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The effects of electropulsing on metallic materials

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Abstract

The influence of electropulsing on metallic materials is reviewed, with a focus on phase transformations and grain refinement. While a large and growing body of literature exists on the topic of electropulsing, the mechanisms governing the process are not currently fully understood. Furthermore, the effects of electropulsing on microstructure and mechanical properties are not yet clearly defined. This review seeks to summarise the existing literature in order to highlight and understand research trends across a variety of metals and alloys, and to clarify the state of the art. Research has shown that the electropulsing process is capable of inducing low temperature recrystallisation in metallic materials at an accelerated rate compared to more traditional heat treatment methods. These microstructural changes often alter the mechanical properties of the materials such as ductility, tensile strength and hardness. Crack healing as a result of electropulsing treatment has also been observed in damaged or work hardened materials and pre-deformation of the sample has been shown to enhance the effects of electropulsing.

Keywords: Electropulsing, Mechanism, Microstructure, Crack healing, Spheroidisation, Grain refinement

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1. Introduction

In the field of metallurgy, electropulsing is a relatively new and developing process. Since the 1960s, researchers have studied the effects of electropulsing, which can be simply described as the passage of electric current through a material. Troitskii pioneered the topic and applied the electropulsing technique to many highly deformed metals.[1, 2] Subsequent electropulsing studies have been carried out on a variety of metals and alloys including copper,[3,4] tin-lead alloys,[5] iron-based amorphous alloys,[6-8] steels,[9-11] titanium and titanium alloys,[12-16] magnesium alloys,[17-26] aluminium alloys,[27] silicon steel[28] and metallic glass of various compositions.[29, 30] Throughout this body of research, many different electropulsing effects have been observed. An accelerated microstructural recrystallisation process is consistently reported across a range of metallic materials, generally stemming from an enhanced nucleation rate, which leads to phase transformation and varying degrees of grain refinement.[5, 6, 14-16, 31-35] Changes to the mechanical properties associated with altered microstructure are also well documented.[10, 11, 27, 34, 36, 37] Additionally, electropulsing has been successfully used to heal microcracks and to reduce defects in a number of metals.[27, 38-42]. Electropulsing treatment has also been applied in combination with other processing methods to achieve various effects. Ivanov *et al.* combined electropulsing treatment with high energy electron beam treatment and applied the processing to various alloys.[43] The physical origin of the effects of combined processing on the improvement of the mechanical properties of steels has been investigated.[44] Ye *et al*. combined electropulsing treatment with ultrasonic shock.[45] The processing enabled significant improvement the surface quality of alloys.

Despite a wealth of research, the underlying mechanism of electropulsing remains unclear. A recent review by Qin assessed and highlighted the outstanding issues in electropulsing processing.[46] Some theories suggest that the effects of electropulsing are produced by the high heating and cooling rates associated with the process.[7, 30, 36, 47] Others focus on properties inherent to the electric current such as current density, additional free energy and the effect of the current on the movement of electrons.[10, 11, 18, 29, 48-51] More recently, multi-factor theories such as those describing the combination of thermal and athermal effects have been developed in an effort to explain the mechanism behind electropulsing. In addition, the effects of pre-deformation (such as cold rolling and extrusion) must also be considered; studies have shown that the effects of electropulsing on recrystallisation can be magnified in pre-deformed materials.[11, 18, 32, 52] Therefore, the specific processing conditions must also be considered; the effects of static electropulsing (where electropulsing is applied separately to any deformation or heating process) have been shown to differ to those of dynamic electropulsing (where electropulsing is applied in combination with deformation).[53, 54]

This literature review will present an overview of the progression of research on electropulsing. The paper will summarise key literature focussing on crack healing; grain refinement through phase transformation and recrystallisation; the effects of pre-deformation; and mechanical properties of electropulsed materials. Processing conditions will be considered throughout the review. Discussion will be presented on relevant theories, and suggestions for further work will be made.

2. Phase transformation: recrystallisation and enhanced nucleation rate

Many studies have demonstrated the capacity for electropulsing treatment to accelerate recrystallisation through enhanced nucleation rate, such as Conrad *et al.'s*[31-33] investigations into the effects of electric current pulses on recrystallisation in copper. In each study, it was found that the application of high density DC electric current pulses (current density $800\mathrm{A \cdot mm^{-2}}$, frequency 2Hz and duration 90μs) during the annealing of cold-worked copper enhanced the recrystallisation rate and reduced the frequency of twinning. Grain refinement was observed at lower annealing temperatures,[32] while grain growth was enhanced at higher temperatures.[31] Electric current was also thought to affect the rate of nucleation of new strain-free grains, by enhancing subgrain coalescence[I] associated with improved dislocation mobility.[33]

[I] Subgrain coalescence refers to a method of subgrain growth resulting from the dissolution of subboundaries. Alternatively, subgrain growth may occur through boundary migration, in which large subgrains consume smaller ones.[55]

It was also considered that localised heating effects and electron-defect interaction might influence the response of copper microstructure under electropulsing.[31] Finally, it was shown that a continuous $10\mathrm{A}\cdot\mathrm{mm}^{-2}$ DC current of equivalent Joule heating rate[II] to the pulsed condition did not produce the same effects on recrystallisation and recovery.[33]

Further research by Lai *et al.*[34] demonstrated the effects of electropulsing on the recrystallisation of iron-based amorphous alloys. The authors noted that low temperature recrystallisation was achieved using different electropulsing parameters for two separate alloys. They proposed that there was some direct action by the electric current over a short pulsing time and the effect could not be attributed to thermal excitation alone. Teng *et al.*[6] found that high current density electropulsing was capable of producing nanocrystallised grains in amorphous $Fe_{78}B_{13}Si_9$; enhanced nucleation rate was thought to be the critical factor here. Similarly, Ma *et al.*[35] observed localized nanocrystalline γ-Fe structure in boron steel after electropulsing at current density $890.1\mathrm{A}\cdot\mathrm{mm}^{-2}$. This effect was also attributed to an enhanced nucleation rate resulting from reduction of the thermodynamic barrier through additional free energy supplied by the applied current.

A review by Conrad[56] discusses the recrystallisation of cold-worked metals and alloys. As part of the review, Conrad states that the most significant effect of the application of electric current is on the nucleation rate as the majority of papers reported the influence of both electropulsing and continuous DC current on the rate of early-stage recrystallisation. Barnak *et al.* [5] cite electropulsing-induced undercooling combined with irregular lamellae spacing reduction as evidence for the effect of nucleation on colony or grain size in Pb-Sn alloy samples. They go on to discuss theories that might explain an electropulsing-enhanced nucleation rate at a relatively low temperature. The authors speculated that the effect of electropulsing on nucleation rate may stem from an increase in liquid-solid interfacial energy, or a reduced free energy difference

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[II] Joule heating is a heating effect that results from the interaction between electrons and atoms as electric current passes through a metal.

between the two states. In addition it was found that skin and pinch effects[III] were limited in their effect on the nucleation rate of the samples. This is supported by Okazaki *et al.'s* earlier work[57] on the electroplastic effect[IV], in which skin and pinch effects were found to have very little influence on electroplasticity in polycrystalline titanium during electropulsing.

In a review of the theory of electroplasticity, Conrad[58] noted that reduced flow stress had been observed in metals as a result of high density electropulsing. Conrad compared calculated and experimentally determined values of the electron wind push coefficient and determined that the reduced flow stress was a product of the electroplastic effect. Conrad proposed that the high density electropulsing produced an interaction between the electron wind and dislocations that improved the dislocation mobility, resulting in a reduced flow stress.

Electron wind is a well-known model used to explain electropulsing-induced recrystallisation.[19, 21, 26, 53-62] The electron wind provides an additional force that acts on dislocations in a material. Electron wind force is capable of enhancing the mobility of dislocations by providing heterogeneous electromagnetic shielding. This provides the extra push during dislocation movement through interaction with drift electrons, assisting dislocations in overcoming obstacles to their motion.[14, 58] In studies focussing on recrystallisation and grain refinement in titanium alloys and pure titanium sheet, Song and Wang[14-16] discuss the effect of electron wind force on nucleation rate and dislocation mobility. They describe how reduction in dislocation density and improved mobility of dislocations, induced by the electron wind force, enhanced the nucleation rate of recrystallisation and produced a refined microstructure.

Zhang *et al.*[53] studied dynamic electropulsing-induced phase transformations in AZ91 magnesium alloy. Electropulsing was applied simultaneously with rolling deformation; a schematic of the apparatus is shown in Figure 1.

[III] Skin and pinch effects are, respectively, the concentration of current in the proximity of the material surface, and the appearance of radial compressive stresses as a result of magnetic field-induced pressure.
[IV] The electroplastic effect describes the interaction between electrons and dislocations.

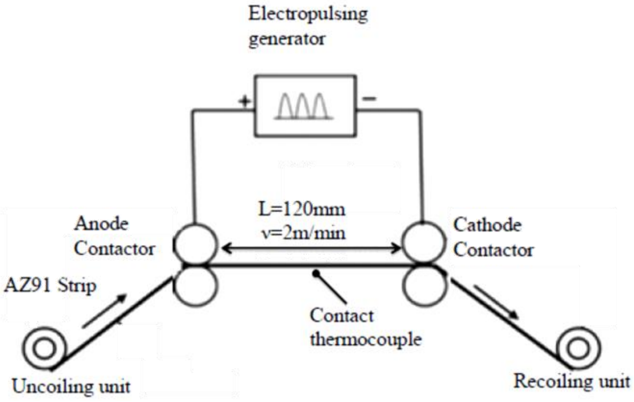

Figure 1. An example schematic of the dynamic electropulsing process. Reproduced from Zhang *et al* (2012), pp.17 Figure 1.

For comparison, static electropulsing was also conducted.[26] It was found that electropulsing was capable of inducing β-phase decomposition with increasing pulse frequency. However, while β-phase decomposition was observed in dynamic electropulsing samples, the static electropulsing samples showed a more accelerated phase transformation. The authors suggested that the accumulation and annihilation of defects in the alloy produced by the electropulsing-induced electron wind resulted in a reduced dislocation density with increasing frequency. Zhang *et al.*[53] believed that as the electric current acted to reduce the dislocation density, Gibbs free energy associated with the electropulsing process was used up through interaction between electrons and defects formed as a result of mechanical deformation. The authors state that the dynamic samples therefore possessed a lower total energy than the static specimens, thus reducing the capacity of the dynamic electropulsing process for acceleration of phase transformation. Xu *et al.*[21] also offer an explanation for a reduction in dislocation density, through the action of the electron wind pushing dislocations towards grain boundaries and resulting in an accumulation of defects. Simultaneously, annihilation of the accumulated defects was thought to occur as a result of the combined action of thermal and electromigration effects.

Similar findings presented by To *et al.*[54] focus on the application of dynamic electropulsing to a eutectic Zn-Al based alloy during tensile deformation. In this case, multiple electropulses of current density ranging between 35.80 and $104.39\mathrm{A \cdot mm^{-2}}$ produced increasing levels of decomposition of the η's phase with increasing current density. When the rate of phase transformation of static and dynamic electropulsing samples were compared, To *et al.* also found that the acceleration of phase transformation observed was much higher in static samples than in dynamic samples. However, it is not clearly stated in this paper whether accelerated phase transformation in the static electropulsing samples was obtained with or without prior deformation.

2.1 Current density

Research by Hao *et al.*[29] on electropulse-induced nanocrystallisation in amorphous alloys suggests that current density may be a critical factor in the crystallisation process. The study found that electropulse-induced low temperature recrystallisation in amorphous alloys a-$\mathrm{Cu_{50}Ti_{50}}$, a-$\mathrm{Pd_{80}Si_{20}}$ and a-$\mathrm{Zr_{60}Cu_{30}Al_{10}}$ was observed when initial current density surpassed a critical threshold value. The authors suggest that the mechanism for this behaviour may revolve around an athermal process based on collective motion of atoms; such concentrated electromigration behaviour was thought to enhance atomic diffusion. Further work by Mizubayashi *et al.*[48] concludes that inherent current density fluctuations exist in amorphous alloys and that high density regions experience a resonant collective motion of atoms due to electropulsing. Similarly to Hao *et al.*, the authors suggest that this resonant collective motion of atoms may induce dynamic atomic diffusion, thereby modifying the thermodynamic free energy of phases.

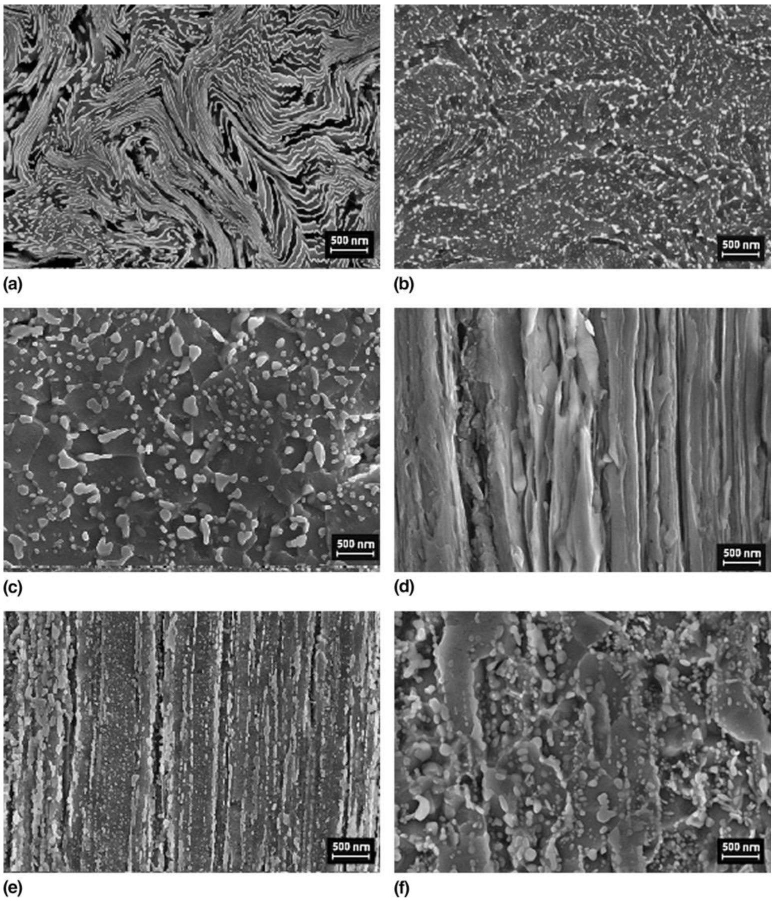

Figure 2. SEM micrographs demonstrating spheroidisation of cementite lamellae in deformed pearlitic steel samples. Reproduced from Samuel *et al* (2010), pp.1022 Figure 2.

In a study on the effects of electropulsing on the microstructure of a heavily deformed pearlitic steel, Samuel *et al.*[11] observed accelerated spheroidisation of the lamellar eutectoid structure. Spheroidisation is described as the breakdown of cementite lamellae in pearlite colonies into smaller globular grains and is visible in the micrographs in Figure 2.

The amount of spheroidisation was found to increase with the amplitude of the current density. In this case, the authors describe the enhanced spheroidisation process as a function of rapid temperature increase associated with increasing current density. It is thought that high rate heating can introduce a large number of vacancies into the steel, which migrate into grains and enhance the ability of elements to diffuse.

Zhou *et al.*[49] found that current density, in combination with high heating and cooling rate, is highly influential in determining the final refined grain size in low-carbon steel; increased current density and average heating rate led to a decrease in average grain size. The authors described the grain refinement process using a schematic illustration, shown in Figure 3.

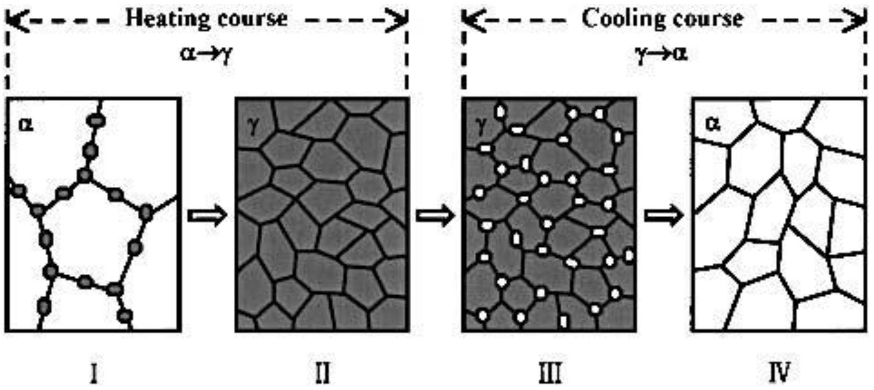

Figure 3. Schematic illustration for grain refinement and ultrafine grained microstructure formation: (I) formation of γ-phase nuclei, (II) formation of γ-phase grains by the growth of γ-phase nuclei, (III) formation of α-phase nuclei, and (IV) formation of α-phase grains by the growth of α-phase nuclei. Reproduced from Zhou *et al* (2002), pp.2110 Figure 5.

Due to the Joule heating effect and a decrease in the thermodynamic barrier, very small γ-phase nuclei form at an accelerated rate in the grain boundaries of the α-phase over the heating course. Grains of γ-phase form from the γ-phase nuclei; the short treatment time prevents grain growth, causing the grains to be very fine as the α-phase fully transforms into γ-phase. The inherent short treatment time necessary for grain refinement here suggests an important advantage to the electropulsing process over more time-consuming processes such as annealing. Over the cooling course, the process acts in reverse; α-phase nucleates at the grain boundaries of γ-phase grains at an accelerated rate due to the increased grain boundary volume fraction provided by the fine γ-phase grains. Finally, the γ-phase completely changes to refined α-phase grains. However, while Zhou *et al.* did emphasise the dominance of current density, heating and cooling rate in determining final grain size, they also stated that a decreased thermodynamic barrier and an enhanced nucleation rate contributed to the effects observed.

Qin *et al.*[10] found that the application of a single electropulse of highest density $9.61 \times 10^3 \mathrm{A} \cdot \mathrm{mm}^{-2}$ was capable of producing nanoscale particles in cold-drawn pearlitic steel, Figure 4.

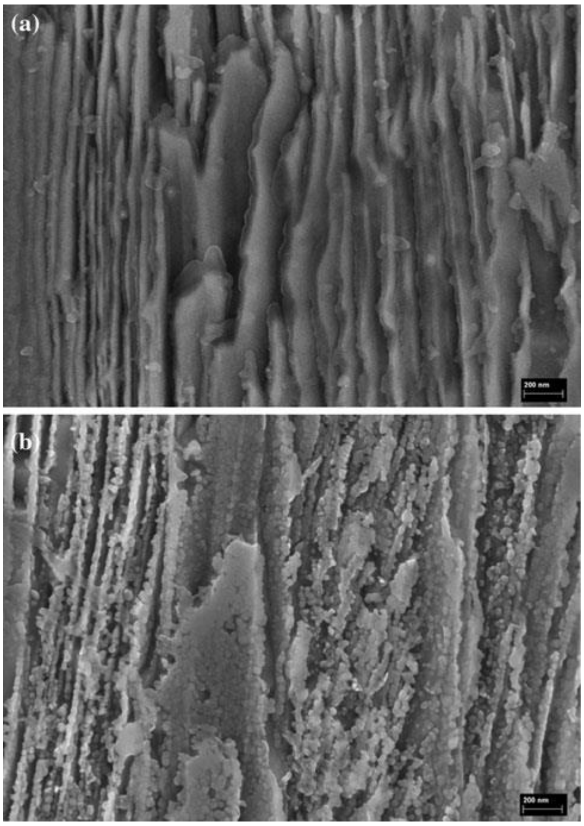

Figure 4. SEM micrographs of electropulse-induced microstructure transformation in pearlitic steel a) before electropulse and b) after electropulse. Reproduced from Qin *et al* (2011), pp.2839 Figure 1.

The application of electropulsing resulted in the formation of fine (approximately 30nm) spheroidised cementite particles that were distributed homogeneously across the sample. The refinement of the cementite particles was thought to stem from the additional free energy provided by the electropulsing process. An increased level of free energy during the microstructural transformation process allows a larger interface area for microstructural transformation, enabling finer particles to form. Further work by Samuel *et al.*[11] supported these findings, demonstrating the same microstructural spheroidisation and grain refinement with increasing current density.

Conrad[56] also highlighted the significance of current density in successful acceleration of solid state phase transformation, stating that a current density greater than $\sim 10^{3}\mathrm{A} \cdot \mathrm{cm}^{-2}$ $(or \sim 10\mathrm{A} \cdot \mathrm{mm}^{-2})$ was capable of accelerating phase transformations using both continuous current and electropulsing. However, work by Teng *et al.*[4] serves to highlight the complexity of the mechanisms describing recrystallisation behaviour in electropulsed samples. High current density electropulsing of $1800\mathrm{A} \cdot \mathrm{mm}^{-2}$ and pulse duration 40μs was applied to two amorphous $\mathrm{Fe}_{78}\mathrm{B}_{13}\mathrm{Si}_{9}$ samples. One sample (pulse frequency 20Hz for 65 minutes, producing an average Joule heating temperature of 493K) was fully recrystallised. The second (treated in two stages with pulse frequency 18 and 14Hz for 5 and 8 minutes, producing average Joule heating temperatures of 483 and 448K respectively) was partially recrystallised. Fine nanocrystallised grains were formed in both samples as a result of the electropulsing treatment. As the current density and pulse duration remained constant for both samples, this study suggests that other factors are capable of affecting the rate of crystallisation in metallic materials.

The research by Teng *et al.*[4] presents differences in crystallisation rate that appear to be linked to the variation of pulse frequency, total pulsing time and average temperature rise due to Joule heating. An earlier study by Conrad *et al.*[63] found a similar dependence on pulse frequency; grain growth in copper was retarded by electropulsing and the observed effect increased withpulse frequency over the range 0.07 to 7Hz. Furthermore, Conrad *et al.* observed that pulse duration had limited influence on the magnitude of electropulsing effects beyond a certain saturation time. As it was observed that the effects of electropulsing were not directly proportional to the number of drift electrons passing through a unit area, the occurrence of which might suggest a dependence on the pulse duration, Conrad *et al.* concluded that the effects of electropulsing were most significant when applied in pulse durations less than 50μs.

It is clear that while current density seems to be a critical factor in initiating recrystallisation, it is not the only component affecting the recrystallisation rate. The research described here shows that other variables, such as heating rate and processing method, are capable of affecting the recrystallisation behaviour of metallic materials. These factors will be considered in the following sections.

2.2 High-rate heating and cooling

The importance of high rate heating and cooling in enhancing recrystallisation has been discussed in depth throughout the literature; it has been suggested as a critical factor affecting the acceleration of recrystallisation. Early research by Xu *et al.*[36] on recrystallisation in cold worked α-Ti combined electropulsing with furnace heating. They found that when the combined temperature of the processes was maintained below 873K for 30 minutes, low current density electropulsing of $10\text{A} \cdot \text{mm}^{-2}$ produced grain refinement resulting from an enhanced nucleation and recrystallisation process. The authors conclude that the enhanced nucleation observed was a result of high heating rate produced by an electron wind.[V] Xu *et al.* also observed that when the temperature was maintained at 873K for 30 minutes, electropulsing produced coarser grains; increasing the current density from zero to $10\text{A} \cdot \text{mm}^{-2}$ led to further grain coarsening. Increasing current density was thought to enhance the rate of impurity migration, thus encouraging grain coarsening.

Results presented by Lai *et al.*[5] suggest that a minimum temperature rise induced by the Joule heating effect is necessary for the initiation of low temperature crystallisation. In tests on various iron-based amorphous alloys, samples cooled to lower the Joule heating effect to below an average temperature of 388K did not exhibit any microstructural change, while low temperature crystallisation was observed in samples that experienced an average Joule heating-induced temperature rise of between 626 and 668K. Zhang *et al.* [47] used an electric current pulse of maximum current intensity $2.6 \times 10^4\text{A}$ and pulse duration 400μs to induce a martensitic phase transformation in a titanium alloy. A phase transformation from α-Ti to β-Ti was observed; after electric current was applied, α-Ti grains were found to contain lamellae of both α-Ti and β-Ti phase. The martensitic phase transformation observed by Zhang *et al.* is stated as being the result of rapid heat treatment. In addition, Takemoto and Mizubayashi[30] found that when crystallisation occurred quickly at higher temperatures (above approximately

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[V] In this case the electron wind is described as a transfer of momentum between electron drift and the crystal lattice.

610K) in amorphous $Cu_{50}Ti_{50}$, nucleation at the early stage of crystallisation was accelerated. This behaviour was observed when an electric current of current density below $50\text{A}\cdot\text{mm}^{-2}$ was applied to the samples.

Research carried out by Zhou *et al.*[64-66] seems to confirm that as previously discussed, high heating and cooling rate alone is not sufficient to induce recrystallisation, phase transformation or grain refinement in metallic materials. The authors attempt to establish the role of high heating and cooling rate in producing enhanced recrystallisation behaviour in a Cu-Zn alloy by presenting a comparison of the effects of electropulsing with those produced by pulsed laser of equivalent heating rate. Initial research describes an electropulsing regime that produced a heating rate of $1\text{x}10^6$ K/s, with pulse duration 800μs, period 120μs and maximum current density $18.0 \times 10^3\text{A}\cdot\text{mm}^{-2}$. A similar heating rate was achieved by a 1000μs pulse from an Nd:glass laser. It was found that an increased number of small, homogeneous β'-phase precipitates developed in the electropulsed samples, but no microstructural changes were observed in the laser samples despite the similarity in heating and cooling rates[65]. The increased nucleation rate was thought to have been achieved through a reduction of the thermodynamic barrier as a result of the electropulsing treatment, rather than the rapid heating and cooling rates associated with the process. A second paper by Zhou *et al.*[64] using a similar electropulsing and laser treatment arrangement also reports the diffusion of atoms between α-phase and β'-phase during electropulsing treatment. Furthermore, Zhou *et al.* observed α-phase ultrafine-grained microstructure in electropulsed samples, but not in those treated by laser pulsing. The authors state that their results indicate that the generation of ultrafine-grained microstructure is dependent on solid-state phase transformations present in the α-phase but absent in the β'-phase, as no β'-phase ultrafine-grained microstructure was observed. Finally, a third paper by Zhou *et al.*[66] describes the observation of a diffusive phase transformation from α and β' phases to β in electropulsed samples, but no transformation in laser pulsed samples. It is important to point out that it is unusual to observe this type of phase transformation under rapid heating conditions, as long-range diffusion is required. The authors theorize that an enhanced diffusion coefficient resulting from the applied electric field may produce the observed phase transformation.

Further work by Zhou *et al.*[37] found that high rate heating, combined with enhanced dislocation mobility and atom migration, accelerates the nucleation rate of recrystallisation. In addition, Zhou *et al*[67] also found that electropulsing produced nanostructured γ-Fe in an initially coarse-grained low-carbon steel over a short treatment time. They suggest that this phase transformation may have resulted from high heating and cooling rates induced by electropulsing. This combined with an enhanced nucleation rate of the γ-phase through reduction of the thermodynamic barrier during electropulsing.

Similarly to current density, the literature shows that while high rate heating is an important factor capable of influencing the characteristics of electropulsing treatment, it is not the only factor capable of doing so. This suggests that the combined effects of current density and high rate heating might produce the electropulsing behaviours previously described.

2.3 Thermal and athermal effects

More recently, the multi-factor mechanism theory of thermal and athermal effects has been given significant consideration in the literature. Mechanisms that describe the thermal and athermal effects revolve around the combined actions of electron wind and Joule heating as discussed previously, combining electromigration with a rapid temperature increase. An early study carried out by Sprecher *et al.*[45] gave some indication of the nature of the relationship between these effects by investigating the factors governing the electroplastic effect. While thermal expansion due to Joule heating was found to be the most significant component of the reversible strain, other factors were also found to enhance the rate of plastic flow, due to an interaction between drift electrons and dislocation motion. Sprecher *et al.* conclude that while dislocations are affected by the athermal electron wind under high density DC current pulses, drift electrons may have also affected other parameters. These parameters were said to include strain rate, activation enthalpy and the effective stress.

Guan *et al.*[18] applied an electropulsing treatment to AZ31 magnesium alloy and concluded that thermal and athermal effects were responsible for the enhanced nucleation rate observed. In this case, the thermal effect was the Joule heating, due to a compressive stress gradient and pulsed electric current, while the athermal effects were attributed to the electron wind. This combination of factors was thought to enhance grain boundary migration as a result of accelerated interchange of vacancies and atoms through the increased driving pressure provided by electropulsing. Accelerated boundary migration then enhanced early stage nucleation and retardation of subsequent grain growth.

Research on the recrystallisation behaviour in electropulsed cold-rolled silicon steel strips by Hu *et al.*[28] discusses a similar theory of electron wind-accelerated dislocation climb leading to an enhanced nucleation rate and retarded recrystallised grain growth. Grain refinement was observed in the electropulsed sample (grain size 0.5-2.0μm) when compared to a non-electropulsed, heat treated sample (grain size 2-6.0μm). Electron migration was thought to be an important factor in electropulse-induced low temperature recrystallisation; this atomic flux was attributed to the thermal and athermal effects. Similar conclusions were drawn in earlier work by Xu *et al.*[2] focussing on dynamic electropulsing of copper wire. In this study, enhanced mobility of vacancies through the combination of thermal and athermal effects was also assumed to be a key factor in promoting dislocation climb and annihilation, leading to an enhanced nucleation rate and retarded growth of recrystallised grains. Moreover, increased atomic flux resulting from the thermal and athermal effects was also stated as being the probable cause of accelerated nucleation rate observed by Jiang *et al.*[23] in a study describing the effects of electropulsing on Mg-9Al-1Zn alloy strip. In this case, fine recrystallised grains were formed over a very short amount of time. Further work by Jiang *et al.*[25] attempted to provide greater detail on the nature of the relationship between the thermal and athermal effects. The authors found that the rate of accelerated recrystallisation observed in the Mg-9Al-1Zn alloy could only be achieved when the thermal effect due to Joule heating was large enough to accelerate dislocation climb and subgrain growth. The influence of the athermal effect on these factors was too small to induce recrystallisation behaviour alone.

A dynamic electropulsing experiment conducted by Xu *et al.*[19] on an Mg-Al-Zn alloy produced similar results, and here the authors go further in their explanation of the combined influence of thermal and athermal effects. They state that the enhanced mobility of dislocations and vacancies induced by the athermal effect acts to decrease the density of piled-up dislocations at grain boundaries, describing the electron wind force as being able to untangle sections of dislocations. The high temperature Joule heating induced by the thermal effect activates dislocations on non-basal planes, rearranging the dislocation structure. The density of dislocations was thought to be reduced by the coupled action of the thermal and athermal effects. Similarly, Liao *et al.*[24] observed that the novel dynamic thermo-electropulsing rolling (TER) process accelerated dynamic recrystallisation in AZ31 magnesium alloy by reducing the dislocation density through annihilation, climb and cross-slip resulting from self-diffusion-controlled thermal and athermal effects.

The thermal and athermal effects mechanism combines high rate heating with electric current-associated phenomena, experimental factors which have both been shown to alter the effects of electropulsing in some form during testing. This multi-factor mechanism therefore seems to be the most valid mechanism theory as yet proposed in the field, but further research is required to fully understand the processes involved.

3. <u>Crack healing</u>

When a material experiences damage or fatigue, microcracks develop. These cracks may grow and accumulate, eventually causing the material to fracture and fail. It has been shown by a number of papers that the application of electropulsing to metallic materials can slow, prevent and even heal cracks, often lengthening the lifespan of the material. Early research by Conrad *et* *al.*[42] suggested that electropulsing was capable of increasing the fatigue life and reducing the amount of intergranular cracking in polycrystalline copper. These improvements were thought to result from a reduction in the geometry and separation of fatigue-induced persistent slip bands, also referred to as an increased homogenization of slip. This behaviour was associated with enhanced dislocation mobility; electron-dislocation interaction or electromigration effects[VI] were proposed as mechanisms for the enhanced mobility required.

A study conducted by Zhou *et al.*[39] focused on the potential crack healing abilities of electropulsing in damaged 1045 steel samples. It was observed that the growth rate of cracks, formed during water quenching after heating to 1113K, was reduced after electropulsing treatment of pulse duration 0.2ms. Furthermore, equiaxial crystals were formed at crack tips, which acted to partially fill or heal the damaged sections as shown in Figure 5.

The researchers also noted the selective behaviour of electropulsing, observing more pronounced effects in damaged areas than in undamaged sections. This property is explained by regional resistivity and electric current detour distance: a damaged area would demonstrate a higher regional resistivity compared to an undamaged section, due to the greater volume of cracks and microstructural defects. As current moves through and detours around an area of higher resistance, a local increase in temperature is induced that acts to increase the observed effects of electropulsing. Zhou *et al.* also discuss the conditions necessary to achieve crack filling under electropulsing; rapid heating and thermal compressive stress resulting from delayed thermal expansion were thought to be capable of producing this effect. The authors propose three mechanisms for the process: dislocation fill, diffusion fill and compression fill.[VII] This theory was later used by Song *et al.*[12, 68] to explain the healing of primary defects and damage resulting from plastic deformation observed in TC4 titanium alloy sheet. Wang and Song[13] also

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[VI] Electromigration is the term used to describe the directed motion of atoms resulting from an applied electric field.[58] Zhou *et al* believed that electromigration was capable of reducing the volume of vacancies in a material.

[VII] During dislocation fill, dislocations experiencing enhanced mobility due to the applied electropulsing current transfer into the crack. During diffusion fill, the diffusional ability of atoms is enhanced by the electropulsing process and atoms diffuse towards the crack. Compression fill involves the movement of atoms into the crack through a thermal compressive stress.

suggested that local recrystallisation due to electropulse-induced temperature increase and thermal compressive stress produced microcrack healing in TA15 titanium alloy sheet.

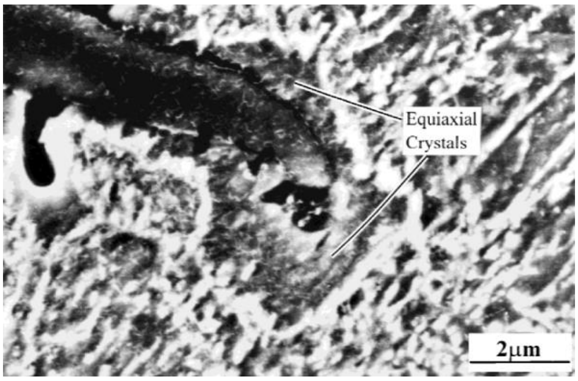

Figure 5. SEM micrograph showing equiaxial crystal formation at a crack tip. Reproduced from Zhou *et al* (2000), pp.1057 Figure 2.

Continued research by Zhou et *al.* has reported similar findings, highlighting the effects of electropulsing on crack healing in steels. Crack healing observed in 1045 steel, as shown in Figure 6 label A, was attributed to the effects of temperature and thermal compressive stress.[40]

Crack healing in another steel sample was described by a further paper as resulting from low temperature plasma produced by electrical breakdown of the crack, or the motion of effective atoms towards the crack. The motion of effective atoms was dependent on sufficient reduction of the crack face separation distance produced by thermal expansion.[41] Here, the motion of atoms theory expands on the idea of diffusion fill previously proposed by the authors. It is also noted that the selective properties of the healing process remove the need for information on location and geometry of a crack prior to healing, as damaged regions are self-identifying due to their higher local resistivity.

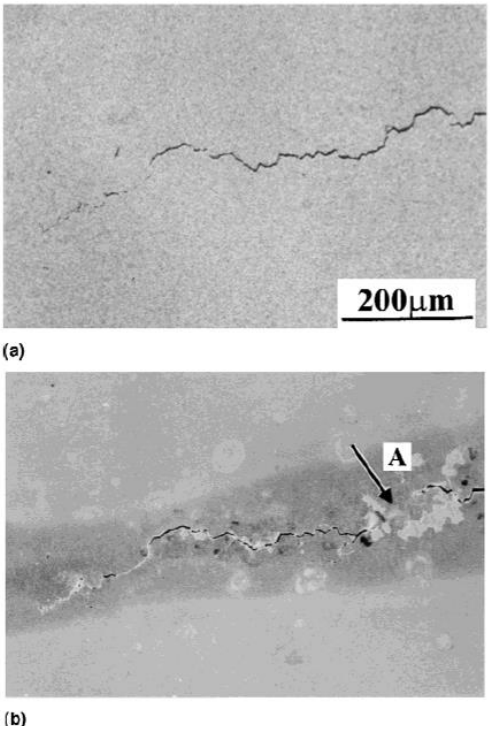

Figure 6. SEM micrographs of a crack in 1045 steel a) before and b) after electropulsing. Reproduced from Zhou *et al.* (2001), pp.18 Figure 1.

Evidence of the healing effect of electropulsing in other metallic materials is also present in the literature. In addition to the work by Conrad *et al.*[42] on polycrystalline copper, research by Qiao *et al.*[27] suggests that damage or work hardening in aluminium alloys 4043 and 2024 was partially healed by electropulsing treatment. In this case, the authors theorized that the damage recovery resulted from a decrease in the volume of defects in the samples due to electropulse-induced recrystallisation.

A theoretical study conducted by Qin and Su[38] produced a thermodynamic model of crack healing, incorporating the relationship between current density, crack geometry and healing driving force. They suggested that a critical current density exists, beyond which crack healing may occur in a range of materials[VIII]. The application of current was thought to reduce the driving force for crack growth, and increase the driving force causing the crack to shrink. Figure

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[VIII] In this study a range of materials were represented through variation in the value of Young's modulus applied in the calculations. Young's moduli of 80, 100, 150 and 200GPa were selected, with the actual Young's modulus of a steel and an Ni-Ti alloy measured as approximately 200Gpa and 83Gpa respectively.

7 suggests that the critical current density lies between approximately 30 and 60$\mathrm{A \cdot cm^{-2}}$ (or 0.3 and 0.6 $\mathrm{A \cdot mm^{-2}}$) for the range of metals considered. A critical current density is also proposed by papers focussing on electropulsing-induced phase transformations, discussed previously.[29, 56]

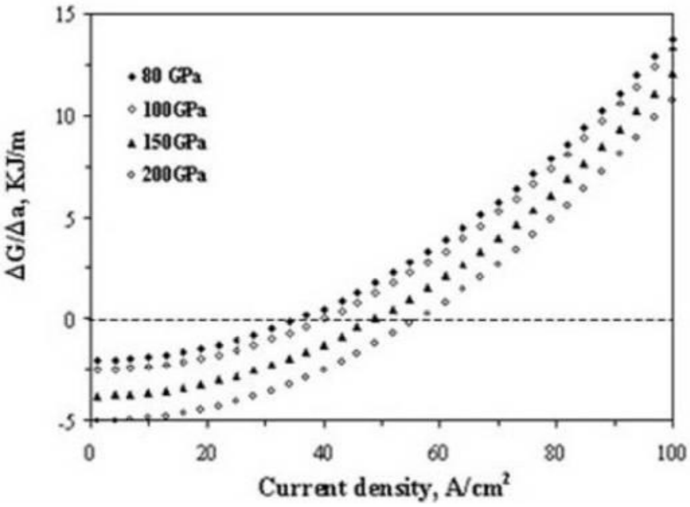

Figure 7. Healing driving force as a function of current density. Reproduced from Qin and Su (2002), pp.2050 Figure 2.

4. Pre-deformation effects

Several studies have shown that pre-deformation of metallic samples, most commonly through cold work, can affect the response of the sample to electropulsing treatment. Early research by Conrad *et al.*[32] studied how cold drawing affected the recrystallisation behaviour of copper during electric current pulsing. Pulses of 800 $\mathrm{A \cdot mm^{-2}}$ and 90μs duration were applied at a rate of two pulses per second to specimens of varying reductions. Increasing cold work produced a corresponding decrease in recrystallisation temperature in both annealed-only and electropulsed samples. However, while the electropulsed samples exhibited enhanced recovery and recrystallisation rates compared to annealed-only samples, the observed effects were less noticeable with an increasing amount of pre-deformation. Conrad *et al.* also observed that electropulsing produced a finer recrystallised grain size, but similarly, while cold work enhanced the response of the copper samples to electropulsing, microstructural changes were less pronounced with increasing cold work as demonstrated by Figure 8.

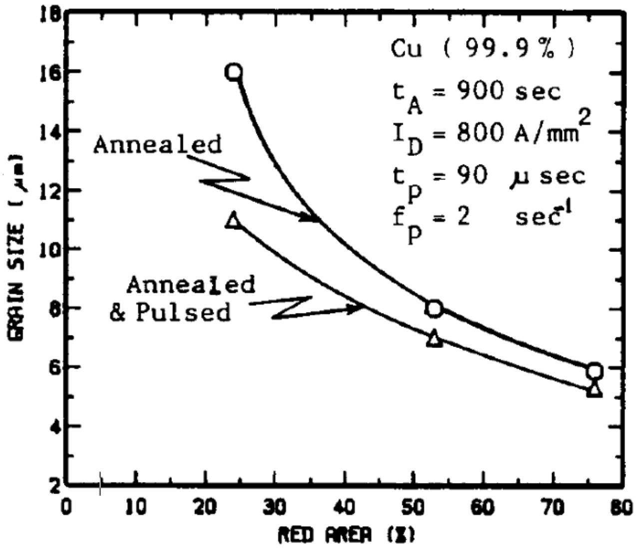

Figure 8. Recrystallised grain size versus amount of prior cold work as a function of current pulsing. Reproduced from Conrad et al (1984), pp.280 Figure 4.

The effects of cold work on the recrystallisation behaviour were explained by Conrad *et al.* to be a result of electric current-enhanced strain-free grain nucleation through subgrain coalescence. With increasing prior cold work, the amount of coalescence required decreases and hence the observed effect of electric current pulsing is lower.

Valeev and Kamalov[52] observed similar thermally-activated recrystallisation results through their work on the structure evolution of copper. Samples were cold rolled to strains of $e = 0.3$ and $e = 0.8$, defined in terms of the initial and final strip thickness, and electropulsed. As for annealed-only samples, the recrystallisation temperature of the electropulsed copper samples was observed to decrease with increasing pre-deformation. However, a reduction in the response of the recrystallisation behaviour with increasing cold work as described by Conrad *et al.*[32] was not considered in this case. More recently, Guan *et al.*[18] assessed texture evolution through electropulsing of cold-rolled AZ31 magnesium alloy strips, of rolling reduction 10%, 22% and 31% respectively. It was observed that at low rolling reduction (10% deformation), incomplete recrystallisation focussed around the grain boundaries occurred while at high rolling reduction (31% deformation), recrystallisation progressed until completion. Similar to previously described crack healing mechanisms, it was thought that the thermal and athermal effects were stronger in areas of higher defect volume due to the increased resistivity and enforced current detour in these regions. Therefore, the thermal and athermal effects would be expected to appear more powerful in the high rolling reduction samples which contain a larger volume of defects, due to this selective effect of electropulsing; a greater driving force is produced, thereby enhancing the rate of recrystallisation. Guan *et al.* also describe potential recrystallisation mechanisms to explain the variation of recrystallisation rate with cold work observed. The authors state that Strain Induced grain Boundary Migration (SIBM), in which an area experiencing strain in one grain boundary initiates the nucleation of previous grain boundaries, is a key factor in low rolling reduction recrystallisation. Fine grained microstructure is formed during electropulsing treatment as SIBM changes to the twin recrystallisation method, in which nucleation sites begin to appear in twins and shear bands.

Samuel *et al.*[11] applied high intensity electropulsing to pearlitic steel deformed to various strain rates. It was found that the application of electropulsing to cold-drawn pearlitic steel induced spheroidisation in the lamellar structure of the material, an effect that increased with current density. Furthermore, the authors observed that samples cold drawn to a higher true strain rate exhibited an enhanced spheroidisation process. As processes such as cold drawing introduce microstructural defects and dislocations, thus increasing the stored strain energy in the metal, the authors theorized that the energy storage associated with the deformation process is critical to the enhancement of spheroidisation. The spheroidisation enhancement effect increased with reduction in sample area and therefore the increased stored strain energy.

5. **Mechanical Properties**

The application of electropulsing has also been found to have various effects on the mechanical properties of different metallic materials. Samuel *et al.*[11] found that increasing the amplitude of current density produced a corresponding decrease in the Vicker's hardness of severely deformed pearlitic steel samples, as shown in Figure 9.

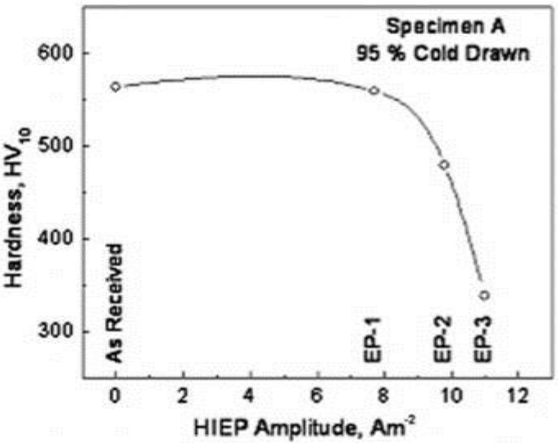

Figure 9. Variation in hardness with increasing amplitude of applied high intensity electric pulses. Vicker's hardness values were: as received sample 564HV$_{10}$; EP-1 559HV$_{10}$, EP-2 480HV$_{10}$, EP-3 339HV$_{10}$. Reproduced from Samuel *et al* (2010), pp.1022 Figure 3.

This behaviour was explained as resulting from microstructural change induced by electropulsing. The observed fine and coarse spheroidisation of pearlite lamellae and cementite particles, respectively, were associated with the decrease in Vicker's hardness. Similar behaviour was observed by Xu *et al.*[36] in α-Ti that was simultaneously electropulsed and furnace heated. A decrease in hardness was observed with increasing current density. Zhang *et al.*[26] also note that the Vicker's hardness of cold-rolled AZ91 alloy was reduced with increasing pulse frequency. In this case, the authors attribute the reduced hardness to the reduction in dislocation density associated with electropulsing treatment.

Conversely, Qin *et al.*[8] found that Vicker's hardness increased after electropulsing in a cold-drawn pearlitic steel, rising from 415HV$_{10}$ to 460HV$_{10}$ after a single pulse of highest density $9.61 \times 10^3 \mathrm{A} \cdot \mathrm{mm}^{-2}$. In this case, the authors state that the increase in hardness correlated with the observed grain refinement. In their study on electropulsing of iron-based amorphous alloys, Lai *et al.*[34] found that the hardness of the alloys increased slightly after electropulsing treatment of current densities 1540 and 758 $\mathrm{A} \cdot \mathrm{mm}^{-2}$ respectively. More recently, Lai *et al.*[5] also found that in general, the microhardness of iron-based amorphous alloys decreased and then increased with the application of electropulsing. However, the authors found no significant difference between the hardness of the electropulsed samples and samples that had simply been heated in a furnace to the same temperature.[5] This suggests that if the application of electric current is indeed capable of enhancing the hardness of metallic materials, the increase may be close to that observed in similarly annealed samples. If this is the case, then the main advantage inherent to this electropulsing property lies in the processing time required to produce such an effect, compared to more traditional methods such as annealing.

Zhou *et al.*[37] found that electropulsing was able to affect a number of mechanical properties of cold-worked brass. As shown in Table 1, electropulsed sample C demonstrated improved properties when compared with annealed samples A and B. In this case it was shown that electropulsing was able to produce improved tensile and yield strength and equivalent percent elongation over the annealing process. Zhou *et al.* attribute the improvement of mechanical properties in brass to the smaller, more uniform grain size formed through the application of electropulsing treatment.

Table 1. Tensile properties of samples. Reproduced from Zhou *et al.* (2004), pp.1950 Table 1.

Sample	Treatment conditions	Tensile strength $\boldsymbol{\sigma_b}$(MPa)	Yield strength $\boldsymbol{\sigma_{0.2}}$ (MPa)	Elongation percent (%)
A	Annealed (3o minutes, 843K)	383	176	41
B	Annealed (3o minutes, 923K)	350	113	47
C	Electropulsed (max. current density $17\times 10^3\mathrm{A\cdot mm^{-2}}$, approx. Joule heating 873K)	395	214	45

Hu *et al.*[28] produced similar results electropulsing cold-rolled silicon steel strips. They found that an electropulsed sample exhibited equivalent tensile strength and elongation properties to a traditionally heat treated sample after both samples had undergone full recrystallisation.

However, research by Qiao *et al.*[27] describes increased elongation in aluminium alloys, but a decrease in ultimate tensile strength. Wang and Song[13] suggest that electropulsing was capable of optimising the relationship between ductility and tensile strength in cold-rolled TA15 titanium alloy sheet, producing samples with improved elongation yet no obvious decrease in tensile strength when compared with samples that had not experienced any electropulsing.

It is also important to consider the effects of static and dynamic electropulsing, as some studies have shown significant differences in the mechanical properties resulting from this processing variation. Zhu *et al.*[61] observed that the elongation of a Zn-Al alloy sample dynamically electropulsed at current intensity 10A (current density $8.13\mathrm{A}\cdot\mathrm{mm}^{-2}$) increased by 437% when compared with a non-electropulsed sample. A supporting paper by Zhu *et al.*[62] found that while static electropulsing of a Zn-Al alloy was also capable of enhancing the plastic elongation by 57%, the dynamic electropulsing process was more effective in enhancing the elongation of the material. However as previously discussed, static electropulsing has been found to be more effective in accelerating phase transformation than dynamic electropulsing.[53, 54] These findings suggest that it is necessary to give careful consideration to the processing methods used. In this case, improved mechanical properties and accelerated processing must be balanced to achieve the desired material characteristics.

6. Discussion

Numerous studies have shown that electropulsing is capable of affecting or altering the microstructure and properties of various metallic materials. The literature available supports the idea that electropulsing can induce phase transformation through enhanced nucleation and recrystallisation rates, often resulting in grain refinement and enhanced mechanical properties. A number of factors have been shown to affect the recrystallisation behaviour of a range of metallic materials. In particular, research suggests that appropriate manipulation of current density is an important consideration when determining electropulsing parameters. Some studies have shown that with increasing current density, the rate of nucleation and phase decomposition may increase and the average grain size may decrease. Furthermore, it is possible that low temperature recrystallisation and the associated effects of electropulsing may only be observed above a certain threshold current density, a theory supported by the results of a theoretical study on crack healing mechanisms. However, studies in which low temperature recrystallisation and/or grain refinement are observed tend to involve a combination of high current density and high heating and cooling rate often resulting from the Joule heating effect.

Other theories available in the literature suggest that additional Gibbs free energy provided by electropulsing treatment may act to reduce the thermodynamic barrier, thus accelerating the nucleation rate. In addition, the range of electropulsing parameters applied across the literature often vary; while some information is available on optimised operating procedure, further research is required to produce appropriate application parameters for specific materials. However, in this respect some theories appear to hold constant over a range of materials. For example, pulse duration is thought to have limited effect on the electropulsing process beyond a saturation point, allowing advantageously short processing times. The majority of studies are able to produce recrystallisation in samples using extremely short pulse duration, suggesting that this variable can be neglected as a critical factor. Similarly, predeformation has been shown to enhance the effects of electropulsing in a range of materials, suggesting that the influence of deformation can be assumed to be relatively constant in that enhanced electropulsing behaviour is observed to different degrees in different materials.

The separation of the effects of key factors such as current density and heating rate is a complex process but the theory of thermal and athermal effects has been developed in an attempt to clarify the critical components of the electropulsing mechanism. Research in this area uses specific aspects of current and heating rate to produce a multi-factor mechanism theory. Generally, it is thought that the athermal electron wind effect acts to enhance the mobility and reduce the density of dislocations, introducing a force on the dislocations and resulting in their accumulation and annihilation at grain boundaries. The combination of electron wind with the thermal compressive stress-induced Joule heating effect results in an enhanced nucleation and recrystallisation rate. Mechanisms defined via the thermal and athermal effects may have particular merit, as numerous studies have highlighted the fact that no single parameter can determine the effectiveness of an applied electropulsing treatment. Moreover, a significant amount of the literature demonstrates that high rate heating and electromigration through high current density electropulsing are critical and interlinked parameters, therefore a multi-factor mechanism such as the thermal and athermal effects theory is particularly appealing.

Research on the influence of pre-deformation in electropulsing studies highlights an interesting opportunity for further optimisation of the process. A number of papers have observed the variation of electropulsing-induced recrystallisation behaviour with the rate of deformation. For each case considered in this review, the recrystallisation temperature under electropulsing was reduced when some form of pre-deformation was applied prior to treatment. Early research also suggests that an upper limit for pre-deformation rate exists, beyond which saturation of the effect occurs. In this case, optimisation of the processing parameters (strain rate or percent elongation/ reduction) would be necessary to improve the time and energy efficiency of electropulsing treatment. A number of theories that seek to explain the exact effects of pre-deformation on recrystallisation temperature under electropulsing are presented in this review. The selective effect of electropulsing is particularly interesting as evidence for the behaviour has been observed through both crack healing and pre-deformation research independently; such a correlation suggests that this theory may have particular merit. Furthermore, the fact that no previous information on crack geometry or location is required before healing may begin is a particular advantage. While it is clear that the selective healing property of electropulsing encompasses particular inherent procedural efficiencies that might encourage further development of an associated healing process for use in the field, the complete mechanism describing crack healing in metallic materials is not yet clear.

While the exact mechanisms behind the electropulsing process are yet to be clarified, it is evident that the microstructural changes induced by such treatment often result in altered mechanical properties. Critically the mechanical property enhancements observed in electropulsed materials are often observed to equal or surpass those obtained by more traditional heat treatment processes such as annealing, which generally require much longer processing time. In particular, the literature highlights elongation as a property consistently enhanced by electropulsing treatment in a range of metallic materials. It is thought that this enhancement is a result of the finer, more uniform grains often observed in electropulsed materials.

However, it is also important to remember that any improvement in ductility should theoretically result in a decrease in tensile strength, as a high tensile strength will generally result in a decrease in elongation or ductility, and vice versa. Despite this, the literature suggests that in some cases ductility can be improved by electropulsing with little or no detrimental effect on the tensile strength. Some research also theorizes that electropulsing may be capable of producing an optimum balance between elongation and tensile strength. Further research is required to clarify this behaviour; a process capable of producing a high tensile strength metal with high ductility would have potential advantages for the manufacture and forming of high impact structures and armour, for example. Electropulsing has also been observed to affect the hardness properties of a range of metallic materials, but the effects are still unclear as various studies have demonstrated both increasing and decreasing hardness as a result of electropulsing. Further research is necessary to understand the hardness properties of metallic materials under electropulsing; in both cases of increased and decreased hardness, grain refinement is cited as most likely to cause the observed change.

7. **Conclusion**

Throughout the literature, a number of factors have been identified as having some effect on the outcome of electropulsing treatment. However, it seems increasingly clear that no single factor can be identified as governing the mechanism behind the process. The theory of thermal and athermal effects seems to be the most robust mechanism theory available in the literature to date, as it takes into account multiple factors that have been shown by previous studies to have some effect on recrystallisation behaviour in metallic materials. However, theories focused on the addition to the system of electropulsing-induced Gibbs free energy must not be ignored; our limited current understanding of the mechanisms involved prevents the theory of enhanced nucleation resulting from a lowered thermodynamic barrier from being excluded.

Further research is required to fully understand and clarify the mechanisms underlying the electropulsing process. It would also be advantageous to optimise the processing parameters for individual materials in terms of pre-deformation, current density and processing method (static or dynamic electropulsing) in order to fully utilise the potential advantages of short processing time and low energy manufacture over traditional methods such as annealing. Research must be carried out to clarify the varying mechanical property changes observed across the literature, as the process cannot be feasibly applied in industry without such data. Finally, further work in the area of crack healing could be implemented, as a fully developed crack healing process could potentially be utilised in industry in a number of pre-existing applications. Crack healing is a reactive process ultimately intended to be applied to a work hardened operational material and, as such, it is possible that there would be limited or no requirement for alterations to current infrastructure or material manufacturing methods.

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