b) 29.5–52.5 µm below the treated surface and (c) 0–20 µm below the treated surface. ID and IPN refers to the impact direction and impact plane normal respectively.
To quantify the results of the texture analysis, $\varphi_2 = 45^\circ$ ODF section was calculated from the same data as the pole figures in figure 9.10. The ODFs are presented in figure 9.11. In the undeformed zone, the ODF (figure 9.11a) shows a partial $\alpha$ fiber between $\{100\}\langle110\rangle$ and $\{111\}\langle110\rangle$, a $\gamma$ fiber pattern with a minor maximum at $\{554\}\langle225\rangle$ with a strongly rotated Cube component $\{001\}\langle110\rangle$. A shear component with a minor maximum is also observed at $\{112\}\langle111\rangle$. Moving into the deformed zone (229.5–52.5 $\mu$m below the treated surface), the ODF (figure 9.11b) shows the development of a $\gamma$ fiber pattern with minor maxima at (111) $\langle1\overline{2}1\rangle$, (111) $\langle0\overline{1}1\rangle$ and (111) $\langle\overline{1}12\rangle$. Figure 9.11c shows the ODF of the top deformed layer (0–20 $\mu$m below the treated surface). A clear and strong $\gamma$ fiber texture is observed to be a maximum at (111) $\langle1\overline{2}1\rangle$. This texture is also observed in iron/steel that was heavily deformed at high strain rates and subsequently recrystallised [307].
9.3 Results

Figure 9.11: ODFs at $\varphi_2 = 45^\circ$ representing (a) 150–400 $\mu$m below the treated surface (undeformed zone), (b) 29.5–52.5 $\mu$m below the treated surface and (c) 0–20 $\mu$m below the treated surface. (d) $\varphi_2 = 45^\circ$ section of the ODF showing the most representative deformed and recrystallised texture components in BCC materials.


9.3.3 Mechanical properties

Micro-hardness

The local hardness at the weld toe before and after ultrasonic impact treatment is shown in figure 9.12. Prior to the ultrasonic impact treatment, the micro-hardness of the fusion zone is significantly lower than that in the heat affected zone. After treatment, the micro-hardness of the fusion zone is increased, thereby decreasing the hardness gradient across the fusion line.

**Figure 9.12:** Micro-hardness indentation at the welding toe. Optical micrograph (a) before and (b) after ultrasonic impact treatment. Corresponding micro-hardness contour maps (c) before and (d) after ultrasonic impact treatment interpolated from measurement position indicated by the symbol ×.
9.3 Results

Nanoindentation

To compare the variation of nano-hardness and reduced modulus $E_r$ [302, 303] as a function of the different zones as described in figure 9.9, the indented zone was divided into layers of the same thickness over which the parameters in figure 9.9 were calculated and averaged. Figure 9.13 shows the variation of nano-hardness and reduced modulus averaged over layer thickness as a function of depth from the surface. The nano-hardness shows variation between 4 and 7 GPa. The thickness of the layers decreases as the surface is approached. It is evident from figure 9.13 that the nano-hardness increases slightly in the transition zone compared to the dynamically recrystallised zone, then levels off to lower values in the bulk material. The reduced modulus shows a similar trend. $E_r$ is lowest (174 GPa) at the top surface. It reaches a peak (192 GPa) at 50 $\mu$m depth from the surface and then decreases to 177 GPa at a depth of 75 $\mu$m, after which it steadily increases with increasing depth until it reaches a plateau at $\sim$195 GPa.

Figure 9.13: Nanindentation at the welding toe. The variation of nano-hardness and reduced modulus averaged over layer thickness as a function of depth from the surface. The dynamically recrystallised zone, the transition zone and the bulk material are marked in pink, yellow and grey respectively.
Residual stress distribution

The biaxial stress distribution in the transverse direction within the ND/TD plane across the weld before and after ultrasonic impact treatment is shown in figure 9.14. The stresses were calculated from the \{200\} bcc plane as detailed in reference [304]. The maximum tensile stress in the middle of the plate is reduced from 460 MPa to 330 MPa after ultrasonic impact treatment. The highly stressed region in the middle of the plate disappears. The region with intermediate stresses (marked by the green colour) becomes larger to maintain the stress equilibrium in the work piece. Similar trends are observed for the other bcc planes and in the longitudinal direction.

Figure 9.14: Biaxial residual stress in the transverse direction within the ND/TD plane across the weld (a) before and (b) after ultrasonic impact treatment. The symbol \(\times\) indicates the measurement positions. White dashed lines mark the outlines of the weld fusion line.
9.4 Discussion

9.4.1 Microstructural characterisation and texture analysis

EBSD characterisation of the sample reveals the presence of a microstructural gradient between surface and bulk material which is a result of the gradual decrease of the applied strain and strain rate with increasing depth from the treated surface layer. Figures 9.4, 9.5, 9.6 and 9.7 cover the whole range of different microstructures which occur between the highly deformed surface layer and the undeformed bulk. The microstructure can be subdivided into three zones (figure 9.9). In the surface zone (the topmost 20 µm) the effects of the surface treatment are the strongest. This zone is characterised by a high fraction of HAGBs (∼83 %) and intermediate GND density values. At around 32 µm a pronounced microstructural change is observed. There is a rise in the GND density that goes along with a corresponding decrease in the fraction of HAGBs. From here on there is a continuous decrease of the GND density down to the level of the bulk material. The fraction of HAGBs remains constant at about 50 % after the sudden drop at a depth of about 32 µm. The grain size continuously increases from the surface inwards. These microstructural findings indicate the occurrence of dynamic recrystallisation in the surface zone. It is well known that recrystallisation can increase the fraction of HAGBs [308–311] and typically leads to ultrafine grains [307]. Static recrystallisation happening after microstructure deformation at elevated temperatures is known to form nearly strain/defect free grains [201]. The fact that the GND density in the surface zone is lower than in the adjacent layer but higher than in the bulk implies that the surface zone has undergone dynamic recrystallisation. In order for dynamic recrystallisation to happen, the local temperature must have reached values high enough for recrystallisation to occur, i.e. the time for thermal diffusion must have been greater than the strain accumulation time scale. To examine this in detail, estimates of the time to conduct heat away from the top recrystallised zone were made and compared with the rate at which the specimens were displaced. The rate of displacement was utilised as a comparator instead of the rate of strain accumulation because in these specimens, strain is not homogeneous. The rate of displacement, while not constant, is calculated to be 4.74 m s⁻¹ from the ultrasonic vibrations using:

\[ v = 2\pi A_0 \nu \]  \hspace{1cm} (9.3)

where \( \nu \) (= 27 kHz) is the vibration frequency and \( A_0 \) (= 28 µm) is the amplitude. Time \( t \) to conduct the heat over a specified distance \( L \) can be calculated using:

\[ t = \frac{L^2}{D} \]  \hspace{1cm} (9.4)

where \( D \) is the thermal diffusivity. With \( D = 1.2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1} \) [206] and \( L = 20 \mu m \) (the length of the top recrystallised zone), it would take 33 µs to conduct heat over
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

a distance of 20 µm. This leads to a conduction rate of 0.6 m s\(^{-1}\). This value, while admittedly a rough estimate, is one order of magnitude lower than the rate of displacement. As such, this estimate is consistent with competitive time scales for deformation and thermal diffusion for these tests and thus confirms the possibility of the occurrence of dynamic recrystallisation.

However, dynamic recrystallisation is confined to the top 20 µm of the material. This is indicated by the sudden increase of the GND density, correlated with a drop of the fraction of HAGBs 32 µm below the surface. Below this depth the combination of accumulated strain and local temperature is no longer high enough to support recrystallisation, as both factors continuously decrease with distance from the impact surface. The continuous decrease of the accumulated strain with distance from the surface is nicely captured by the continuously decreasing GND density in the transition zone.

The hypothesis of surface recrystallisation is further supported by texture analysis. Analysis of the pole figures (figure 9.10c) revealed the presence of a well-developed shear texture at the top layer of the deformed zone (0–20 µm below the treated surface). The texture is similar to those observed in heavily deformed low carbon steel by similar high strain rate processes [312]. The ODFs figure 9.11 show the development of a clear and strong \(\gamma\) fiber texture with a maximum at (111) \([121]\). This texture is also observed in iron/steel that was heavily deformed at high strain rates and subsequently recrystallised [307].

The most critical parts with respect to crack initiation in welds is the weld toe [313]. Prior to the ultrasonic impact treatment, the weld toe had a coarse grained martensitic microstructure (see figure 8.1) with high hardness (\(\sim 410 H_{V50g}\)). The high frequency peening causes dynamic recrystallisation on the top surface. The heavy deformation causes the microstructure to recrystallise as ultrafine grained ferrite, thereby improving the ductility of the heat affected zone. Additionally, the peening induces compressive stresses into the surface that overcompensate the tensile stresses needed for the formation of surface cracks [18].

At a depth of 29.5–52.5 µm below the treated surface (figure 9.6), where the strain and strain rate is lower than at the surface zone, the pole figures (figure 9.10b) show a developing shear texture. Such textures are similar to the ones observed during dynamic shear of high purity iron [314]. The transition zone is a microstructural region between the undeformed matrix and the fine grained top recrystallised zone and is characterised by elongated grains with a low fraction of high angle boundaries (figures 9.3, 9.6 and 9.9). Such a shear-affected zone dominated by elongated grains with no well-defined shear texture has been observed previously in other metals such as Ta [315] and Cu [316] during high strain rate deformation processes. It is well known that during deformation, dislocations, forming cell walls, will begin to form substructures dominated by low angle boundaries such as subgrains or microbands. With contin-
9.4 Discussion

ued deformation, the misorientation across these dislocation boundaries will increase. However, for the formation of HAGBs by dislocation motion, diffusion-controlled dislocation processes such as climb are necessary which are too slow during dynamic shear loading to explain the high amounts of newly formed HAGBs. A subgrain rotation model called PriSM (Progressive Subgrain Misorientation) was proposed to explain the evolution of microstructure during high strain rate deformation [317]; this model comprises several steps. At the onset of shear band formation, cell subdivision takes place along the direction of the shear. This can be seen from figure 9.10, where body centred cubic fibers representative of well-defined simple shear textures (\{011\} \parallel IPN and \{111\} \parallel ID) develop as the distance to the surface decreases. The original grains break up into a cell structure in order to accommodate the strain. As deformation continues, cells become narrower, evolving into subgrains (as evidenced in figure 9.9 by the decrease in HAGBs in the region between 29.5–52.5 \( \mu \)m below the treated surface) by recovery processes and the latter subdivide into more equiaxed cells. Once a critical cell size is achieved (approximately 200 nm) [318], deformation cannot be accommodated any longer by substructure subdivision, and thus the equiaxed cells start to rotate (this can be qualitatively visualised by comparing pole figures 9.10b and 9.10c). This critical cell size is comparable to the grain sizes in the top recrystallised zone (approximately 200 nm, see figure 9.9). With increasing deformation increasingly higher misorientation angles between some subgrains are developed. The final stage is boundary refinement (recovery) provided the temperature rise and time necessary for dislocation rearrangement during deformation is sufficient. In the transition zone (29.5–52.5 \( \mu \)m below the treated surface), while a fully recrystallised shear deformed substructure was not observed, features that correlate with the early stages of the PriSM model were seen. While these observations certainly are not validation of such a model, they are consistent with the substructural evolution that would occur prior to shear banding according to the PriSM model.

Based on the ODFs of the deformed zone shown in figure 9.11b and 9.11c, both the layers (0–20 \( \mu \)m and 29.5–52.5 \( \mu \)m below the treated surface) show a \( \gamma \) fiber. However, compared to the top recrystallised layer, the grains in the region between 29.5–52.5 \( \mu \)m below the treated surface are weakly oriented to the \( \gamma \) fiber (also shown in the \{111\} pole figure 9.10). This result could also indicate an intense restoration process within this transition zone (dynamic recovery) lowering the intensity of the \( \gamma \) fiber.

9.4.2 Mechanical response

The ultrasonic impact treatment causes surface nanocrystallisation resulting in the formation of a heavily deformed surface layer to a depth of \( \sim \)75 \( \mu \)m. Such a treatment significantly alters the mechanical properties of the sample. Figures 9.12c and 9.12d show that the micro-hardness of the fusion zone increases with such treatment, thereby decreasing the hardness gradient across the fusion line. The micro-hardness
Microstructural gradient induced by surface severe plastic deformation at the toe of a high-strength steel weld

in the scanned zone (around 100 µm from the fusion line) was lowered from 413 to 321 $H_{V_{50}}$. The lowering of the micro-hardness is attributed to recrystallisation. The nanoindentation tests also confirm the lowering of the nano-hardness and the reduced modulus of the top recrystallised layer (figure 9.13). The nano-hardness and the reduced modulus $E_r$ reaches a peak in the transition zone, due to an increase in the dislocation density (figures 9.9 and 9.13) and then levels off to lower values in the bulk material. Compressive surface stresses are known to retard surface crack formation, tensile surface stresses to accelerate them [319]. After the ultrasonic impact treatment, the maximum tensile stress in the middle of the plate is reduced. In some regions on the top surface below the treated zone even compressive stresses are observed (figure 9.14b). The overall tensile stress reduction contributes to the positive effect of ultrasonic impact treatment on the mechanical properties of a weld.

9.4.3 Improvement in fatigue strength of welded components due to UIT

The fatigue cracks in welded joints usually initiate at the specimen weld-toes, in the HAZ [313]. High tensile residual stresses are produced by welding at the weld toe [320]. The near surface tensile stresses tend to accelerate the initiation and growth stages of fatigue cracks [319]. A further reason for the initiation of fatigue cracks at weld-toes is the formation of coarse grained brittle martensite in the heat affected zone, which is formed due to rapid cooling rates.

Post weld ultrasonic impact treatment (UIT) has already been shown to improve the fatigue life of welded components due to the elimination on the tensile residual stresses by generating compressive residual stresses [319, 321]. The current study reveals several further reasons for the enhancement of the fatigue life of welded components by UIT. The surface layer has been shown to undergo dynamic recrystallisation, which is a diffusional process. The crystal phase of the newly formed surface grains is thus ferritic, which is much more ductile than the original martensitic microstructure prior to UIT. According to figure 9.9, UIT results in significant grain size refinement, which is known to enhance a material’s strength without sacrificing ductility. This provides a further reason for the enhanced mechanical properties. The dislocation density in the recrystallised surface layer is lower than in the transition zone (figure 9.9). Hence the surface layer has work-hardening capacity, as it is not yet completely saturated with dislocations. Recrystallisation results in an increased fraction of high angle grain boundaries at the surface. Fatigue crack propagation being primarily transgranular in nature [18], the generation of high angle grain boundaries at the surface would also contribute to improving the resistance to fatigue crack propagation.
Fatigue cracks in metals are generally initiated from the surface, through the formation of intrusions and extrusions under an alternating stress \cite{18}. To improve the fatigue strength, the surface must be protected from the generation of slip steps at low stress amplitudes. This can be achieved by arresting the dislocations escaping from the inside to the surface. The restraint of dislocations is generally achieved either by the increase of friction stress or by the reduction in applied stress. While surface modification methods such as nitriding or carburising and quenching are mainly based on the former effect, the stress applied to the surface can be decreased by the formation of surface-layers with a Young’s modulus lower than that of the substrate. In this case, since the surface can be protected by the stress redistribution due to the difference of Young’s moduli, an improvement in the fatigue strength is expected. Studies by Morita et al. have shown the improvement in fatigue strength due to decreased surface layer elastic modulus of pure iron by the formation of a titanium layer \cite{322}. Nanoindentation tests show that the nano-hardness and the reduced modulus decreases in the top surface layer as a result of recrystallisation which provides further proof for the improvement in fatigue strength of welded high strength steel by ultrasonic impact treatment.

9.5 Conclusions

Surface modification by the generation of a nanostructured surface layer induced via UIT was performed at the weld toe. Such high frequency peening techniques are applied to improve the fatigue life of welded components. The nanocrystallise structure at the surface layer was characterised via high resolution FEG–EBSD. The observations can be summarised as follows:

1. Surface modification by ultrasonic impact treatment creates a microstructural gradient which can be explained by the gradual decrease of the applied strain and strain rate with distance from the UIT surface.

2. A surface layer of 20 μm thickness is characterised by refined ultrafine grains, a high fraction of HAGBs, GND density levels lower than the next deeper layer but higher than in the bulk, and a well-defined simple shear texture (\{011\} \parallel IPN fiber). All of this is correlated evidence for the occurrence of dynamic recrystallisation within 20 μm distance of the surface.

3. The layer between 20 μm and 150 μm depth is characterised by a continuous increase in grain size, a constant fraction of HAGBs at intermediate levels, a sharp increase in GND densities compared to the surface layer, that levels off to values of the bulk material in depth; and a developing shear texture. The microstructural changes in this zone can be described by recovery.

4. The microstructural changes caused by ultrasonic impact treatment significantly alter the mechanical properties of the sample. The micro-hardness of the fusion
zone increases with such treatment, thereby decreasing the hardness gradient across the fusion line. The lowering of the micro and nano-hardness in the top 20 µm from the surface is attributed to recrystallisation. The maximum tensile residual stress in the middle of the plate is reduced. Some surface regions contain compressive stresses.
Chapter 10

Conclusion and recommendations for future work

10.1 General conclusions

The experimental investigation of the microstructural development during welding and high frequency post weld impact treatments for high strength quenched and tempered structural S690QL1 steel was carried out in this work, and the following conclusions are derived:

1. The temperature dependent plane specific diffraction elastic constants (DECs) of ferrite in a high strength quenched and tempered structural S690QL1 steel have been determined with a high degree of precision for the first time. For this purpose, in-situ tensile tests have been carried out at different temperatures in a high energy synchrotron X-ray diffractometer. The data reported are of considerable significance, since they lead to a more accurate determination of the residual stress states from the measured local d-spacings between lattice planes than when using averaged elastic constants.

2. The evolution of local d-spacings between lattice planes of bainitic ferrite in a high strength quenched and tempered structural S690QL1 steel has been used to determine the thermal expansion behaviour. For the first time it has been shown that thermal anisotropy exists in bainitic ferrite.

3. Surface modification by the generation of a nanostructured surface layer induced via ultrasonic impact treatment was performed at the weld toe of a welded high strength quenched and tempered structural S690QL1 steel. The nanocrystallised structure as a function of depth from the top treated surface
was characterised. A grain refinement mechanism induced by plastic deforma-
tion during the ultrasonic impact treatment is proposed involving the formation
of low angle misoriented lamellae displaying a high density of dislocations fol-
lowed by the subdivision of microbands into blocks and the resulting formation
of polygonal submicronic grains. These submicronic grains further breakdown
into nano grains.

4. Severe surface plastic deformation produces grains with an average size of \(\sim 35\) nm
at the surface. Analysis shows that these grains are more prone to grain rotation
than in-grain deformation. The change in deformation mechanism, avoiding the
formation of slip band intrusions and extrusions, is an important contributing
factor in improving fatigue lifetime.

5. Surface nanocrystallisation generates an increased fraction of high angle grain
boundaries at the surface, resulting in improved resistance to fatigue crack
propagation, which is ordinarily transgranular.

6. Surface severe plastic deformation induced via ultrasonic impact treatments
resulted in the formation of a dynamically recrystallised zone down to a depth of
20 \(\mu m\) from the surface and the formation of strain free grains. Hence the surface
layer has work-hardening capacity as it is not yet completely saturated with
dislocations. The recrystallisation results in the formation of nanocrystalline
ferritic grains in place of the original brittle martensitic microstructure; hence
a higher ductility is achieved.

7. It has been shown for the first time that surface nanocrystallisation induced via
ultrasonic impact treatment leads to an increased fraction of low-angle and
low energy coincident site lattice (CSL) boundaries on the top surface. In
comparison with non-coincidence boundaries, a high number of CSL boundaries
can improve many material properties, such as cracking and corrosion resistance.

8. Nanoindentation tests show that the nano-hardness and the reduced modulus
decreases on the top surface layer as a result of recrystallisation which provides
further proof for the improvement in fatigue strength of welded high strength
steel by ultrasonic impact treatment.
The experimental investigation on the effect of tensile deformation with in-situ ultrasonic treatment on the microstructure of low carbon steel was also carried out in this work. It has been shown for the first time that the in-situ ultrasonic treatment results in:

i a decrease in dislocation density,

ii substantial reduction in sub-grain formation,

iii significant decrease in the fraction of low-angle grain boundaries,

iv concentration of dislocations in the vicinity of the grain boundaries,

v significant reduction in the extent of grain deformation and in-grain orientation gradient,

vi spreading of the orientation gradients along the grain boundaries in the direction of propagation of ultrasound,

vii inhomogeneous deformation, as shown by the mixture of equiaxed grains and very elongated grains giving rise to a bimodal grain size distribution. It was shown through EBSD that plastic deformation is accommodated by ultrasound induced grain rotation.

10.2 Recommendations for future work

Although, an extensive experimental research on the microstructural development during welding and post weld high frequency ultrasonic impact (HFUI) treatment of high strength steel was carried out in this work, there is scope for future developments that can be pursued on the basis of the present study.

In the present study, the microstructural evaluation has been performed on one model HFUI treatment parameter. Residual stress measurements have shown that the stress state changes with the treatment parameters. The study can be further extended to investigate the effect of different treatment parameters on the microstructural evolution.

Another interesting field of further study could be residual stress distributions close to the surface. In the present study, the residual stress measurements were carried out in beamline ID15b at ESRF, Grenoble. A high energy white beam, in transmission mode, was used to study the stress state in the welded and post HFUI treated plates. However, one drawback of the procedure was that the measurements could not be performed within the first 1 mm of the surface. Microstructural evaluation has shown that the depth of the nanocrystalline zone (after HFUI treatment) is approximately 75 µm. It would be interesting to probe into the residual stress distribution of this nanocrystalline zone using a high energy synchrotron source with a nano beam in
Conclusion and recommendations for future work

reflection mode.

In the present study, the HFUI treatment was carried out post welding. Another area of investigation could be the use of the treatment during welding. It would be worth investigating if the ultrasonic vibrations affect the weld pool flow and solidification and its effect on the final properties.

Although the present use of post weld high frequency impact treatments at the toe of a high strength steel weld proved to improve the microstructure, it is unknown how the treatment will affect other materials or phases. Excessive peening is known to damage the surface and hence be detrimental to the microstructure and subsequent properties. More research should be performed on the effect of post weld high frequency impact treatments on different materials/phases to determine whether the improvement in microstructure is universal. Also the limit in degree of severity of the treatment for which it is beneficial to the microstructure/properties should be examined.
References


[31] A. V. Kulemin, Ultrasonics and diffusion in metals, Metallurgiya.


References


[76] F. S. LePera, Improved etching technique for the determination of percent martensite in high strength dual-phase steels, Metallography 12 (1979) 263–268.


[83] Dillimax 690: High strength fine grained structural steel, quenched and tempered, Dillinger Hütte GTS.


[85] ASTM, Standard specification and temperature - electromotive force (EMF) tables for standardised thermocouples.


noise camera as a versatile x-ray detector for time resolved dispersive extended x-ray absorption fine structure and diffraction studies of dynamic problems in materials science, chemistry, and catalysis, Review of Scientific Instruments 78 (9).


References


168


[190] P. A. Gordon, T. Neeraj, Y. Li, J. Li, Screw dislocation mobility in bcc metals: The role of the compact core on double-kink nucleation, Modelling and Simulation in Materials Science and Engineering 18 (8).


References


[252] E. F. Rauch, M. Veron, Coupled microstructural observations and local texture measurements with an automated crystallographic orientation mapping tool attached to a tem, Materialwissenschaft und Werkstofftechnik 36 (10) (2005) 552–556.


[300] B. C. Ng, B. A. Simkin, M. A. Crimp, Application of the electron channeling contrast imaging technique to the study of dislocations associated with cracks in bulk specimens, Ultramicroscopy 75 (3) (1998) 137–145.


Summary

1. Base material microstructure and thermal and mechanical properties

The as-received S690QL1 is a quenched and tempered martensitic steel with 0.16 wt. % carbon and a carbon equivalent ($CE_{IIW}$) of 0.42. The average hardness is $\sim 260 \, H_{V_{50gf}}$. The tensile behaviour of S690QL1 base metal samples showed a typical stress-strain curve with continuous yielding and a completely ductile failure. The results show that the average yield stress of the S690QL1 base metal at 0.2 % offset is 828 MPa. With an average ultimate tensile stress of 868 MPa, the base metal showed a maximum average elongation of 9.81 mm with a total tensile strain of 11.8 %. The average Young’s modulus was 210 GPa. The temperature dependent elastic properties were calculated from the elevated temperature tensile tests. Young's modulus and the yield stress both decrease with increasing temperature. Grain coarsening occurs with increasing temperature which results in a decrease of the yield stress. Dilatation measurement shows the formation of bainite (of varying fractions) at cooling rates between 8 and 30 °C s$^{-1}$ and martensite at cooling rates above 30 °C s$^{-1}$.

The temperature dependent plane specific diffraction elastic constants (DECs) were determined with a considerable degree of precision. For this purpose, in-situ tensile tests have been carried out at different temperatures in a high energy X-ray diffractometer. For the planes aligned with the plane-normal, parallel to the loading axis, a greater value of $A_{hkl}$ implies a greater stiffness $E_{hkl} = \sigma / \varepsilon_{hkl}$. Hence for ferrite, the $\langle 200 \rangle$ direction is the most compliant crystallographic direction in the axial loading direction with $A_{200} < A_{110} = A_{211} = A_{220}$ and therefore $E_{200}^\alpha < E_{110}^\alpha = E_{211}^\alpha = E_{220}^\alpha$. Figure 5.6 confirms these predictions. Along the loading direction, the $\{200\}$ ferrite plane is most strained by the applied stress at all temperatures. A comparable strain development for the $\{110\}$, $\{211\}$ and $\{220\}$ ferrite planes is observed up to the yield point. The qualitative elastic response observed experimentally is well predicted by a cubic elastic anisotropy factor.

The evolution of local d-spacings between lattice planes during continuous cooling from austenite to bainitic ferrite in S690QL1 steel were determined to calculate the thermal expansion behaviour. For this purpose, in-situ continuous cooling tests carried out in a high energy synchrotron X-ray diffractometer. The results indicate
anisotropic thermal expansion coefficients in the bainitic ferrite planes. The aniso-
tropic CTE could arise due to the tetragonality in bainitic ferrite. Recently tetragonal
or slightly orthorhombic unit cell of bainitic ferrite was observed [116]. The tetragon-
ality in bainitic ferrite could arise from the excess carbon that persists in the ferrite,
which is in contact with austenite. This is a consequence of an increased solubility
due to the change in symmetry from the conventional cubic unit cell.

2. Welding experiments and microstructure

Multi-layer deposition was performed for welding of 16 mm thick S690QL1 plates.
The first weld pass was made with a relatively low heat input to avoid burn through.
Subsequent weld passes, were carried out under similar heat inputs but with different
metal transfer characteristics (spray mode). No preheating was required. Successive
thermal cycles introduced additional local re-melting, solid state phase transform-
ations, grain growth, grain refinement, recrystallisation and recovery, all of which
results in a complicated and evolving stress state. Analysis of weld cross sections re-
vealed little visible porosity. Microstructural analysis showed a gradient from acicular
ferrite in the fusion zone to primarily Widmanstätten ferrite and martensite in the
heat affected zone and tempered martensite in the base metal. The fusion zone con-
tains complex inclusions. Morphological analysis indicates that the inclusions in the
fusion zone consist of aluminium, silicon and copper oxides and sulphides.

The overall hardness in the fusion zone ranges between 240 and 280 $H_{V_{500g_f}}$, which
primarily consists of acicular ferrite. The hardness rises to a range of 280 to 320
$H_{V_{500g_f}}$ in the weld metal close to the fusion boundary. This could be due to the
formation of martensitic structures in this region because of the faster cooling rates.
In the fusion zone, the heterogeneity in the grain sizes and the presence of martensite
and acicular products dictates the variation of hardness in the solidified microstruc-
ture. The hardness values drop again to a range between 200 to 240 $H_{V_{500g_f}}$ close
to the fusion boundaries due to grain coarsening in the heat affected zone combined
with the effects of the formation of polygonal ferrite in the inter-critical heat affected
zone.

3. The effect of tensile deformation with in-situ ultrasonic treatment on
the microstructure of low carbon steel

The softening effect in metals due to ultrasonic vibration is used in many indus-
trial applications. The existing understanding of such an acoustoplastic effect is one
in which the ultrasonic treatment either imposes additional stress waves to supple-
ment the quasi-static applied load or causes heating of the metal. The effect of an
in-situ ultrasonic treatment on the microstructure of low-carbon steel (Fe-0.051C-
0.002Si-0.224Mn-0.045Al (wt. %)) under tensile deformation was investigated by a
combination of optical microscopy, scanning electron microscopy, crystal orientation
mapping by electron backscattered diffraction and X-ray diffraction. The results show
that the dislocation density and the fraction of low-angle grain boundaries decrease significantly, accompanied by preferential grain rotation. The softening effect of the ultrasound is found to drive recovery associated with significant reduction in sub-grain formation during deformation. By comparing the microstructures of samples deformed with and without simultaneous application of ultrasound, the reduction in sub-grain formation is shown to occur due to the combined application of the quasi-static loading and the ultrasound, but is not a simple addition of the two factors acting separately. The effect of the ultrasound can be attributed to its ability to enhance dislocation dipole annihilation. The superimposed ultrasound causes dislocations to travel longer distances thereby increasing the probability of annihilation. A bimodal grain size distribution is observed with relatively small equiaxed grains with an average diameter of 10 µm at the grain boundaries of large elongated grains. The formation of these relatively small equiaxed grains is interpreted in terms of dynamic recrystallisation by lattice and sub-grain rotation.

4. Microstructural evolution at the weld toe of S690QL1 steel due to high frequency ultrasonic impact treatment (UIT)

UIT at the weld toe causes severe plastic deformation at the surface. This introduces nano grains and grain size gradients into the surface region of bulk materials. The formation of a nanocrystalline microstructure can result in a considerable improvement in mechanical properties, such as the low and high temperature strength, fatigue resistance and tribological properties. The nano grains introduced at the top layers at the weld toe were studied along the RD × TD cross section of the weld using automated crystallographic orientation mapping in a transmission electron microscope (TEM). With automated crystal orientation mapping in TEM it is possible to couple the advantages of the TEM and EBSD, i.e. to create orientation maps with the high spatial resolution of TEM.

Surface modification by the generation of a nanostructured surface layer induced via ultrasonic impact treatment was performed at the weld toe of a welded high strength quenched and tempered structural steel, S690QL1 (Fe-0.16C-0.2Si-0.87Mn-0.33Cr-0.21Mo (wt. %)). Such high frequency peening techniques are known to improve the fatigue life of welded components. The nanocrystallised structure as a function of depth from the top treated surface was characterised via a recently developed automated crystal orientation mapping in transmission electron microscopy. Based on the experimental observations, a grain refinement mechanism induced by plastic deformation during the ultrasonic impact treatment is proposed. It involves the formation of low angle misoriented lamellae displaying a high density of dislocations followed by the subdivision of microbands into blocks and the resulting formation of polygonal submicronic grains. These submicronic grains further breakdown into nano grains. The results show the presence of retained austenite even after severe surface plastic deformation. The average grain size of the retained austenite and martensite is ~17 nm and ~35 nm respectively. The in-grain deformation mechanisms are different in
larger and smaller grains. Larger grains show long range lattice rotations while smaller grains show plastic deformation through grain rotation. Also the smaller nano grains exhibit the presence of short range disorder. Surface nanocrystallisation also leads to an increased fraction of low-angle and low energy coincident site lattice (CSL) boundaries especially in the smaller grains (D<50 nm).

The above technique presents many advantages, compared to the classical EBSD technique in the scanning electron microscope (SEM). The most notable is the high spatial resolution (∼4 nm in the present case). However, with such a technique, the measured area is rather small and hence it is difficult to obtain statistical representativeness of the obtained data. High resolution electron backscattered diffraction (EBSD) was therefore carried out in the TD × ND cross section of the weld. This was possible due to the slightly larger grain sizes along the normal direction. The nanocrystalline structure and texture was characterised as a function of depth from the impacted surface via high-resolution electron backscatter diffraction (EBSD). Results indicate that the top 20 µm undergo dynamic recrystallisation. The processes occurring in 20-175 µm depth are consistent with progressive subgrain misorientation (PriSM). Recrystallisation results in lowering of hardness in the top treated zone.
1. Basismateriaal, thermische en mechanische eigenschappen

Het in dit onderzoek gebruikte staal, S690QL1, is een gehard en ontlaten staal met 0.16 gew. % koolstof en een koolstofequivalent (CEIIW) van 0.42. De gemiddelde hardheid is ongeveer $260 \, H_{V500gf}$. Onder trekbelasting laat dit basismetaal een typische spanning-rek curve zien met continu vloeien en een volledig taaie breuk. Uit de resultaten komt naar voren dat de gemiddelde rekgrens van het basismetaal 828 MPa is. Bij de gemiddelde treksterkte van 868 MPa ondergaat het materiaal een gemiddelde maximale verlenging van 9.81 mm en een totale rek van 11.8 %. De gemiddelde elasticiteitsmodulus was 210 GPa. Uit trekproeven op verhoogde temperatuur zijn de temperatuurafhankelijke elastische eigenschappen bepaald. De elasticiteitsmodulus en de vloeigrens nemen beide af met toenemende temperatuur. Korrelgroei treedt op bij temperatuurtoename en resulteert in een afname van de vloeigrens. Uit dilatatiemetingen blijkt dat bij afkoelingsnelheden tussen 8 en 30 °C s$^{-1}$, bainiet in variërend fracties wordt gevormd en bij afkoelsnelheden boven 30 °C s$^{-1}$ martensiet.

De temperatuurafhankelijke vlakken specifieke elastische diffractie constanten zijn met een grote mate van nauwkeurigheid bepaald. Hiertoe zijn in een hoogenergetische Röntgen diffractometer bij verschillende temperaturen in-situ trekproeven uitgevoerd. Voor vlakken samenvallend met de normaalrichting van een vlak, parallel aan de richting van belasting, impliceert een grotere waarde van $A_{hkl}$ een grotere stijfheid $E_{hkl} = \sigma/\varepsilon_{hkl}^\parallel$. Voor ferriet is de $\langle 200 \rangle$ richting de meest meegevende kristallografische richting bij axiale belasting waarbij $A_{200} < A_{110} = A_{211} = A_{220}$ en dus is $E^{\alpha}_{200} < E^{\alpha}_{110} = E^{\alpha}_{211} = E^{\alpha}_{220}$. Figuur 5.6 bevestigt deze voorspelling. In de belastingrichting ondervindt het $\{200\}$ ferrietvlak bij alle temperaturen de grootste rek onder de aangelegde spanning. Een vergelijkbare ontwikkeling van de rek is voor de $\{110\}$, $\{211\}$ en $\{220\}$ ferrietvlakken tot aan de vloeigrens waargenomen. De experimenteel bepaalde kwalitatieve elastische respons wordt goed voorspeld door de kubisch elastische anisotropiefactor.

De evolutie van de lokale afstanden tussen de vlakkenscharen in S690QL1 staal tijdens continue afkoeling van austeniet naar bainitisch ferriet zijn bepaald om het thermisch uitzettingsgedrag te berekenen. Hiertoe zijn in-situ continue afkoelingsexperimenten
uitgevoerd in een hoogenergetische synchrotron Röntgen diffractometer. De resultaten tonen anisotrope thermische uitzettingscoëfficiënten van de bainitische ferrietvlakken. De anisotropie in de thermische uitzettingscoëfficiënten kan optreden door de tetragonaliteit in bainitisch ferriet. Recentelijk zijn tetragonale of licht orthorombische eenheidscellen in bainitisch ferriet waargenomen [116]. De tetragonaliteit in bainitisch ferriet kan het gevolg zijn van de overmaat aan koolstof dat aanwezig blijft in ferriet dat in contact is met austeniet. Dit is een consequentie van een verhoogde oplosbaarheid door de verandering in symmetrie ten opzichte van conventionele kubische eenheidsceel.

2. Lasexperimenten en microstructuur

Meerlagenlassen zijn vervaardigd in 16 mm dik S690QL1 staalplaat. De eerste las is gemaakt met een relatief lage warmteïnbreng om doorbranden te voorkomen. De daaropvolgende lassen zijn vervaardigd met vergelijkbare warmteïnbreng maar met een ander type metaaltransport (sproeiboog). Voorverwarmen was niet noodzakelijk. De thermische cycli van de successiefelijke lassen zorgen voor lokaal hersmelten, vaste stof fasetransformaties, korrelgroei, korrelverfijning, rekristallisatie en herstel. Hierbij ontwikkelt zich de spanning op een complexe manier. Analyse van lasdoorsneden van de las liet nauwelijks de aanwezigheid van porositeit zien. De microstructurele analyse toont een gradint van naaldferriet in het lasmetaal, via hoofdzakelijk zijplaatferriet en martensiet in de warmtebeïnvloede zone naar ontlaten martensiet in het basismateriaal. Het lasmetaal bevat complexe insluitsels. Morfologische analyses hiervan wijzen op aluminium-, silicium- en koperoxiden en -sulfiden.

De gemiddelde hardheid in het hoofdzakelijk naaldferriet bevattende lasmetaal ligt tussen de 240 en 280 $H_{V500g_f}$. De hardheid neemt toe tot waarden tussen 280 en 320 $H_{V500g_f}$ in de buurt van de smeltlijn. Dit kan het gevolg zijn van de vorming van martensiet in deze zone door de hogere afkoelsnelheden. De variatie in hardheid in het lasmetaal komt door de heterogeniteit in korrelgrootte en de aanwezigheid van martensiet en naaldvormige transformatieproducten. De hardheid neemt in de warmtebeïnvloede zone, in de buurt van de smeltlijn, af tot 200–240 $H_{V500g_f}$ door korrelgroei en de vorming van polygonale ferriet in de interkritische warmtebeïnvloede zone.

3. Het gecombineerde effect van deformatie onder trekbelasting en in-situ ultrasone behandeling op de microstructuur van laagkoolstofstaal

Het effect dat metalen zwakker worden door ultrasone trillingen wordt in tal van industriële toepassingen aangewend. De verklaringen voor dit acoustoplastische effect door de ultrasone behandeling worden gezocht in de introductie van aanvullende spanningsgolven op de quasi-statische opgelegde spanning of in verhitting van het metaal. Het effect van een in-situ ultrasone behandeling tijdens deformatie onder een trekbelasting op de microstructuur van een laagkoolstofstaal (Fe-0.051C-0.002Si-0.224Mn-0.045Al...
(gew. %)) is onderzocht door middel van optische microscopy, scanning elektronenmicroscopie en het in kaart brengen van de kristaloriëntatie met ‘electron backscattered’ diffractie en Röntgendiffrahtie. De resultaten tonen dat de dislocatiedichtheid en de fractie kleine-hoek-korrelgrenzen significant afnemen, waarbij voorkeurskorrellrotatie optreedt. Het verzwakkende effect van ultrasound bevordert herstel, gepaard gaand met een significante afname van de vorming van subkorrelgrenzen tijdens deformatie. Door het vergelijken van de microstructuur van preparaten vervormd met en zonder het toepassen van ultrasonic trillingen is aangetoond dat de reductie in de vorming van subkorrels niet eenvoudigweg de som is van beide afzonderlijke effecten. Het effect van ultrasonic trillingen kan worden toegeschreven aan het vermogen tot het bevorderen van dislocatiedipool annihilation. Ultrasonic trillingen maken het mogelijk dat dislocaties een grotere afstand afleggen waarbij de kans op annihilation toeneemt. Een bimodale korrelgrootteverdeling is waargenomen met relatief kleine gelijkvormige korrels met een gemiddelde diameter van 10 µm aan de korrelgrenzen van grote uitgerekte korrels. De vorming van deze kleine korrels kan geïnterpreteerd worden in termen van dynamische rekristallisatie door rooster en sub-korrel rotatie.

4. Evolutie van de microstructuur aan de lasteen van S690QL1 staal door de hoogfrequente ultrasonic impact treatment (UIT)

UIT van de lasteen resulteert in zware plastische deformatie aan het oppervlak. Dit introduceert nano-korrels en korrelgroottegradiënten in de zone van het oppervlak. De vorming van een nano-kristallijne microstructuur kan leiden tot een aanzienlijke verbetering van de mechanische eigenschappen, zoals de lage en hoge-temperatuursterkte, de vermoeiingsweerstand en tribologische eigenschappen. De nano-korrels aan het oppervlak bij de lasteen zijn bestudeerd in een bovenaanzicht van de las door middel van het geautomatiseerd in kaart brengen van de kristallografische oriëntatie in een transmissie elektronen microscoop (TEM). Dit maakt het mogelijk de voordelen van TEM en EBSD te combineren, te weten het creëren van oriëntatiekaarten met de hoge ruimtelijke resolutie van een TEM.

Oppervlaktemodificatie door het genereren van een nano-structuur door middel van UIT is uitgevoerd aan de lasteen van een gehard en ontlaten hoge sterkte constructiestaal, S690QL1 (Fe-0.16C-0.2Si-0.87Mn-0.33Cr-0.21Mo (gew. %)). Het is bekend dat de vermoeiingslevensduur van gelaste componenten toeneemt bij het toepassen van dergelijke hoogfrequente impact technieken. De nano-kristallijne structuur is gekarakteriseerd als een functie van de afstand tot het behandeld oppervlak door middel van de recentelijk ontwikkelde methode van het geautomatiseerd in kaart brengen van de kristaloriëntatie in een transmissie elektronen microscoop. Gebaseerd op de experimentele waarnemingen is een korrelverfijningsmechanisme ten gevolge van plastische deformatie tijdens UIT voorgesteld. Het houdt de vorming van lamellen met een kleine hoek misoriëntatie en een hoge dislocatiedichtheid, gevolgd door het opdelen van microbanden in blokken en de resulterende vorming van polygonale submicron grootte korrels. Deze submicron-korrels breken verder af tot nano-korrels. De res-
ultaten tonen de aanwezigheid van restausteniet, zelfs na zware plastische deformatie van het oppervlak. De gemiddelde korrelgrootte van het restausteniet en het marten- siet is respectievelijk $\sim 17$ en $\sim 35$ nm. De deformatiemechanismen in de korrel zijn verschillend voor grotere en kleinere korrels. In de grote korrels vindt lange afstand roosterrotatie plaats, terwijl in kleinere korrels plastische vervorming plaatsvindt door korrelrotatie. De kleinere nano-korrels laten ook de aanwezigheid van wanorde over korte afstand zien. Nano-kristallisatie aan het oppervlak geeft ook aanleiding tot een toename in de fractie kleine hoek en lage energie ‘coincident site lattice’ (CSL) grenzen, vooral in kleinere korrels ($D<50$ nm).

De bovengenoemde techniek heeft vele voordelen in vergelijking tot de klassieke EBSD techniek in de scanning elektronenmicroscoop (SEM). Het meest opmerkelijke is de grote ruimtelijke resolutie ($\sim 4$ nm in het huidige geval). Met zo’n techniek is het gemeten gebied echter klein en dus is het moeilijk data te verkrijgen die statistisch representatief is. Hoge resolutie electron backscattered diffractie (EBSD) is daarom uitgevoerd in een dwarsdoorsnede van de las. Dit was mogelijk door de enigszins grotere korrels in de dikterichting. De nano-kristallijne structuur en textuur is gekarakteriseerd als functie van de afstand tot het behandelde oppervlak door middel van hoge resolutie ‘electron backscattered’ diffractie. Uit de resultaten komt naar voren dat de 20 µm dikke toplaag dynamische rekristallisatie plaats vindt. De processen die optreden in de laag van 20 tot 175 µm zijn te verklaren door een toenemende (progressieve) subkorrel misoriëntatie (PriSM). Rekristallisatie zorgt voor een daling van de hardheid van de bovenste laag in de behandelde zone.
Acknowledgement

The Ph.D. research project, which I started roughly four years ago in September 2010 has finally culminated in the publication of this thesis. My journey in Delft started six years ago, in the August of 2008, when I started with my M.Sc. in the department of MSE. During the course of my PhD research project, there have been numerous ups and downs. A number of people have supported me throughout and they deserve special mention. I would like to express my deepest appreciation to all those who provided me the possibility to complete this research.

First, I would like to thank Monica Reulink from M2i. Without her persistent but professional approach, this would never have started. I would also take the opportunity to thank the whole team from M2i, namely Gaby Cuss, Brigitte van Uden, Bert van Haastrecht, Gitty Bouman and Oscar Ruigrok, who made it possible to start my research in Delft University of Technology.

During my PhD, I have learned that the success of the candidate and the project is as dependent on the candidate himself as is on the supervisor(s). In that respect, I would like to express my deepest appreciation and gratitude to my daily supervisor dr. ir. Marcel Hermans and my promoter prof. dr. Ian Richardson. To both of you, I am forever indebted for providing a stimulating and challenging research environment all the while supporting me through the more risky endeavours. Both of you have pushed me to think independently and decisions on my own from the beginning. This helped me to develop a critical attitude towards my own work and evolve into a more independent researcher than would have been otherwise possible.

I also greatly appreciate the support of Allseas Engineering bv for this project and special thanks to dr. Natalia Ermolaeva from Allseas, who provided very important inputs for the project. I am also thankful to Werner Smakman of Applied Ultrasonics Europe for providing the ultrasonic impact treatment tool and the training.

A special mention goes to dr. Murugaiyan Amirthalingam (Muru), Richard Huizenga and my office mate He Gao, with whom I worked very closely during my PhD. Even though I kept on disturbing you at times over journal papers and data analysis, you were always willing to help and engage in fruitful discussions.
My thanks to prof. dr. ir. Jilt Sietsma for his advice and guidance with his knowledge of phase transformation and microstructure evolution. The use of SEM/EBSD was necessary for this research work. Special thanks to prof. dr. ir. Roumen Petrov from Universiteit Gent for his guidance with the interpretation of the EBSD results. I would also like to thank dr. ir. Rob Delhez for all the useful discussion on X-ray diffraction results.

I am thankful to the joining and mechanical behaviour group members for all the help that led to the completion of my research project. Our secretaries Anneke, Marian, Linda, Yneke and Fraukje helped me with all the administrative matters. I would also like to extend sincere thanks to Frans Bosman and Jurriaan van Slingerland for their help in conducting welding experiments, fixing power sources and helping in building experimental setup and Ton Riemslag for all the help in performing the mechanical tests. Without their help, this work would remain incomplete. I would also like to thank experienced technicians for helping in different experiments: Nico (dilatometer), Hans (gleeble), Erik (optical microscope), Kees (SEM) and Ruud (XRF and XRD). Thanks to all my colleagues in the group, He, Masoud, Chuangxin, Sepideh, and Vera, for all the nice discussions.

I enjoyed the experiments carried out at neutron (PSI) and synchrotron (ESRF) beamlines very much and for that I thank the team members over the years: Masoud, Marcel, He, Muru, Vera, Sepideh and Richard. Even though the experiments were very taxing, it was only due to your involvement that the experiments were successful. I would like to thank Jon Wright, Andrew King and Thomas Buslaps, our local contacts at the ESRF for their help during the experiments, sometimes even in the middle of the night.

I would also like to extend my thanks to Loïc Malet, Didier Robert and Prof. dr. Stéphane Godet of the 4MAT group in Université Libre de Bruxelles for their collaboration work which resulted in chapter 8. Special thanks to M. Herbig and P. -P. Choi of Max-Planck-Institut für Eisenforschung GmbH, for their collaboration work on high resolution EBSD on the nanocrystalline grains which resulted in chapter 9.

Finally, I would like to take the opportunity to express my gratitude to my family in India for their unfailing encouragement and support, specially my parents, Sandip and Rudrani Dutta, tha and my sister, Kaju. Also thanks to Rosanne for her understanding and patience and the indispensable support I received from her during my research career.
List of publications

Journals


- **R. K. Dutta**, R. M. Huizenga, R. H. Petrov, M. Amirthalingam, A. King, H. Gao, M. J. M. Hermans and I. M. Richardson, *In-situ synchrotron diffraction studies on transformation strain development in a high strength quenched and


**Conference proceedings**


Conference


CV

RANGAN KAUSHIK DUTTA

Born on 25\textsuperscript{th} August, 1983 in Kolkata, WB, India

2010 - 2014 PhD Researcher, Materials innovation institute,
Delft, The Netherlands.

2008 - 2010 Master of Science in Materials Science and Engineering,
Delft University of Technology, Delft, The Netherlands.

2006 - 2008 Software Engineer, Infosys,
Bangalore, India.

2002 - 2006 Bachelor of Engineering, Jadavpur University,
Kolkata, India.

1991 - 2002 M. P. Birla F. H. S. School,
Kolkata, India.