Investigation of the electroplastic effect using nanoindentation

D. Andrea, T. Burleta, F. Körkemeyer, G. Gerstein, J.S.K.-L. Gibson, S. Sandlöbes-Haut, S. Korte-Kerzel

Institute of Physical Metallurgy and Materials Physics, RWTH Aachen University, Germany
Institute of Materials Science, Leibniz University Hannover, Germany

**Highlights**

- A novel set-up for electro-plastic nano-indentation was developed and applied to Al, Al$_2$Cu, and eutectic Al + Al$_2$Cu.
- The study shows for the first time that electro-plasticity is relevant not only to metals, but also to intermetallics.
- Current intensity, pulsing interval and loading rate determine the magnitude of load drops due to electric current pulses.
- The main contribution to electro-plasticity is de-pinning of dislocations from obstacles, not thermal effects.

**Abstract**

A promising approach to deform metallic-intermetallic composite materials is the application of electric current pulses during the deformation process to achieve a lower yield strength and enhanced elongation to fracture. This is known as the electroplastic effect. In this work, a novel setup to study the electroplastic effect during nanoindentation on individual phases and well-defined interfaces was developed. Using a eutectic Al-Al$_2$Cu alloy as a model material, electroplastic nanoindentation results were directly compared with macroscopic electroplastic compression tests. The results of the microscopic and macroscopic investigations reveal current induced displacement shifts and stress drops, respectively, with the first displacement shift/stress drop being higher than the subsequent ones. A higher current intensity, higher loading rate and larger pulsing interval all cause increased displacement shifts. This observation, in conjunction with the fact that the first displacement shift is highest, strongly indicates that de-pinning of dislocations from obstacles dominates the mechanical response, rather than solely thermal effects.

**Keywords**: Electroplasticity, Nanoindentation, Metallic-intermetallic composites, Al-Cu alloys

1. Introduction

The influence of electric current on the plastic deformation of metals, termed as the electroplastic effect (EPE), was first investigated by Troitskii and Likhtman [1]. The EPE was subsequently found to influence a wide range of mechanical properties, such as flow stress [2,3], stress relaxation [4], creep [4], dislocation generation and mobility [5], brittle fracture [6], and fatigue [7]. Systematic studies were carried out to study the nature of the EPE over a variety of loading conditions and materials and the EPE was also found to affect the materials’ behaviour during processing [8,9]. Though several mechanisms for the EPE were proposed, our understanding of its nature remains at a nascent stage. Local adiabatic temperature increase due to fast joule heating was thought to be

https://doi.org/10.1016/j.matdes.2019.108153
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the major contributor to the EPE due to local thermal softening [10]. However, the EPE was also observed at low temperatures and high-density current pulses [1], during which the joule heating is negligible, thus indicating that a part of the EPE is intrinsically related to the interaction between drift electrons and dislocations. Early work by Troitskii et al. [1,11] and later by Conrad et al. [5,12] considered the effect of the ‘electron wind’, i.e. the drift of conduction electrons upon application of an electric potential, in assisting dislocations to overcome lattice obstacles. The ‘electron wind’ theory has since been widely applied and is able to predict key features of the EPE in a variety of materials and loading conditions [12]. More recently, Molotskii [13] also proposed the depinning of dislocations from paramagnetic obstacles by the magnetic field induced by the electric current as an alternative mechanism for the EPE in metals. Beyond the EPE, electric currents can also induce phase transformations through increased nucleation and recrystallisation rates, often resulting in grain refinement and enhanced mechanical properties [14–16]. Furthermore, the effects of an electric current applied separately to any deformation or heating process has been shown to differ from those where an electric current is applied in combination with deformation [17].

Several key factors in the enhanced dislocation motion were identified, such as the effects of drift electrons on the driving force for dislocation motion, the activation energy barrier of dislocation obstacles, and the depinning of dislocations from paramagnetic obstacles [13]. However, until now, no method has been available which allowed a quantitative investigation of the EPE in a well-defined local environment, such as an individual grain or eutectic region, selected crystal orientations or isolated grain and phase boundaries.

Electroplastic forming, in which the EPE is exploited by passing a current through the work piece during processing, is particularly promising for materials that are otherwise brittle. An example are metallic-intermetallic composite materials that are — at least theoretically — capable of combining the high formability of metals and high strength of intermetallic phases. Plasticity in such composite alloys is predominately carried by the formable metallic components while the brittle intermetallic phases strengthen the material. Microstructurally weak points are the interfaces between metallic and intermetallic phases due to stress concentrations promoting strain localization and failure initiation [18–23]. In general, the constituent phases and the morphology and spacing of the eutectic structures control the mechanical behaviour of eutectic composite alloys [24,25]. Structural refinement is reported to cause an increase in strength concomitant with a loss in ductility [24,25].

As an example of metallic-intermetallic composite materials, Al–Cu eutectic alloys exhibit low density, good castability, corrosion resistance and a wide range of available phases and microstructural morphologies. The main microstructural constituents of eutectic Al–Cu composite alloys are cubic (fcc) Al and tetragonal (bct) Al2Cu [26]. Depending on the composition, solidification and possible heat treatment conditions different volume fractions and morphologies of fcc Al and bct Al2Cu form. Additionally, some more copper-rich intermetallic phases might form as precipitates [25,27–29].

The main drawback of Al–Cu eutectic composite alloys is brittle deformation at ambient temperatures [18,23,30,31]. Early detailed characterisation of deformation microstructures of Al–Cu eutectic alloys revealed dislocation-dominated deformation in Al grains with higher dislocation densities at Al-Al2Cu interfaces while the eutectic lamellae deformed by kinking to accommodate compressive strain. Consequently, fracture was observed along Al2Cu interfaces or through Al2Cu grains [23,31]. The Al–Cu composite is therefore a candidate material to benefit from electroplastic forming if the process enables plastic flow in the brittle phase and its interfaces, such that formability is enhanced and damage during forming reduced.

Here, we present the first local studies of the EPE at the scale of an alloy's individual microstructural components. For this, we developed a novel in-situ nanoindentation setup in which high current densities are passed through the contact during deformation. This setup therefore opens new opportunities to investigate the EPE and the underlying mechanisms. In order to allow a direct comparison across the scales, we study and compare the effects of electric pulses during deformation in a eutectic Al–Cu alloy on both, the macroscopic and microscopic scale.

2. Experimental procedure

2.1. Macroscopic electroplasticity experiments

Macroscopic electrically pulsed compression experiments were performed on bulk eutectic Al–Al2Cu samples with a composition of 33.1 wt% copper and 66.9 wt% aluminium. The as-cast material was cut into specimens with dimensions of 2 mm × 2 mm × 4 mm (width × length × height) using electric discharge machining. Compression experiments with and without electric current pulsing during deformation were performed at a deformation rate of 0.1 mm/min using a Walter & Bai 100 universal testing machine at the Institute of Materials Science, Leibniz University Hannover. After the application of a pre-force of 80 N and an offset time to ensure proper electrical contact, electric current pulses of 7 kA, 55 V and a duration of 0.5 ms were applied every second using a high current impulse generator (further information regarding the high current impulse generator are available in [32]). The resulting

Fig. 1. Al-Al2Cu eutectic alloy used for nanoindentation experiments including nine indents performed without application of electric current pulses.

Fig. 2. True compressive stress-strain curves of samples deformed with and without the application of electric current pulses. The electric current pulses had a current of 7 kA, voltage of 55 V and duration of 0.5 ms at a frequency of 1 Hz. The initial strain rate was 0.1 mm/min.
current densities amounted to $1.66 \pm 0.01 \text{kA/mm}^2$ when considering the contact area prior to deformation. For comparison, compression experiments on the same material with the same sample geometry and machine settings were performed without electrical pulses. During the compression tests, the sample temperature was recorded using a FLIR Systems, ThermaCam SC3000 at a frequency of 250 Hz.

2.2. Microscopic electroplasticity experiments

The same eutectic Al-$\text{Al}_2\text{Cu}$ alloy with a composition of 33.1 wt% copper and 66.9 wt% aluminium as used for the macroscopic tests, was investigated using electroplastic nanoindentation experiments (Fig. 1). Furthermore, electroplastic nanoindentation was also performed on as-cast samples of the $\text{Al}_2\text{Cu}$-phase and the $\text{Al}$-phase.

The samples were metallographically prepared and deformed using a nanoindenter (NanoTest, Micromaterials Ltd., UK) modified as part of this work (see Section 3.2.1 ‘Technical development’) to allow application of electric current pulses of 100 ms in length. Using this new nanoindentation setup, four different current intensities in the range of $1.5 \times 10^3 \text{A}$ and two different pulsing intervals of 1 s and 2 s using a loading and unloading rate of 20 mN/s to a final load of 500 mN were applied. Experiments on the individual phases were conducted using a loading rate of 20 mN/s ($\text{Al}_2\text{Cu}$-phase) and 2 mN/s ($\text{Al}$-phase), respectively, with a holding period of 5 s at maximum load before unloading. Additionally, experiments with a current intensity of 2.5 A during loading and unloading with loading and unloading rates of 10 mN/s and 20 mN/s were carried out to identify the effects of electric current pulses during unloading. In order to investigate the interplay of indentation depth, current and resulting current density, additional experiments were carried out with a constant displacement rate of 20 nm/s until maximum displacements of 2000 nm and 1200 nm. To evaluate possible area functions (projected contact area as used in the Oliver and Pharr analysis [33], true area of contact between material and indenter or circumference of the plastic zone), increasing intensities of the electric current pulses scaling with each potential area function at each depth/time increment were used (see Supplementary Materials).

3. Results

3.1. Macroscopic electroplasticity experiments

3.1.1. Macroscopic stress-strain response

Fig. 2 shows the true stress-strain curves of macroscopic compression experiments with and without electric pulses. In the beginning of the compression experiments, before electric current pulses were applied, all samples show similar stress strain responses. When applying the first electric current pulse, large stress drops of about $47.8 \pm 4.8 \text{MPa}$ occurred (highlighted by the red box in Fig. 2). The subsequently applied current pulses caused smaller stress drops of about $18.3 \pm 1.6 \text{MPa}$ during compression. The stress-strain curves of the samples with and without electric current pulses have similar slopes. At a strain of 5% - 6% the electric current pulses were turned off to avoid electric arcing during fracture. With the shut-off of the electric current pulsing, the stress of the samples maintains a lower stress level than that of samples compressed without electric current pulses at the same strain value and fracture shortly after the final electric current pulses, see blue box in Fig. 2.

Generally, samples exposed to electric current pulses during compression reached higher fracture strains and exhibited lower flow stresses than samples compressed without electrical current pulses.

Shear band induced fracture was observed for all compression samples. The corresponding fracture planes were inclined by approximately 45° towards the loading direction. Since the electrical current pulses were turned off before fracture occurred, it...
cannot be determined if fracture was initiated before or after the electric current pulses were switched off.

3.1.2. Temperature evolution during macroscopic electroplasticity experiments

Fig. 3 shows the temperature evolution of the third sample given in Fig. 2 as a typical example for all electroplastically (EP) deformed samples. The temperature, which was measured at the sample surface in the centre of the gauge section, rises abruptly and then drops back to nearly room temperature after each electric current pulse.

The maximum temperature measured during the EP compression tests was 76.0 °C (at a minimum temperature of about 29.5 °C). The increase in minimum temperature measured during and after the EP compression experiments is assumed to be due to residual heat present in the sample.

3.2. Micromechanical electroplasticity experiments

3.2.1. Technical development

The development of an experimental setup enabling nanoindentation experiments with electric current pulses offers a fundamentally new approach to investigate the electroplastic effect. It allows us to evaluate the effect of electric current pulses on the mechanical properties and deformation mechanisms of defined microstructural constituents. Furthermore, due to the small dimensions it is possible to apply high current densities.

Fig. 4 a) shows a schematic drawing of the commercial nanoindenter adapted for this setup (NanoTest P3, MicroMaterials Ltd.). The system is controlled by passing a current through the coil, which is then attracted towards the permanent magnet, resulting in a rotation of the pendulum around the frictionless pivot. This movement leads to the indenter tip being pushed towards/into the sample and can be tracked via the displacement sensor at the lower end of the pendulum. The new setup also contains a sample holder designed and built in-house, a dedicated tip holder developed for this application and a controllable power supply.

The newly designed sample holder ensures electric insulation of the sample towards the indenter due to the use of insulating ceramics and plastics (Fig. 4c)). The added stub for scanning electron microscopy (SEM) facilitates easy sample transfer to the microscope after indentation. The electrical parts of the tip holder are insulated towards the indenter by using an alumina rod (Fig. 4 b)). A program was developed to start the current reproducibly at a desired time after the start of the indentation by automatically detecting the contact between tip and sample. Additionally, the achieved current values can be recorded and correlated with the mechanical data.

Similar to the macroscopic EP experiments, the electric current was applied in short pulses with a length of 100 μs each to minimize the effect of joule heating. It should be noted that the pulse duration applied in the micromechanical EP experiments is five times shorter than that applied during macroscopic electropolastic compression experiments to minimize joule heating at the increased current density. The current intensity was varied between 1.0 A and 3.0 A and applied at a defined frequency during loading and unloading (Fig. 5). The indenter tip material was tungsten carbide, as it combines high electrical conductivity with high hardness.

3.2.2. Nanoindentation experiments with different current intensities and pulsing intervals

The nanoindentation load-displacement curves obtained when applying different current intensities or pulsing intervals on a eutectic Al-Al2Cu sample are shown in Fig. 6. The application of electric current pulses during indentation causes a step-like increase in displacement with constant loading rate (see also Fig. 5 above). A similarly step-like displacement curve is observed during unloading. During loading, these steps in the load-displacement curve are characterised by a rapid displacement shift towards higher displacements at constant load upon each electric current pulse, followed by a period of low displacement response to the applied load in the time between two consecutive electric current pulses. During unloading, the direction of the displacement shift is reversed and becomes more noticeable the smaller the remaining applied load (see Section 3.2.3. Pulsed unloading experiments).
Variation of the current intensity reveals that the load-displacement curves corresponding to higher currents show "broader" steps, i.e. higher displacement shifts, than those corresponding to smaller currents. As similarly observed during macroscopic EP compression experiments, the displacement shift induced by the first electric current pulse is larger than the displacement shifts induced by subsequent electric current pulses, independent of the applied current (blue box, Fig. 6a and b)).

Fig. 6 b) shows the effect of the interval between two electric current pulses on the load-displacement behaviour of the eutectic Al-Al2Cu alloy. A larger pulsing interval results in larger displacement shifts but affects the maximum displacement values only marginally.

Indentation load-displacement data of the individual phases Al2Cu-Θ and fcc-aluminium are shown in Fig. 7 using also different current intensities and replicating the described effect that a greater current intensity yields larger displacement shifts. In addition to the displacement shifts associated with electric pulses, the load-displacement data exhibits serrations. As these are present also in the indentations without electric current pulses, they appear to be associated with the underlying plasticity mechanisms of the intermetallic Al2Cu phase. In other intermetallics, a similar behaviour was observed and is often associated with localised dislocation slip on distinct slip planes [34,35].

Due to the difference in hardness these experiments were conducted to different maximum loads to achieve comparable displacements. Similarly, the loading rates were scaled, using 20 mN/s on the Al2Cu-Θ phase and 2 mN/s on the fcc-aluminium phase. Quantitatively, nanoindentation experiments on (110)\textlangle ND\rangle Al2Cu-Θ phase grains reveal hardness values of 5.3 ± 0.09 GPa at a depth of 1100 nm for four indents performed without the application of electric currents and hardness values of 5.07 ± 0.53 GPa at a depth of 1100 nm for four indents performed with electric current pulses of 1.5 A (a)). Note that for each value given here with a standard deviation, the individual data points are shown in Fig. 8. As the number of indentations was limited to ensure tip wear would not be significant, the standard deviations are calculated only to convey a measure of repeatability or scatter and are not statistically relevant calculations with four indentations averaged on the Al2Cu-Θ phase and two on the Al α-phase. The elastic modulus fitted for the upper 80% of the unloading curve ranged from 101.3 ± 3.6 GPa for indents without the...
application of electric current pulses to 118.5 ± 2.9 GPa for indents with 1.5 A current pulses during application (Fig. 8 b).

The hardness values for the Al α-phase (oriented [352]||ND) accounted for 0.34 ± 0.00 GPa for two indents performed without current pulses and 0.33 ± 0.01 GPa for two indents performed with 1.5 A current pulses during deformation (Fig. 8c). The elastic modulus amounted to 72.6 ± 1.4 GPa without application of current pulses and 70.4 ± 1.3 GPa for indents performed with 1.5 A (Fig. 8d)).

Fig. 9 shows the hardness drops induced by electric current pulses of both individual phases. The first hardness drop is marked in red, as it is always larger than the subsequent ones. The hardness drops furthermore increase in size with increasing current intensity. For low current intensities (0.5 A), it is not possible to distinguish between the current induced displacement shifts and serrations that occur independent of electric current pulses, and consequently, no hardness drops were calculated for 0.5 A.

3.2.3. Pulsed unloading experiments

The application of electric current pulses of 2.5 A during unloading causes a similar step-like displacement response as observed during loading. Fig. 10 shows load-displacement curves with loading and unloading rates of 10 mN/s and 20 mN/s, respectively. The displacement shifts occurring during loading and unloading at a loading (and unloading) rate of 10 mN/s are smaller and half in height compared to the displacement shifts occurring at a loading (and unloading) rate of 20 mN/s. The direction of the displacement shifts changes during unloading, specifically, the first displacement shift during unloading continues to higher displacement (blue box, Fig. 10), but changes towards lower displacement during progressive unloading (red box, Fig. 10). Furthermore, the displacement shifts during unloading are smaller than those during loading and increase as the applied load decreases. The maximum displacement seems to be unaffected by the loading rate.

4. Discussion

4.1. Macroscopic EP experiments

Our experimental results reveal that electric current pulses can reduce the stresses needed for plastic deformation and enhance plastic deformation of a eutectic Al-Al2Cu alloy under compressive load. The stress-strain curves of the EP deformed specimens show rapid stress drops upon each electric current pulse. These stress drops are followed by a sudden increase in stress until the next electric current pulse was applied. A similar phenomenon has been reported by several other researchers before [1,2,36,37] and confirms the applicability of EP assisted forming for Al–Cu eutectic alloys.

The EP stress-strain curves further reveal that the stress drop induced by the first electric current pulse amounts to more than two times the stress drops induced by the following electric current pulses. A similar effect has been reported by Livesay et al. [38], who investigated the effects of electrical current pulses on the plastic deformation of Al thin films. Their results [38] reveal that the increase in elongation induced by the first current pulse is higher than those induced by subsequent electric current pulses.

The high stress drop induced by the first electric current pulse observed in the current study might be caused by the electron wind effect [1,5,11,12], which is assumed to help dislocations to overcome obstacles. We therefore assume that during the first current pulse dislocations present in the microstructure are de-pinned from short-range obstacles, hence, inducing a major stress drop. During the following electric current pulses, the number density of pinned dislocations is consequently lower, resulting in smaller stress drops during subsequent electric pulsing. The slope of the EP compressed samples is the same as the slope of samples compressed without electric current pulses indicating that short electric current pulses (0.5 ms) have only a short-term effect and do not influence the overall strain hardening behaviour of the eutectic Al–Cu alloy investigated. However, for a continuous electric field, Conrad et al. [5] observed a lowered strain hardening in metals. Similarly, Liu et al. [39] have reported a decrease of the strain hardening exponent with increasing current density for a TRIP steel when applying electric current pulses during deformation. However, in our...
experiments, the interaction of electrons with dislocations and interfaces are expected to dominate the EP deformation behaviour, whereas in the latter, the TRIP effect might show additional and/or different EP-induced mechanisms.

The observed maximum temperature induced by electric current pulses was 76 °C and after each applied electric current pulse the temperature decreased again to near room temperature. Since the heat loss due to radiation from the sample surface can be neglected [40], it is assumed that the measured surface temperature corresponds to the temperature inside of the sample.

This temperature increase causes thermal expansion of the samples, which can be estimated by applying the rule of mixtures for the thermal expansion coefficient of the composite material giving the composite coefficient as \( \alpha_C = 2.38 \times 10^{-5} \text{ K}^{-1} \) with the thermal expansion coefficients of the constituents taken as \( \alpha_{Al} = 2 \times 10^{-5} \text{ K}^{-1} \) and \( \alpha_{Cu} = 2.7 \times 10^{-5} \text{ K}^{-1} \) [41] and volume fractions of \( \chi_{Al} = 0.55 \) and \( \chi_{Cu} = 0.45 \)

\[ \text{assuming a maximum temperature increase, } \Delta T, \text{ of } 46.5 \text{ °C. The resulting maximum thermal strain amounts to } 0.11\%. \]

The modulus of the composite, \( E_C \), was also calculated using the rule of mixtures with \( E_{Al} = 75.15 \text{ GPa} \) and \( E_{Cu} = 99.28 \text{ GPa} \) [23] giving \( E_C = 86.01 \text{ GPa} \). The additional thermal stress, \( \sigma_{th} \), was calculated using Eq. (2)

\[ \sigma_{th} = E_C \cdot \varepsilon_{th} \]  

\[ \text{(2)} \]

to be 94.5 MPa.

When considering these induced thermal expansion and thermal stresses, the change in stress upon electric current pulsing is assumed to be due to three different effects, namely, the thermal expansion of the sample, the thermal softening of the material and the electroplastic effect. These results were not corrected for the unknown thermal expansion of the machine.

Thermal expansion of the samples induces an increase in stress, whereas thermal softening and the EPE cause a stress reduction. The stress increase due to thermal expansion amounts to 94.5 MPa at the maximum temperature observed during EP compression experiments when assuming that the temperature measured at the surface of the sample is similar to the temperature inside the sample.

In order to quantify the effect of thermal softening during the experiments, hardness measurements of Al and Al\(_2\)Cu at RT and 200 °C reported by Chen et al. [42], were further analysed. The hardness of the intermetallic Al\(_2\)Cu phase decreased from 5.77 GPa at room temperature to a value of 5.33 GPa at 200 °C whereas the hardness of the aluminium phase decreased from 1.45 GPa at RT to 0.68 GPa at 200 °C [42]. When assuming that the deformation mechanisms for both materials are unaffected by a temperature increase to 200 °C, the hardness drop can be calculated via linear interpolation. The assumption that the active deformation mechanisms are not affected by an increase in temperature to 200 °C is based on the results by Bai et al. [43] for Al and by Chen et al. [42] for Al\(_2\)Cu who reported no changes in hardness within the standard deviation indicating no change in the active deformation mechanisms. The resulting hardness value of Al\(_2\)Cu at 76 °C is accordingly calculated to amount to 5.66 GPa and the one of Al to amount to 1.25 GPa. The resulting composite hardness amounts to 3.24 GPa according to the rule of mixtures. This corresponds to a decrease in hardness of 0.16 GPa when the temperature increases from room temperature to 76 °C. When assuming a small strain hardening, this loss in hardness, \( H \), can be converted into a decrease in yield strength, \( \sigma_y \), according to Eq. (3) [44]:

\[ H = 3\sigma_y \]  

\[ \text{(3)} \]

It should be noted that the assumption of a low strain hardening coefficient overestimates the yield strength. The resulting upper bound on the decrease in yield stress due to thermal softening amounts to 52.8 MPa. As has been mentioned above, the stress increase induced by thermal expansion amounts to 94.5 MPa.

The stress drops measured during EP assisted compression experiments amount to 47.8 ± 4.8 MPa for the first electric current pulse and to 18.3 ± 1.6 MPa for the subsequent electric current pulses. When considering a decrease in stress due to thermal softening of 52.8 MPa and an increase in stress due to thermal expansion of 94.5 MPa, the softening induced by the EPE amounts to about 89 MPa during the first electric current pulse and about 60 MPa during subsequent electric current pulses. This supports the hypothesis that the observed stress drops are not solely induced by thermal softening but that the EPE causes an additional stress decrease. This assumption is further supported by the fact that the temperature increase observed during the first electric current pulse is similarly high as those observed during the subsequent electric current pulses. Therefore, the large stress drop occurring during the first electric current pulse cannot be a result of merely thermal softening.

4.2. Micromechanical EP experiments

4.2.1. Nanoindentation experiments with different current intensities and different pulsing intervals

An increase in the applied current intensity (Fig. 6a for the eutectic microstructure and Fig. 7 for the individual phases) results in larger displacement shifts at constant load. This observation is assumed to be caused by enhanced plasticity induced by electric current pulses.

Similarly, Okazaki et al. [10] and Troitskii [45], have reported larger stress drops with increasing current density. In the present study, an increase in current intensity is interpreted as increased current density at a constant pulse length.

The observed periods of low displacement response, which follow the rapid displacement shifts upon electric current pulsing are interpreted as the response of the material after the effect of the short-term electric current pulse is exhausted.

The observation that the applied current intensity influences the magnitude of the displacement shift indicates a dislocation motion related EPE mechanism. This is assumed to be either related to the de-pinning of dislocations from obstacles or an increased dislocation mobility or a combination of both.

Independent of the applied current intensity, the displacement shift induced by the first electric current pulse during indentation is significantly larger than those of the subsequent electric current pulses. This is consistent with our macroscopic EP assisted compression experiments that show that the first electric current pulse induces a stress drop that is more than two times larger than the subsequent electric current pulses. As has been discussed above, this effect is assumed to be most likely related to the de-pinning of dislocations from short-range obstacles induced by electric current pulses.

A longer inter-pulse interval results in larger displacement shifts (Fig. 6b)). This further supports our hypothesis that electric current pulses might induce the de-pinning of dislocations from short-range obstacles: a longer inter-pulse interval corresponds to a longer time between two electric current pulses during which more dislocations might become pinned than during shorter inter-pulse intervals. Consequently, more dislocations are assumed to be de-pinned during a subsequent electric current pulse resulting in a larger displacement shift.
Similarly, Troitskii [6] has reported that increasing the inter-pulse interval resulted in higher stress drops up to an interval of 5 s, beyond which no further increase in the stress drop magnitude for higher inter-pulse intervals was observed.

The measured hardness of the Al2Cu-θ phase at a depth of 1100 nm amounts to 5.3 ± 0.09 GPa without application of current pulses and 5.07 ± 0.53 GPa with application of 1.5 A (Fig. 8 a). The hardness value without application of current pulses corresponds well to those observed by Chen et al. [42] who reported hardness values of about 5.77 GPa at room temperature. The small differences might arise from the different solid solution content of the alloys as well as different crystallographic orientations. The observed decrease in hardness upon application of electric current pulses and increased standard deviation is assumed to be caused by experimental scatter due to the application of electric current pulses. Specifically, a current pulse might affect the measured hardness due to displacement shift and at the time increase the standard deviation (see Fig. 7). We assume that a pulsed electric current does not affect the overall hardness measured at the end of an experiment because, during further loading after a current pulse, the material is initially re-loaded elastically across the increased contact area and the dislocations are re-pinned during subsequent plastic deformation. If the lattice resistance was influenced by the electroplastic deformation, an effect on the overall hardness may be expected, but this has not been observed in our experiments.

The elastic moduli measured for the Al2Cu-θ phase without application of electric current pulses correspond well to the results by Chen et al. [42]. With applied electric pulsing, no change in elastic modulus is expected for drift electron — dislocation interactions [46] as these do not influence the atomic bonding strength that determines the material’s elastic response and therefore elastic modulus value.

However, a small increase in average modulus was observed for the Al2Cu-θ phase. A closer inspection of the load-displacement curves (Fig. 7a)) revealed that a different and linear gradient is indeed observed towards the top of the unloading curve. This indicates that the increase in modulus is not related to any time-dependent relaxation processes after the final pulse, as seen regularly in indentations affected by creep. Whether this unexpected increase in stiffness is related to the electroplastic deformation, e.g., through changes in the pile-up behaviour, or artefacts, e.g., tip wear, could not yet be resolved. However, we assume that the electroplastic effect is not influencing the atomic bonding strength, as the effect is mostly related to the interaction of the electric current with dislocations [46].

The hardness values obtained for the fcc-Al phase amounted to $0.34 ± 0.00$ GPa (Fig. 8 c), which is consistent with hardness values reported in literature where Liu et al. [47] reported hardness values of 0.47 GPa at an indentation depth of 100 nm for Al and Pharr et al. [48] reported a hardness value of 0.21 GPa for Al. The small deviations in the hardness values reported in the literature are assumed to arise from the different crystal orientations as well as different compositions investigated.

The elastic modulus measured for the Al α-phase remained approximately constant, as expected, and the value of 72.6 ± 1.4 GPa without application of current pulses corresponds well to literature where modulus values of 68.0 GPa [49] and 70.4 GPa [48] are reported. The calculated local hardness drops for the individual phases (Fig. 9) underline the effect that the first current pulse induces the largest drop in hardness. It is further observed that the hardness drops increase with increasing current intensity which is consistent with the experiments conducted on the eutectic microstructure.

The magnitude of the displacement shifts induced by electric current pulses during nanoindentation is observed to be rate-dependent (Fig. 10); a high loading rate induces larger displacement shifts during loading and unloading than a low loading rate when the same current intensity is applied.

This observed strain rate dependence cannot be explained by merely thermally activated dislocation motion. Higher loading rates would reduce the probability of dislocations to overcome the Peierls barrier by thermal fluctuations [50], hence, resulting in increased yield and flow stresses rather than decreased flow stresses as observed in the present study. This again indicates that the observed EPE cannot be only induced by thermal activation.

Further, the strain rate dependence of the EPE is controversially discussed in the literature [44–48]. Specifically, Troitskii [51], Ross et al. [52] and Varma et al. [53] have reported that higher current densities are required to induce the same EPE at higher strain rates indicating that the EPE decreases with increasing strain rate. In contrast, Cao et al. [54] have observed an increasing EPE with increasing strain rate for Nb, whereas Okazaki et al. [55] have observed no strain rate dependence of the EPE. It is therefore assumed that the strain rate dependence of the EPE is not controlled by a general mechanism but is controlled by the material and the predominant deformation mechanisms.

4.2.2. EP unloading experiments

Our nanoindentation experiments further reveal that electric current pulses during unloading also induce load-displacement

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**Fig. 11.** Comparison of the EPE during unloading of a tensile test and unloading during nanoindentation. (a) adapted from Reference [58] with permission by Elsevier, a) load-elongation curve of a tensile test on a polycrystalline titanium wire, current pulses ($12 \times 10^3$ A/mm², 80 μm) were applied during the test; b) nanoindentation load-displacement curve on a eutectic Al-Al2Cu sample, electric current pulses (3 A, 100 μs) were applied during loading and unloading. The inset in (b) shows an enlarged image of the unloading curves, they are off-set by a load increment to enhance distinctiveness.
steps, Fig. 10 and Fig. 11. The direction of the displacement shifts changes from displacement shifts towards higher displacements to displacement shifts towards lower displacements during unloading. Thermal effects such as thermal expansion of the tip and/or sample would not induce a change in the direction of the displacement shifts. It is therefore assumed that the displacement shifts during unloading are caused by increased reverse plasticity generated by the electric current pulses. This is consistent with the residual stresses accumulated during the displacement of the confined sample volume of the impression into the material. A driving force for reverse plasticity therefore naturally exists during indentation as shown, for example, during high temperature indentation in direct comparison with uniaxial micropillar compression [56,57]. In addition, residual stresses may be introduced due to anisotropy of flow in different crystal directions and within the different phases of the eutectic microstructure. A similar observation has been reported by Okazaki et al. [58] for polycrystalline titanium wires (Fig. 11 a)). They [58] observed short-term load drops in the load-time curve upon electric current pulsing during unloading in a tensile test. In the beginning of unloading, an electric current pulse induced a short-term load drop followed by an increase in load to a level slightly below the load level before the electric current pulse, \(\sigma_{p1}\). Whereas in the end of unloading, the electric current pulses induced load drops which were followed by an increase in load to load levels above the load before the electric current pulse was applied, \(\sigma_{p2}\). They [58] attributed the latter to reversed plastic flow.

This agrees to our observation that the direction of displacement shifts induced by electric current pulses changes its direction from towards higher displacements to towards lower displacements, Fig. 11.

Okazaki et al. [58] suggested that internal long-range stresses in the material are responsible for this effect. Specifically, if unloading exceeds the level of internal long-range stresses, electric current pulsing induces forward plastic flow, whereas at load levels below the existing internal long-range stresses reversed plastic flow is induced by an electric current pulse [58].

In the eutectic composite microstructure investigated in the present study, the two phases deform inhomogeneously and strain gradients are assumed to form during deformation, particularly in the vicinity of phase boundaries. These strains and those induced upon casting due to different thermal expansion coefficients of the constituent phases existing in the eutectic microstructure might induce long-range internal back stresses on dislocations [41,59]. Hence, electric current pulses might promote the motion of dislocations under these long-range internal stresses and cause the observed displacement shifts during unloading. Thermal softening alone would not cause such back stresses during unloading.

The difference in the maximum displacement when compared to the experiments shown in Fig. 6 and Fig. 10 are assumed to be caused by the composite microstructure, where the phase fractions and crystallographic orientations present in the microstructure below the indents vary.

5. Conclusions

We have performed macroscopic and micromechanical electroplastic deformation experiments on a eutectic Al-Al\(_2\)Cu alloy. To this end, we have developed a new nanoindentation setup to apply electric current pulses during nanoindentation. From our study we draw the following conclusions:

- Macroscopic compression experiments using electric current pulses (current density \(1.66 \pm 0.01\) kA/m\(^2\), pulse length 0.5 ms) show stress drops upon electric current pulses, as well as higher fracture strains and lower flow stresses than during compression experiments without electric current pulses.
- The temperature increased from room temperature to up to 76°C due to electric current pulses during macroscopic EP compression tests.
- Both the macroscopic and nanoindentation EP experiments show that the first current pulse induces a larger stress drop than the subsequent electric current pulses. This is assumed to be caused by the de-pinning of dislocations from obstacles.
- In electroplastic nanoindentation experiments (pulse length 100 µs), a higher current intensity, pulsing interval and loading rate result in larger displacement shifts. This indicates an increased dislocation mobility and de-pinning of dislocations from obstacles.
- Electric current pulses during unloading induce displacement shifts that change their direction during unloading from towards higher displacements to towards lower displacement. This is assumed to be caused by long-range internal stress fields present in the deformed microstructure.

CRediT authorship contribution statement

**D. Andre:** Methodology, Data curation, Formal analysis, Investigation, Writing - original draft. **T. Burlet:** Methodology, Software, Writing - review & editing. **F. Korkemeyer:** Investigation, Writing - review & editing. **G. Gerstein:** Investigation, Writing - review & editing. **J.S.K.-L. Gibson:** Methodology, Formal analysis, Writing - review & editing. **S. Sandløes-Haut:** Conceptualization, Funding acquisition, Supervision, Writing - review & editing. **S. Korte-Kerzel:** Conceptualization, Funding acquisition, Supervision, Writing - review & editing.

Acknowledgement

The authors gratefully acknowledge the funding of the priority program “Manipulation of matter controlled by electric and magnetic field: Towards novel synthesis and processing routes of inorganic materials” (SPP 1959/1) by the German Research Foundation (DFG). This work was supported by grant number 319419837 and grant number 319282412.

Data availability statement

The raw data required to reproduce these findings are available on request from the corresponding author of this study.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matdes.2019.108153.

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