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Low cycle fatigue behaviour of a precipitation hardened Cu-Ni-Si alloy

Maxime Delbove^{a,b}, Jean-Bernard Vogt^{a,*}, Jérémie Bouquerel^a, Thierry Soreau^b, François Primaux^b

^a Unité Matériaux Et Transformations, Université de Lille - 1 Sciences et Technologies / UMR CNRS / ENSCL / INRA, 59655 Villeneuve d'Ascq, France

^b Le Bronze Industriel, Z.I. voie de Châlons, RD 977, 51600 Suippes, France

* Corresponding author. Tel.: +33 320 43 40 35; fax: +33 320 43 40 40

E-mail addresses: jean-bernard.vogt@ensc-lille.fr (J.-B. Vogt)

Abstract

Low cycle fatigue tests were performed at room temperature to investigate the role of the microstructure of a Cu-Ni-Si alloy on the stress response to strain cycling and on the fatigue resistance. The cyclic accommodation consisted in a hardening followed by a softening. TEM analysis showed that in some grains dislocations remained isolated and confined between precipitates while in other grains dislocations piled up at δ -Ni₂Si precipitates and then cut them. Repetitive cutting allows their dissolution and formation of precipitate-free bands where the plastic deformation is localised. The Manson-Coffin diagram exhibited two regimes according to the proportion of grains involved in the plastic deformation accommodation.

Keywords

copper alloys - cyclic properties - low cycle fatigue – microstructure - microscopy

Highlights

Investigated Cu-Ni-Si contains δ -Ni₂Si nano precipitates – Cyclic hardening followed by softening is observed – Dissolution of δ -Ni₂Si results from repetitive cutting – Deformation is localised in precipitate-free bands

1. Introduction

A large amount of engineering applications, like railway equipment, marine hardware or lead frames, requires materials that exhibit high mechanical strength as well as good electrical and thermal conductivities. In that way, copper alloys are widely used although the enhancement of mechanical properties usually comes along with a decrease of the electrical performances [1,2]. To face this problem, oxide-dispersion-strengthened (ODS) copper alloys, where aluminium oxide nanoparticles are dispersed in the matrix, is one of the existing solutions [3–5]. However, their mechanical properties might not be enough for the cited applications. To overcome this aspect, precipitation hardened copper alloys have been developed, such as copper-beryllium alloys. Those latter exhibits a good balance between mechanical, thermal

and electrical properties [1,6]. But due to the price and the toxicity of beryllium and its compounds, alternative copper alloys have been formulated. Cu-Ni-Si alloys, with a nickel content from 1.5 to 8.0 wt.% and a quantity of silicon included between 0.3 and 1.8 wt.%, are known to be one of the best replacement options [4–6]. Their good property balance is mostly attributed to the formation of nanosized disc-shaped coherent δ -Ni₂Si precipitates identified by transmission electron microscopy (TEM) [7–11]. Currently, the scientific literature on these alloys mainly deals with the microstructure evolution combined with the optimisation of the mechanical (hardness and tensile properties) and electrical behaviour thanks to their elaboration process [12–18], their heat treatments [7,12,19–21] or their chemical composition [14,22–28]. The scope of applications of Cu-Ni-Si alloys contains the transport sector, in electric engines for example. It implies that they are submitted to cyclic loading and are therefore subjected to fatigue failure. The fatigue behaviour of Cu-Ni-Si alloys has however only been investigated in few articles, especially on high-cycle fatigue (HCF) [29,30]. Low cycle fatigue (LCF) was just briefly studied on polycrystalline material by Lockyer and Noble [29], and on Cu-Ni-Si single crystals by Fujii et al. [31]. The LCF behaviour seems to exhibit cyclic softening but has not been fully investigated at this point. Very recently, Goto et al. [32] investigated the role of microstructure on short crack propagation in a Cu-Ni-Si alloy by pointing out the importance of precipitation process. Indeed, particles and precipitate-free zones formed in their material as a result of the high level of Ni and their heat treatment. Both induced localised high stress/strain distribution which led to the crack initiation, followed by the growth along slip planes in grains sharing grain boundaries.

The aim of the present work is therefore to improve the knowledge on a Cu-Ni-Si alloy by providing a complete description of the LCF behaviour. The study of cyclic accommodation and of fatigue resistance will be linked to the microstructure investigated by scanning and transmission electron microscopes in order to propose a fatigue mechanism of the Cu-Ni-Si alloy.

2. Material and experimental procedures

The Cu-Ni-Si alloy used in the present work is a CuNi₂Si (CW111C) provided by the company Le Bronze Industriel (France). The casted material was first solution treated around 950°C for 2 h, then hot formed, rapidly quenched and finally aged at a temperature between 450°C and 500°C for a duration between 2 h and 4 h. At last, the plate fatigue specimens were machined with a thickness of 3 mm, a width of 6 mm and a gauge length of 12 mm. This design of specimen ensures that no bending occurred during fatigue tests.

Before testing, each specimen was mechanically polished on both sides in order to remove the work-hardened surface due to the machining step. A mirror-like surface was obtained by using successively silicon carbide papers, diamond suspensions and colloidal silica suspension. For the metallographic investigation, samples were lastly etched during 10 s using a solution made of 5 g of FeCl₃, 50 mL of HCl and 100 mL of deionised water.

Fatigue tests were carried out on an electromechanical Schenck Trebel RMC10 fatigue machine using an Instron 8800 mini-controller. Low cycle fatigue tests were conducted at room temperature under axial total strain control by using a 10 mm gauge extensometer. A triangular waveform, a fully push-pull mode ($R_\epsilon = -1$), a total strain variation ($\Delta\epsilon_t$) value included between 0.6 % and 1.5 %, and a constant strain rate of $4 \cdot 10^{-3} \text{ s}^{-1}$ have been selected as testing parameters.

The fatigue life N_f has been defined as the number of cycles necessary to reach a difference higher than 25% between the maximum stress and the absolute value of the minimum stress during a cycle. The reference loop for the measurement of strain components and stress values was that recorded at 80% of the fatigue life.

The scanning electron microscopy (SEM) observations were conducted at an accelerating voltage of 20 kV on a FEI Quanta 400 tungsten-SEM fitted with an electron backscatter diffraction (EBSD) detector from Oxford Instrument. TEM observations were performed on a Philips CM30 at an operating voltage of 300 kV, while automated crystal orientation mapping (ACOM-TEM) were conducted at an accelerating voltage of 200 kV on a FEI Tecnai G2-20 equipped with the ASTARTM acquisition system developed by NanoMEGAS [33,34]. TEM thin foils were beforehand prepared by jet-polishing using Struers D2 commercial solution at 15 V and 15°C. The orientation data was post-treated using the TSL OIMTM Analysis 7 commercial software provided by EDAX.

3. Results and analysis

3.1 Metallographic characterisation and microstructure

The microstructure of the material is shown in Fig. 1. EBSD data reveal that the studied alloy presents equiaxed grains with a size ranging from 20 to 50 μm . A large number of annealing twins is also observed.

Fig. 1. EBSD unique grain map of the studied Cu-Ni-Si alloy

Concerning the precipitates formed during the aging treatment, they have been identified by using selected area diffraction mode in TEM. Disc-like δ -Ni₂Si intended for the hardening of the alloy have been indeed detected. The precipitate density is high and their width is close to 5 nm. For example, Fig. 2 shows the dark field micrograph and the diffraction pattern for a beam direction parallel to $\langle 111 \rangle_{\text{Cu}}$. The bright spots correspond to the diffraction of the copper matrix whereas the dotted star pattern is due to the diffraction of the three variants of δ -Ni₂Si. Moreover, the nature of the precipitates is in agreement with the thermodynamic calculation of Lu et al [35].

Fig. 2. Dark field micrograph (a) and corresponding selected zone on the indexed diffraction pattern (b) of the Cu-Ni-Si alloy for a beam direction parallel to $\langle 111 \rangle_{\text{Cu}}$

3.2 Monotonic tensile behaviour

Tensile tests have been carried out to characterise the monotonic properties of the material. Tests were performed up to fracture on a servo-hydraulic machine under displacement mode control in order to have a calculated strain rate equal to 10^{-3} s^{-1} . An additional test where an extensometer was stuck to the gauge length was performed to measure accurately the Young modulus. Monotonic tensile properties values are summarised in Table 1.

Table 1. Monotonic tensile characteristics of the Cu-Ni-Si alloy

E (GPa)	YS _{0.2 %} (MPa)	UTS (MPa)	A _u (%)	A (%)	n	K (MPa)
130	540	700	11.5	12.7	0.12	930

The material presents a good balance between a high ultimate tensile strength (UTS) and a uniform elongation (A_u) higher than 10 %. But the low difference between A and A_u reveals a low toughness of the material. This is confirmed by the observations of the fracture surfaces. Fig. 3 clearly shows that the tensile fracture surface comprises a mixture of brittle and ductile zones. The surface presents at the same time respectively intergranular fracture and ductile dimples.

Fig. 3. SEM micrographs of the monotonic tensile fracture surface of the Cu-Ni-Si alloy presenting intergranular rupture (a) and dimples (b)

3.3 Low cycle fatigue behaviour

3.3.1 Cyclic accommodation

The graphical representations of the stress amplitude σ_a as a function of the number of cycles N in a logarithmic scale and versus the fatigue life fraction in linear scale are plotted in Fig. 4 for the different total strain variations. The behaviour during tensile and compressive phases was nearly symmetrical.

For the high total strain variation tests ($\Delta\epsilon_t = 1.2\%$ and 1.5%), the fatigue life of the alloy can be divided into four parts. At the beginning of the test, a hardening behaviour is observed during the first 50 cycles. This step is followed by a primary softening behaviour, until 20 % of the fatigue life fraction. Then, a secondary softening step with a lower intensity occurs until 90 % of the fatigue life fraction. At the end, the last 10 % of the fatigue life fraction are attributed to the propagation of the main crack in the bulk. This LCF behaviour confirms the cyclic accommodation observed by Lockyer and Noble but not their fatigue life [29]. Their alloy, with a similar chemical composition but a different thermomechanical treatment (the as received commercial alloy was re-solution treated at $800\text{ }^{\circ}\text{C}$ for 2 h and then aged at $450\text{ }^{\circ}\text{C}$ for 2 h), exhibits indeed a number of cycle to failure way lower than the one in the present study (for a test carried out at 1 % total strain amplitude on hour-glass specimens). This difference can be attributed to the microstructure variation (grain size, precipitate size and density) resulting from the thermomechanical treatment. This result highlights the

significance of the material process and initial microstructure descriptions for Cu-Ni-Si alloys.

At lower total strain variation ($\Delta\epsilon_t = 0.8\%$), no hardening stage is observed at the beginning of the test and the alloy continuously softens during the cyclic loading at a rate close to the second softening step at high total strain variation. Moreover, at the lowest tested total strain variation ($\Delta\epsilon_t = 0.6\%$), the softening effect is very slight. Therefore, for the considered conditions, the material exhibits no real stable state during the cyclic loading.

Fig. 4. Evolution of the cyclic stress amplitude versus the number of cycles (a) and the fatigue life fraction (b) for different total strain variations

To appreciate more accurately the hardening and softening, the evolution of plastic strain variation $\Delta\epsilon_p$ during cyclic loading has been plotted (Fig. 5).

Fig. 5. Evolution of the cyclic plastic strain variation versus the number of cycles for different total strain variations

For a total strain variation $\Delta\epsilon_t$ included between 0.9 and 1.5%, the plastic strain decreases at first and then increases, which correspond respectively to the hardening and softening steps. This clearly indicates that the hardening and softening are very strong. However, hysteresis loops exhibited a linear shape during the entire fatigue life for the test conducted at $\Delta\epsilon_t = 0.6\%$ and no macroplasticity has been observed. The absence of stabilised state needs to specify a criterion for the selection of the hysteresis loops for the drawing of the cyclic stress-strain curves presented in Fig. 6. In the present case, it has been decided to measure strain and stress values on hysteresis loops recorded at the hardening peak and at the end of the softening period, just before the crack propagation period, which corresponds to 90% of the fatigue life. It can be clearly seen that the softening effect during cyclic loading is much more important than the slight hardening resulting from the beginning of fatigue testing.

Fig. 6. Monotonic and cyclic stress-strain curves for the Cu-Ni-Si alloy tested at room temperature

3.3.2 Fatigue resistance

The graphical representation of the total, the plastic and the elastic strain variations (respectively $\Delta\epsilon_t$, $\Delta\epsilon_p$ and $\Delta\epsilon_e$) with the fatigue life is presented in Fig. 7a. The Manson-Coffin relation (Eq. 1) must be considered according two regimes ($\Delta\epsilon_t \leq 1\%$ or $\Delta\epsilon_t > 1\%$) while only one regime can be considered for the Basquin relation (Eq. 2).

$$\Delta\epsilon_p = K_p(N_f)^{C_p} \quad (\text{Eq. 1})$$

$$\Delta\epsilon_e = K_e(N_f)^{C_e} \quad (\text{Eq. 2})$$

The evolution of the stress amplitude σ_a versus the number of cycles to failure N_f is also plotted in Fig. 7b. It follows the pseudo-Wöhler relation (Eq. 3).

$$\sigma_a = K_{PW}(N_f)^{C_{PW}} \quad (\text{Eq. 3})$$

Finally, the plastic energy dissipated per cycle ΔW_p , measured by the area inside the hysteresis loop, is represented against N_f in Fig. 7c. As for the Manson-Coffin approach, two regimes must be considered ($\Delta\epsilon_t \leq 1\%$ or $\Delta\epsilon_t > 1\%$). The Golos and Ellyn relation (Eq. 4) has been employed to measure the fatigue resistance of the alloy.

$$\Delta W_p = K_{Wp}(N_f)^{C_{Wp}} \propto \sigma_a \cdot \Delta\epsilon_p \quad (\text{Eq. 4})$$

The Manson-Coffin relation and the plastic energy dissipated per cycle approach appear to be the two best criteria to quantify the fatigue resistance of the tested Cu-Ni-Si alloy, but two distinct regimes ($\Delta\epsilon_t \leq 1\%$ or $\Delta\epsilon_t > 1\%$) have to be considered.

Fig. 7. Fatigue resistance diagrams of the Cu-Ni-Si alloy: (a) Manson-Coffin diagram, (b) Pseudo-Wöhler diagram, (c) energy diagram

In the four cited equations, the factors K and C represent respectively the coefficient and the exponent of fatigue resistance associated to the relation. All these results are summarised in Table 2.

Table 2. Fatigue resistance coefficients and exponents of the Cu-Ni-Si alloy

	Manson-Coffin		Basquin		Pseudo-Wöhler		Golos and Ellyn	
$\Delta\epsilon_t$ (%)	K_p (%)	C_p	K_e (%)	C_e	K_{PW} (MPa)	C_{PW}	K_{Wp} (GJ.m ⁻³)	C_{Wp}
≤ 1	41700	-1.44	1.30	-0.070	958	-0.096	545	-1.54
≥ 1.2	23.1	-0.474					0.569	-0.646

3.3.3 Crack initiation

Slip marks produced by cyclic loading at the external surface of the fractured specimens at $\Delta\epsilon_t = 0.8\%$ and $\Delta\epsilon_t = 1.5\%$ are displayed Fig. 8. The external surfaces are covered by extrusions the density of which was very high at high strain range. Compared with pure polycrystalline copper [36], they appeared much thinner and much straighter and had a similar aspect to those produced in other precipitate-strengthened alloys [37,38]. This suggests that

cross slip is disfavoured for planar slip. As well, secondary slip was frequently observed in the highly strained specimens.

Fig. 8. SE-SEM micrographs of specimen surfaces after failure at $\Delta\epsilon_t = 0.8 \%$ (a) and at $\Delta\epsilon_t = 1.5 \%$ (b). The double white arrow indicates the loading axis.

The evolution of the slip marks pattern has been analysed thanks to interrupted tests at different fatigue life fractions for the test performed at $\Delta\epsilon_t = 1.5 \%$. The corresponding SEM observations are gathered together in Fig. 9. During the hardening period, the slip activity developed in one slip system in some grains while the other ones appeared less active or may be inactive. Fig. 9a shows that the slip marks formed within the activated grains after 25 cycles consist of straight slip bands the emergence of which is mostly weak. Among them, a few ones are more developed. After 50 cycles (Fig. 9b), that is at the transition between the hardening and the softening periods, the slip marks pattern is nearly the same but more pronounced and secondary slip started occurring. With further cycling, the slip band density and the slip band height increase then with the number of cycles until the specimen fracture. Crack initiation took place at slip band roots and propagation was mainly transgranular and occasionally intergranular. The crack mode is similar to the one observed in α -brass [39,40].

Fig. 9. SE-SEM micrograph evolution of the specimen surface as a function of the number of cycles for a test conducted at $\Delta\epsilon_t = 1.5\%$. The double white arrow indicates the loading axis.

3.3.4 TEM investigation

TEM thin foils were taken off from post-mortem specimens to study the dislocation structure after fatigue testing. Contrary to fatigued polycrystalline copper [41] or ODS copper alloys [42], the cyclic deformation did not result in well-formed dislocations arrangements such as walls or cells in the investigated strain ranges. Globally, the grains were featuring two kinds of deformation structures. The first one is shown in Fig. 10, where some grains contained a high density of dislocations segments or debris next to the δ -Ni₂Si precipitates but dislocations remained isolated and did not rearrange into a low energy dislocation structure. In addition, some dislocations appear to be pinned between two precipitates. The alloy fatigued at $\Delta\epsilon_t = 0.6\%$ had all the grains with this structure observed in Fig. 10.

Fig. 10. TEM observations of fatigued Cu-Ni-Si specimens after failure at $\Delta\epsilon_t = 0.6\%$ (a) and at $\Delta\epsilon_t = 1.5\%$ (b)

For the other specimens tested at strain ranges higher than $\Delta\epsilon_t = 0.6\%$, in addition to the former observations, the other grains were decorated with a grid pattern of white bands as shown in Fig. 11. The more the specimen is strained, the higher is the amount of grains presenting this pattern. These bands crossed the entire grain from grain boundary to opposite one and propagated from neighbouring grains or twins. Close observations showed that they were free of precipitates.

Fig. 11. TEM micrographs of a fatigued Cu-Ni-Si specimen (after failure at $\Delta\epsilon_t = 1.5\%$) showing bands without precipitates

The nature of the white precipitate-free bands has been determined using the ACOM-TEM technique. A zone $4\ \mu\text{m}^2$ in area including precipitate-free bands was scanned at a 5 nm step with the ASTARTM software. Fig. 12a is virtual bright field image and Fig. 12b is the inverse pole figure for copper crystals. No misorientation is pointed out between the band and the matrix, or is below the angular resolution of the system, evaluated at 1° with the selected experimental conditions [34]. It is therefore clearly concluded that these precipitate-free bands are not deformation twins but intensive slip bands.

Fig. 12. ACOM-TEM virtual bright-field image (a) and corresponding inverse pole figure map (b) of a zone scanned with a 5 nm step containing precipitate-free bands

4. Discussion

The present investigation which aimed at identifying the low cycle fatigue properties of a Cu-Ni-Si alloy shows that the material exhibited high stress values. This is the result of the efficient hardening heat treatment that precipitated δ -Ni₂Si nanosized particles. Compared to other ODS copper such as Cu-Al₂O₃ material, the Young modulus as well the yield stress of the considered Cu-Ni-Si alloy are respectively 30 % and 65 % higher [3]. Both materials exhibit nearly the same size range of nanoparticles but the interaction of dislocations with the precipitates is different. In Al₂O₃ ODS copper alloys, dislocations can bypass the Al₂O₃ particles but do not interact similarly with the δ -Ni₂Si precipitates of the tested Cu-Ni-Si. Due to their coherency with the copper matrix [7,43] and their high volume fraction, the interaction of dislocations with δ -Ni₂Si nanosized particles is cutting [44]. This impacts the cyclic stress response to strain cycling and the fatigue life dependence with the applied strain of the Cu-Ni-Si alloy.

For the lowest strain range test ($\Delta\epsilon_t = 0.6$ %) where the stress amplitude is nearly constant during cycling, all the grains exhibited the same deformation structure. Dislocations moved under the applied shear stress and remained confined in their slip plane, so that no substructure formed. The plastic deformation is so small that dislocations accommodated it just by a quasi-reversible motion and did not interact between them. As well, the dislocation displacement is less than the average distance between precipitates so that they nearly did not interact with them.

For the other tests, the mechanical response, the cyclic stress-strain curve and the Manson-Coffin curve, points out two regimes whether tests were performed in the range 1.2% - 1.5% or in the range 0.8% - 1.0%. If we consider the strain variations ranging from 1.2% to 1.5%, that is a situation totally opposite to the one described above, the assigned deformation is so high that most of all the grains are activated and accommodate large plastic strain. The number of involved dislocations is high and should glide on large distance but have a limited mean free path because of the high density of nanoscale δ -Ni₂Si precipitates [44]. A strong

hardening is thus observed as the result of dislocations-dislocations and dislocations-precipitates interactions. At the same time, as the dislocations pile up at precipitates, an increasing shear stress is generated. When the critical value for shearing is reached, here after 50 first cycles (the cyclic peak stress), then precipitates are cut providing a path for dislocations. The blocked dislocations can therefore escape leading to stress relaxation and to the observed strong primary cyclic softening in Fig. 4. A strong localisation of plastic deformation occurs in narrow bands which behave as persistent slip bands in pure copper. Such bands have already been reported under LCF in a Cu-Ni-Si single crystal [31], under LCF and high-cycle fatigue in a polycrystalline Cu-Ni-Si alloy [29], but also in other precipitate hardened copper (Cu-Co) [45,46], aluminium [37,47,48] or nickel alloys [38,49]. The moderate secondary softening corresponds to dislocation behaviour in these bands and it turns out that annihilation process is still prevailing and that equilibrium is not reached. Therefore, due to successive cutting of δ -Ni₂Si by the dislocations going forward and backward in the softening stage, precipitates are reduced under the critical size of stability, leading to their dissolution into the matrix and finally to the formation of precipitate-free bands.

When the applied strain is moderate, typically $\Delta\epsilon_t = 0.8\%$ or 0.9% , most grains accommodate the plastic deformation. Since the assigned deformation is quite small, dislocations glide on rather short distance and are not obstructed as much as in the range of deformation $\Delta\epsilon_t = 1.2\% - 1.5\%$, which gives rise to a very moderate or even to a quasi-absence of hardening. Only few well oriented grains have dislocations moving on larger distance with a limited mean free path as described previously. These grains cyclically harden but since they are in minority, the macroscopic average cyclic response is a cyclic softening. The transition between the two regimes in the cyclic accommodation curves diagram and in the Manson-Coffin diagram low and high strain regimes reflects the proportion of grains involved in the hardening/softening process is majority.

5. Conclusion

The investigation of the microstructure of the tested Cu-Ni-Si alloy has exhibited a high density of δ -Ni₂Si precipitates resulting in the fatigue properties summarised as follows.

1. The cyclic accommodation of the alloy is composed of a hardening step at the beginning of the test, follow by a continuous softening until the specimen fracture. Moreover, two plastic regimes have to be considered ($0.8\% \leq \Delta\epsilon_t \leq 1.0\%$ and $1.2\% \leq \Delta\epsilon_t \leq 1.5\%$) in order to characterise its fatigue resistance. Planar slip is favoured in this alloy and the slip band density at the surface increase with the strain level and the number of cycles. Finally, grains exhibit two different strain patterns. The first one presents lots of short dislocations but not rearranged into a sub structure. The other one is composed of precipitate-free bands and only appeared if $\Delta\epsilon_t$ is higher than 0.8% . Moreover, the number of grains exhibiting the second strain patter increased with the strain level.

2. The microstructure evolution during cyclic loading can be described as follow. At the lowest tested strain range ($\Delta\epsilon_t = 0.6\%$), each grain can accommodate the deformation by quasi reversible slip of dislocations. At higher strain ranges ($\Delta\epsilon_t \geq 0.8\%$), some grains present dislocations piled up on precipitates, leading to an alloy hardening. Then precipitate cutting can occur resulting in a cyclic softening of the alloy. The successive cutting of the precipitates

leads to their dissolution into the matrix and then precipitate-free bands where the deformation is localised. The two regimes exhibited by the Manson-Coffin diagram reflect the proportion of grains involved in the plastic deformation accommodation. The above described presented phenomenon are indeed localised in a few number of grains at low strain rates ($0.8 \% \leq \Delta \epsilon_t \leq 1.0 \%$), whereas they are generalised at higher strain ranges ($1.2 \% \leq \Delta \epsilon_t \leq 1.5 \%$).

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Highlights

Investigated Cu-Ni-Si contains a high density of δ -Ni₂Si precipitates – Cyclic hardening followed by softening is observed – Dissolution of δ -Ni₂Si results from repetitive cutting-localisation of deformation in precipitate-free bands