PORTEVIN–LE CHATELIER EFFECT IN ALUMINIUM ALLOYS

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Abstract:

Due to their excellent corrosion resistance, lightweight, hardenability, and high recyclability, aluminum alloys have become a staple in the automotive industry, particularly in manufacturing car bodies. However, the Portevin–Le Chatelier (PLC) effect, which manifests in certain aluminum alloys during plastic deformation, poses significant challenges. This phenomenon can lead to undesirable visual and structural defects, thereby limiting the broader application of these alloys in the automotive sector. Understanding the PLC effect is crucial for enhancing the usability of aluminum alloys in vehicle production. This paper addresses the PLC phenomenon, exploring the various parameters that influence its occurrence during plastic processing. By investigating these factors, the aim is to provide better insight into this phenomenon.

1 Introduction

Aluminum alloys are increasingly used in the automotive industry, especially in the production of car bodies, to reduce the weight and fuel consumption of vehicles [1]. Due to their corrosion resistance, lightweight, hardenability and high recycling rate, aluminum alloys are widely used for various applications [2]. Aluminum alloys are divided into groups such as 1xxx, 2xxx, 3xxx, 4xxx, 5xxx, 6xxx, 7xxx and 8xxx, depending on the leading alloying element in the alloy [3]. The 5xxx series are alloys with magnesium as the leading alloying element, which are used for various applications in the automotive industry due to their high strength-to-weight ratio, good corrosion resistance, weldability, and good formability. The addition of magnesium to these alloys increases the strength through the hardening mechanism due to the interaction of dislocations with each other or with precipitates of dissolved elements and different phases [4]. As at most engineering metal materials, there is a normal plastic flow in the deformation zone of aluminum alloys during plastic processing. However, it has been found that there are exceptions for certain alloys where the plastic deformation process is inhomogeneous under certain conditions and plastic instabilities occur. Examples of such instabilities are the Lüders bands [5] and the Portevin - Le Chatelier (PLC) effect [6]. AlMg alloys of the 5xxx series are an example of alloys in which unstable plastic flow occurs under certain conditions of plastic deformation, which is associated with the PLC effect in the literature. The PLC effect manifests itself as the localization of deformations in the form of deformation lines that lead to repetitive serrations on the stress-strain curve during tensile tests [7, 8]. This phenomenon limits the application of these alloys in the automotive industry, as rough and undesirable marks can appear on the surfaces of products made from certain aluminum alloys where the PLC effect occurs due to deformation during the forming process. The localization of the deformation significantly reduces the quality of the surface and leads to different roughness. These surface changes represent initial cracks and places of stress concentration during the processing of such materials, which can potentially lead to premature failure due to material fatigue [10]. In addition, the occurrence of the effect on materials leads to an increase in stress and hardness, affects the tensile strength and hardening rate, while on the other hand it reduces the ductility of the metal and the sensitivity coefficient to the deformation rate [11, 12].

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2 Instabilities of plastic flow of AlMg alloys

Plastic deformation is observed in most metallic materials as uniform flow of the material in deformation zone, which is manifested as uniform strain hardening on the stress-strain diagram. During the process of plastic deformation of some the AlMg alloys, an unstable and inhomogeneous plastic flow occurs under certain deformation conditions, which manifests itself in the stress-strain diagram as serrations on the strain hardening curve. In such a plastic flow, various phenomena arise that cause visual and structural changes, and they are the subject of ongoing research. Examples of such phenomena are Lüders bands and the Portevin-Le Chatelier (PLC) effect, which is characteristic of AlMg alloys, where instabilities manifest themselves as deformation localization in the form of deformation bands [13]. Repeated increases and decreases in stress lead to discontinuities in the strain hardening curve [14,15]. A comparison of the hardening curve during homogeneous and inhomogeneous plastic flow of the material is shown in Figure 1.

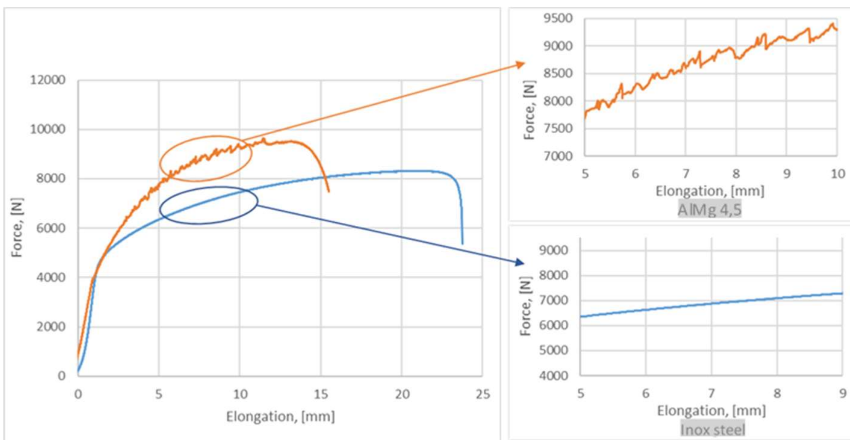

Figure 1. Force-elongation diagram of inox steel and AlMg 4.54 alloy

The Portevin-Le Chatelier effect, also known as the PLC effect, was originally observed by F. Savart (1837.) and A. Philibert Masson (1841.) in an experiment on samples of brass, iron, steel, copper and zinc subjected to a tensile dead-weight loading, which gave it the name "staircase phenomenon". But this plastic instability attracted more attention later, with the publication of the work of F. Le Chatelier (1909) on repetitive oscillations in the plastic flow of mild steels tested at elevated temperatures during the study of blue brittleness, and later, following the work of A. Portevin and F. Le Chatelier (1923), (1924), on the serrations of the hardening line in duralumin samples tested at room temperature at different stretching rates [16]. The PLC effect occurs not only in AlMg alloys [17], but also in other alloys of great industrial importance, such as some types of steel, e.g. TRIP steels, in which plasticity is caused by phase transformations, and TWIP steels, in which plasticity is caused by twinning [18], as well as in copper alloys [19]. In addition, the occurrence of the PLC effect has also been observed in nickel-based superalloys, which play an important role in the manufacture of aircraft engines and industrial gas turbines [20]. Considering the occurrence of the effect in different alloys with a specific application, it is necessary to investigate the influence of the deformation parameters on the PLC effect and to avoid the deformation range in which the effect occurs.

2.1 The mechanism of the PLC effect

The generally accepted and one of the first mechanisms at the microscopic level of the PLC effect is the Cottrell model, which is based on the interaction between mobile dislocations and atoms of dissolved elements in the alloy [21]. By this mechanism we can say that in AlMg alloys, dissolved magnesium atoms and/or formed precipitates interact with dislocations. This model is also referred to as dynamic strain aging (DSA) [21, 22]. In the case when the concentration of solute atoms around the dislocations is sufficient, the movement of the dislocations is blocked, leading to a decrease in the mobile dislocations and an increase in the stress. When the stress reaches the value at which the blocked dislocations are released or the multiplication mechanisms are activated, the number of mobile dislocations increases, causing the stress to decrease. The alternating increase and decrease of stress lead to a discontinuity on the hardening curve, which corresponds to the occurrence of deformation aging [18, 23]. According to new theories in the literature, DSA corresponds to the interaction resulting from the diffusion of solute atoms around dislocations. This leads to the creation of a atmosphere of solute elements and temporarily stops the movement of dislocations, resulting in the appearance of negative strain rate sensitivity (nSRS) [24]. This is explained by two main diffusion mechanisms associated with DSA: volume diffusion and pipe diffusion [7, 25]. The mechanism of volume diffusion is favoured by the formation of vacancies during plastic deformation, while the mechanism of pipe diffusion corresponds to the diffusion of additional atoms of dissolved elements along stationary dislocation lines towards moving dislocations while they are stopped. Each of the above mechanisms or their combination, the diffusion of atoms of solute elements on mobile dislocations, causes their temporary pinning. Dislocations can overcome these obstacles by thermal activation due to applied stress which results in unpinning of them.

The repeated stopping and restarting between moving dislocations and solute atoms lead to a serrations in the stress-strain curve, which is ultimately known as the PLC effect [7, 25]. McCormick's model [22] additionally considers the interaction between the waiting time required for the solute atoms to diffuse into the dislocation and the time required for the diffusion of solute atoms itself. Therefore, DSA occurs when the waiting time allows the solute atoms to diffuse into the arrested dislocations and effectively pin them. DSA occurs when the waiting time and the characteristic diffusion time of the solute atoms are of the same order of magnitude. The comparison of these two times is an important method for understanding the microscale mechanisms underlying experimentally observed phenomena. At the microscopic scale, dislocations move according to the stop-and-go principle described earlier, and diffusion of solutes occurs when dislocations are temporarily stopped [22, 26]. A lower strain rate usually results in slower dislocation movement, making dislocation stopping more efficient, and vice versa. In the current picture of dynamic stress aging according to the contribution of Cottrell and McCormick, solute atoms diffuse either through the bulk or along the dislocation to form mobile dislocations that are temporarily stopped at obstacles during the waiting time. The longer the dislocations are stopped, the more stress is required to set them in motion again. If the DSA is large enough, the critical stress for dislocation movement increases with increasing waiting time or decreasing strain rate. When these dislocations break out, they move at high speed until they stop again. At high strain rates, or low temperatures, the time available for solute atoms to diffuse to the dislocations to stop them is reduced and therefore the stress required to restart them is reduced. PLC instability therefore manifests itself in the range of deformation rates and temperatures in which these two-time variables are of the same order of magnitude [27]. According to the DSA theory, strain rate sensitivity becomes negative (nSRS) in a certain range of temperature and/or strain rate. Under these conditions, unstable plastic flow is observed, and the material loses its deformation properties. The stress-strain dependence then takes the form of a curve in the shape of the letter N [17].

2.2 Types of PLC bands

During the visualization of plastic flow, localized deformations are observed in the form of bands that move through the sample during the plastic deformation. Bands are mainly characterized by their appearance on the stress-strain curves, and accordingly there are three main types of strain lines A, B and C. The type of lines that appear depends on the alloy, although they can change in the same alloy, which ultimately depends on the strain rate and temperature [28]. The rarer band types E and D are also mentioned in the studies on these phenomena [7]. Although numerous studies have been carried out on their occurrence the parameters that influence the occurrence of the bands themselves and that cause them to change from one type to another are not yet fully understood. Research [29] has proposed a new method to quantitatively describe the bands by estimating the speed of the bands and relating the property of the bands to the properties of the material. Authors state that this new approach could provide an efficient quantitative analysis of the band properties and could be applied to any material exhibiting the PLC effect [30]. Type A bands occur at higher strain rates or lower temperatures, type B bands occur at medium strain rates or temperatures and finally type C bands occur at low strain rates or high temperatures. Type A bands are bands that propagate, i.e. when a deformation band appears, usually at one end of the specimen, the band propagates continuously along the specimen until it reaches the other end or until a new band appears, which then carries another deformation [24]. Bands of this type are observed on stress-strain curve by periodic increases and decreases in stress. Each stress rise precedes the nucleation of a new line. The subsequent propagation towards the opposite end occurs at lower stress and is usually accompanied by irregular stress fluctuations [17, 31]. Type B bands are of hopping nature, they do not spread like type A across all length of the specimen, they only move over short distances after nucleation [31, 32]. Type B bands can form anywhere on the specimen during plastic deformation, and a new band nucleates at an almost constant frequency along the previous one. As the strain rate decreases, more regular stress oscillations are observed, and each stress drop can be associated with a single band [31]. At the lowest strain rate, deep drops below the nominal stress level are observed, which is characteristic of type C bands. Each of these drops is caused by a separate deformation band. In contrast to type B bands, however, the nucleation is not clearly associated with earlier bands. The lower strain rate, more random nucleation of the bands becomes. Therefore, type C bands are stochastic in nature and the nucleation occurs randomly along the measured length of the sample. The loss of spatial and temporal correlations significantly distinguishes them from type A and B bands [33]. Type D bands occur at a high imposed strain rate. When this type of line occurs, the stress-strain curve looks like a staircase where the stress remains constant during propagation of the deformation band through the specimen. Finally, type E lines correspond fairly closely to type A bands, but with a more gradual decrease in stress [28, 33].

3 Microstructure of AlMg alloys

AlMg alloys have requirements for sufficient strength and for corrosion resistance. Thanks to the good solubility of magnesium in an aluminium solid solution, AlMg alloys have a pronounced ability to harden by alloying and deformation, a good ratio of strength and ductility, a great ability to form in complex operations, corrosion resistance and good weldability. Most alloys of this type have a microstructure consisting of a solid solution (α). The solubility of magnesium in aluminium increases with temperature and reaches a maximum value of 17.4 wt. % at the eutectic temperature of 450 °C. In the composition range 34.8-37.1 wt. %, Mg leads to the formation of Al_3Mg_2 phases (β) [34]. The decomposition of a supersaturated solid solution (α) is associated with the nucleation and growth of precipitates (β), which can occur by various mechanisms. An increase in Mg concentration in aluminium alloys promotes homogeneous precipitate nucleation. The presence of precipitates contributes to the occurrence of the PLC effect and increases the localization of deformation during plastic deformation. In solid solution, magnesium tends to slow down dynamic recovery as it affects the increase in dislocation density. The increase in dislocation density leads to material strain hardening in the case of cold deformation, while the reduction in the mobility of the dislocations reduces the possibility of plastic deformation [23, 34]. In addition to alloying, aluminium alloys can also be hardened by precipitation, aging or by strain aging. The decomposition of the AlMg alloy produces a series of precipitates depending on the applied aging temperature:

$$\alpha_{ss} \rightarrow \text{solute-rich clusters} \rightarrow \beta'' \rightarrow \beta' \rightarrow \beta$$

Where α_{ss} is the aluminium matrix with Mg in a supersaturated solid solution, β'' is the chemical composition Al_3Mg, β' is the intermediate phase of the chemical composition Al_3Mg_2 and β is the equilibrium phase of the chemical composition Al_3Mg_2. AlMg alloys that do not harden by ageing usually harden at Mg contents above 7-8 wt. %, which is due to the formation of coherent and semi-coherent AlMg precipitates, while no hardening effect at all is observed in alloys with a lower Mg content after aging [34]. It is known that work hardening is accompanied by an increase in plastic yield stress due to the formation of a larger number of dislocations that accumulate at obstacles within alloys such as dissolved atoms, precipitates, grain boundaries, etc. [35]. As the stress and deformations increase, the dislocations interact and can accumulate at the grain boundaries. When saturation is reached, the deformation can continue in a steady state where the rate of dislocation formation is equal to the rate of accumulation during dynamic recovery, in which we can observe a normal and uniform plastic flow. In the presence of dissolved atoms and small particles, the recovery rate is reduced, and attachment of the solute and particles occurs, leading to solute hardening which can be observed in AlMg alloys with a higher Mg content. Solute atoms are considered immobile at lower temperatures, while their diffusional mobility increases at higher temperatures and effectively attracted to dislocations, then the velocity of dislocations decreases because they have to move with the solute atmosphere [34-36]. The optimal percentage of magnesium in alloys is around 4.5 %. At this percentage, it is possible to achieve high strength without significantly reducing plasticity [14]. The addition of Mg influences the stacking fault energy and thus the strength, recovery and recrystallization characteristics of aluminium alloys. The presence of dissolved magnesium atoms can lead to the occurrence of plastic instabilities, which manifest themselves in a serrated hardening curve and a phenomenon known as the Portevin-Le Chatelier (PLC) effect [37, 38].

3.1 Influence of Mg and other alloying elements on the effect

The influence of Mg on the effect has already been shown in the previous chapters. It can have a significant influence on the hardening mechanisms, and it is similar for other alloying elements. The influence of alloying elements on AlMg alloys and the occurrence of the PLC effect can be explained by the interaction of Mg atoms and dislocations. The higher the magnesium content, the more obstacles occur, and the DSA becomes more effective, resulting in a strengthening of the PLC effect.

The increased activity of the DSA causes a stronger propagation of the deformation bands and increases the differences in values between the rise and drops of the stress [8]. The addition of a high Mg content increases the strength, but the ductility and formability decrease due to the occurrence of nSRS and the PLC effect. In study [39] on AlMg alloys with 1%, 3% and 5% Mg, the influence of Mg and the additional elements Sc and Zr on the occurrence of the PLC effect was investigated. An increase in the magnesium content in the alloys leads to an increase in the PLC effect, which has been reported in several studies [8, 39-41]. The results with additions of Sc and Zr show that they not only reduce the intensity of the PLC effect but also lead to a lower intensity of the stress drop at all deformation rates. In general, it can be concluded that the addition of certain elements leads to a reduction of stacking fault energy, which ultimately leads to a weakening of the PLC effect. Furthermore, the addition of Sc and Zr contributes to grain refinement and to a lower diffusivity of the Mg atoms. This is due to the presence of dispersoids (AlSc) at empty places in the crystal lattice, which simultaneously increase the dislocation density, reducing the waiting time required for stopped mobile dislocations to restart [39]. In the study [23], the influence of precipitates on the PLC effect in an Al-Zn-Mg-Cu alloy was investigated. It was found that the presence of precipitates contributes to the occurrence of the PLC effect and enhances the localization of deformation during plastic deformation. A similar study was carried out on Al-Mg-Si alloys in which the influence of various precipitates on the PLC effect was also investigated. It was found that the formation of precipitates reduces the initial stress and that the serrations are more pronounced. The results indicate that the deformation-induced dissolution of precipitates and their new formation play an important role in the development of the PLC effect.

The PLC effect is influenced by deformation, deformation rate and temperature [12, 25, 42], then by grain size, dislocation density, type, size and distribution of the dispersoids [43, 44-46]. The occurrence of the effect takes place under different deformation conditions, and dislocations interacting with solute atoms in the alloy are considered to be the main contributors to plastic deformation [44]. Although many factors influence the occurrence of the PLC effect, they do not necessarily have a significant influence. On the AlMg alloy as an example, the phenomenon can be observed as a function of the Mg content. According to [44], the DSA and the occurrence of a negative sensitivity to the strain rate increase with increasing Mg content, which leads to a strengthening of the PLC effect. Looking at the results of the tensile tests, it is found that the critical deformation at a certain temperature increases with increasing strain rate, which we consider as normal behaviour. However, when the critical strain is reduced by increasing the applied strain rate, the inverse behaviour is observed, resulting in a U-shaped curve of critical strain [17, 47].

3.2 Influence of temperature and strain rate on the effect

The temperature of plastic deformation itself has a significant influence on the PLC effect. In [44], where aluminium alloys with 4, 6 and 9% Mg were tested at different temperatures and different strain rates, in addition to the obvious influence of temperature and strain rate on the effect, it was observed that during the static tensile test, the maximum stress values increased with the increase of Mg content in the alloy. In a study on AlMg alloys with different Mg contents [44], it was shown that an increase in the deformation temperature decreases the stress values compared to the values at room temperature. Dynamic recrystallization is visible in the samples with higher temperatures, and a reduction in the serrations on the strain curve is observed during the static tensile test. Xu (2022) [12], Cho (2020) [44], Bakare (2021) [39] have investigated the influence of temperature and deformation rate on various AlMg alloys with different magnesium content in their works. It was found that both parameters have a strong influence on the occurrence of DSA. According to [12], faster diffusion of the dissolved atoms of the elements at higher temperatures or sufficient time for diffusion at low temperatures promote DSA. Looking at the influence of strain rate, it is found that there is a negative strain rate sensitivity (nSRS) in AlMg alloys, which leads to a decrease in the amount of stress when the strain rate is increased, which is not typical for metallic materials. The literature shows that there is a change at which strain rate the effect occurs, i.e. a transition from positive to negative sensitivity to strain rate, and that this change is also related to temperature. This can be confirmed by studies [44] in which the effect occurred at room temperature at a strain rate of 10-5 s^{-1}, while at a temperature increased to 50 °C, the PLC effect occurred at a strain rate of 10-4 s^{-1}.

This shows the influence of temperature on the strain rate range in which the PLC effect predominates. With a further increase in temperature and an increase in strain rate, the transition from a positive to a negative strain rate sensitivity was only observed at the highest strain rates of 10^{-1} s^{-1}. The results of investigation [12] carried out on AlMg alloy with 4.55% Mg at different temperatures from 20 °C to 150 °C and different strain rates from $1x10^{-1}$ to $1x10^{-5}$ s^{-1} also show the influence of temperature and strain rate to the occurrence of the effect. At room temperature and different strain rates, serrations occur on the hardening curve during the tensile test. It can be seen that the amount of stress decreases as the strain rate increases, i.e. nSRS occurs. It was observed that the amplitude of serrations is high at lower strain rate values, while milder serrations are observed at higher values.

4 Constitutive models of the PLC effect

In order to accurately predict the behaviour of plastic flow of metal materials during deformation, studies based on the creation of constitutive models that can describe the effects of deformation rate, deformation temperature, deformation and strain rate on the behaviour of the material during deformation are increasingly being carried out. However, due to the non-linear characteristics of material behaviour during plastic deformation, it is difficult to fully describe the influence of all deformation parameters with a constitutive model. For this reason, research on the deformation behaviour of materials focuses on the creation of new constitutive models or the modification of existing constitutive models using data from macroscopic and microscopic tests. The nonlinear behaviour during the plastic deformation of different metals or alloys under many influencing factors takes into account the particular influence of the parameters of the deformation process [48].

The occurrence of the PLC effect is very complex and depends on numerous influencing factors, which imposes the need for further research on this phenomenon in order to better understand the phenomenon itself and the parameters at which the effect could be avoided during plastic processing. Previous research has presented certain constitutive models [48, 49] which attempted to represent the complex thermomechanical behaviour of materials in which the PLC effect occurs, but the results are still far from satisfactory. Of interest are the proposed models that have attempted to represent the behaviour and prediction of PLC bands [50]. The proposed model was based on the ABBM model of avalanche dynamics (Alessandro-Beatrice-Bertotti-Montorsi model of avalanche dynamics). The results of this study show that mobile dislocations interact to generate avalanches of deformations in metal alloys, the necessary conditions being the values of temperature and strain rate within a certain frame in which a PLC band of type A is observed. In view of this, avalanches follow the principle of the ABBM model [51], and the stopping of the bands is random and depends on the local, heterogeneous properties of the material. In the study [12], a model was proposed that shows the prediction of the PLC domain as a function of strain rate and temperature. The proposed constitutive model is based on the mechanical threshold stress (MTS model) and is used to calculate the stress-strain response at different strain rates and temperatures. The modified MTS model captures the dependence of strain rate sensitivity (SRS) on strain, temperature and strain rate within and outside the domain of the PLC effect. The mentioned models are among the newer ones and the simulations relatively coincide with the experiments, which can be seen as a major advance in this field. In addition, both models open up new directions for studying the effect.

5 Conclusion

This comprehensive review of the existing literature on the Portevin-Le Chatelier phenomenon illustrates the complex and multi-layered nature of the PLC effect and shows considerable progress in understanding the underlying mechanisms. However, despite this progress, there are still significant gaps in the understanding of the interplay between microscopic mechanisms and macroscopic deformation behaviour during dynamic strain aging. In particular, the conditions under which the PLC effect transitions between different types of bands, as well as its dependence on strain rate and temperature variations, are not yet fully understood. Therefore, future research should focus on understanding the effects of dynamic strain aging under different processing parameters, including the conditions under which the PLC effect does not occur, in order to establish a clearer boundary for its occurrence. In addition, efforts should be directed towards the development of a comprehensive model that can accurately describe the formation and evolution of PLC bands, taking into account both experimental observations and theoretical findings.

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