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Fractal Analysis of Strain-Induced Microstructures in Metals

Ricardo Fernández, Gaspar González-Doncel and Gerardo Garcés

Abstract

The deformation of materials is a key topic for different industrial sectors. The correlation between specific thermomechanical processes, like extrusion, rolling or additive manufacturing, and the resultant material's microstructure, is particularly interesting. In these thermomechanical processes, the microstructure of the materials depends mainly on the applied stress, the magnitude of strain achieved in a given time period, and the temperature. In the case of metals and alloys, plastic deformation can be described microscopically based on the dynamics of a huge population of moving dislocations. Plasticity is characterized at the mesoscale by the avalanche-like collective behavior of dislocations, which is a typical case of self-organized systems. Dislocations are organized into cells named grains or subgrains that greatly influence the mechanical behavior of metals and alloys. The existence of these fractal structures of dislocations in metals is well established. However, it is very complex to conduct a fractal analysis of these microstructures. This is actually done by looking for a compromise between a detailed description of the complex fractal microstructure and the development of a practical procedure that avoids unaffordable extensive characterization in a different time and spatial scales. Several cases will be described considering different alloys and experimental conditions.

Keywords: fractal, metal, strain, dislocations, mesoscale

1. Introduction

The concept of fractal refers to structures whose appearance is independent of the scale at which it is observed. In these geometrical objects, the same pattern appears regardless of whether we move away to have a more global vision of the observed structure or if we approach to expand a particular detail of it. Fractals are very common in many aspects of nature. They appear, for example, in the crystal-line structure that develops in snowflakes or in the crystals that form frost during cold and wet sunrises. The rough, superficial structure of broccoli is another recurring example in living beings where a fractal nature is revealed. The structure of the veins through which the sage passes on the leaves of the trees is another very clear example of a fractal. The coastline can be described, in some cases, in the frame of a fractal object. Fractals appear not only in nature and are not restricted to physical objects, but they are also present in other fields, such in financial markets; the repeated and cyclic behavior of the stock market also falls in a fractal pattern. Music is other field in which fractals have been reported, for example, when shorter passages are reflected in expanded form in longer passages. It is surprising that,

being fractals so common objects and present in so many fields, the concept of fractal was initially proposed as late as in 1975 [1]. This “self-similarity” of fractal geometry was proposed by the IBM mathematician Benoît Mandelbrot (the term fractal derives from the Latin word *fractus*).

The widespread appearance of the concept of fractal in many areas is also observed in different microstructural features of materials. In fact, the appearance of fractal structures in metals is widely accepted [2]. Solid state metals have an orderly crystalline structure that provides their physical properties. The strongest metal bond is formed when the metallic atoms are ordered as close as possible, usually in cubic or hexagonal lattices. These lattices are described by the unit elemental cell characterized by the lattice parameter, dimension measured in nm and their symmetry typically represented by cubic or hexagonal unit cells. Most of the metals we find in our daily lives are polycrystals. This means that not all the unit elemental cells of the material have got the same orientation with respect each other. Actually, there are domains of atoms/unit cells where their orientation is the same. These domains, which are present in millions in a metallic component, are called grains. These grains are separated from each other by grain boundaries. All this microstructural complexity is a fertile breeding ground for fractal structure appearance. One example is the presence of fractals in the solidification/crystallization structure generated in the manufacturing process of polycrystalline metals during cooling [3]. Another example of fractal structures in metals is provided by the distribution of reinforcement in discontinuously reinforced composite materials. The physical properties of these composite materials are governed by the properties of the clusters that are formed when contacts are established among the reinforcing particles. The percolation of the system greatly modifies its properties. The formation of these clusters depends on the fractional content of the composite material [4]. Another classic example of fractal structures in metals is the fracture surface [5]. These fracture surfaces are generated from the interaction of macroscopic defects, mainly cracks due to the application of mechanical stresses that generate a progressive deformation of the material until its breakage. These surfaces are formed by facets of different sizes and heights that can be accurately described by a fractal. The knowledge of the fractal nature of fracture surfaces has allowed the design of fractal surfaces to achieve certain properties such as a better air flow avoiding turbulence [6].

The fractal nature of the fracture surfaces is an indication that the deformation process generates fractal structures in the deformed microstructure, which ultimately give rise to the fractal fracture surface. In the case of metals, the deformation process begins with an elastic deformation that occurs in small intervals typically smaller well below 0.2% strain. The stress value achieved at this point is known as the yield stress, and it has been agreed that it reflects the beginning of plastic deformation of metals in the macroscale (homogeneous deformation). From this strain/stress value, the deformation is mainly caused by the movement of dislocations. The main defects in metals are vacancies and dislocations. These defects correspond to the lack of a semi-plane of atoms in its crystalline structure, respectively. When the temperature is high and the applied stress is low, this movement can be controlled by the motion of vacancies (diffusion). Dislocations are found not only inside the polycrystalline grains but also at the grain boundaries. A characteristic of dislocations, the fundamental element in metal deformation, is that their collective arrangement has a fractal nature. Sevillano et al. [7] were the first to recognize the possibility of fractal geometry of the cell structure based on the measurement of the fractal dimension of the microstructure of cold worked Cu. These dislocations are organized during the deformation process and form cell structures that have a fractal nature [8]. Moreover, the tangled

arrangement of dislocations in metals has been proposed to be associated with a fractal structure. Examples are found in various usual metal forming processes such as rolling [9].

It is important to understand the role of the collective motion and interaction of a large number of dislocations in the generation of fractal structures [10]. The sample size is one of the features that determine the fractal dimension of these dislocation structures [11]. In that study, carried out in iron micropillars, it has been observed that the fractal dimension of the dislocation structure changes approximately 40%, from 1.1 to 1.5, when the sample diameter increases from 300 to 1500 nm. This reveals the strong size effect on fractal substructures of dislocations in metals.

2. Origin of fractal structures in metals: stochastic vs. thermodynamic approach

Materials, in particular metals, can be considered complex systems from the mechanical point of view, considering that they contain a large number of elements and dislocations that interact with each other. A complex system has properties that arise from the interaction between its elements and the system itself with the environment. In particular, one of these properties is the emergency. The emergency can be defined as the generation of spontaneous patterns resulting from the self-organization of the elements of the system. Classic examples of an emergency in nature are snowflakes. These patterns have different geometries with a high degree of symmetry indicating the ability to organize the system. Surprisingly, a similar behavior has been found in the case of gallium-based liquid metals. By applying an electric potential to this liquid metal, snowflake-shaped fractals have been found [12]. These structures have encouraged the introduction of the term fractalized metals. It has been proposed that this concept defines a new state in which metals can be found. In this state, all positions in the metal lattice are coordinated granting properties such as chiral transport [13].

Metal microstructures have a great amount of defects. Dislocations, linear defects, are the most important ones from the point of view of plastic deformation and mechanical behavior. An individual, isolated, dislocation has an associated elastic stress field. For an edge dislocation, is described, in Cartesian coordinates, by a tensor where the components, σ_{ij} , are given by Eq. (1) [14].

$$\begin{aligned}\sigma_{xx} &= -\frac{Gby(3x^2 + y^2)}{2\pi(1 - \nu)(x^2 + y^2)^2}; \sigma_{yy} = -\frac{Gby(x^2 + y^2)}{2\pi(1 - \nu)(x^2 + y^2)^2}; \sigma_{xy} = \sigma_{yx} \\ &= \frac{Gbx(x^2 - y^2)}{2\pi(1 - \nu)(x^2 + y^2)^2}; \sigma_{zz} = \nu(\sigma_{xx} + \sigma_{yy}); \sigma_{xz} = \sigma_{zx} = \sigma_{zy} = \sigma_{yz} = 0\end{aligned}\quad (1)$$

where G is the shear modulus, b is the Burgers vector, ν is Poisson's ratio, and x , y , and z are the coordinates, perpendicular in-plane, perpendicular out-of-plane, and parallel directions, with respect to the dislocation line, respectively, in the reference system of the dislocation.

The specific stress field depends on the character of the dislocation, i.e., screw or edge. The stress fields of a huge amount of dislocations, in combination with the external applied stress, play a relevant role in the formation of fractal deformation patterns in metals. Through these stress fields, interactions occur with other dislocations or with second phases of the material, such as precipitates. The interaction among typically 10^{10} – 10^{14} m⁻² dislocations generates fractal patterns in the

mesoscale. The formation of these fractals follows the same fundamental rules as any other physical system which develops a fractal structure. Here, it is particularized to the specific characteristics of metals. The formation of fractals in physical systems can be explained from a stochastic or a thermodynamic point of view. The concept that represents the susceptibility of uniform systems to spontaneously develop patterns in uniform nonlinear systems is known as Turing instability [15]. These patterns are found in reaction–diffusion systems describing, for example, the transformation of substances by chemical reactions in which the substances are transformed among each other and spread out over by diffusion. In the case of metals, a reaction–diffusion model of coupled nonlinear equations has been used to describe the formation of forest (immobile) and gliding (mobile) dislocation densities in the presence of cyclic loading [16]. This description is done assuming that a metal undergoing both an internal and external stress is far from equilibrium. The second law of thermodynamics, states that any physical system can spontaneously increase its entropy, i.e., from order to disorder. The apparent conflict between this postulate and the generation of ordered fractal patterns is dismissed by considering that the accompanying decrease in entropy (i.e., ordering) is compensated by a corresponding increase somewhere else, so that the net entropy always increases. However, very recent theories [17] postulate that the apparent conflict between the second law of thermodynamics and ordered systems evolution is due to the consideration that the Universe is near equilibrium. This new theory postulates that there is an energy/matter flow comprising the whole Universe that evolves from disorder to order via self-organization. This process must be described by the empirical laws of non-equilibrium thermodynamics [18]. In this situation, a stress gradient in relatively simple non-equilibrium systems, as deformed metals, causes a flux of energy/matter in the system. As a consequence, it emerges a countervailing gradient. These conjugated processes result in spatio-temporal macroscopic order that spontaneously emerges provided that system is driven far away enough from equilibrium [19, 20]. Some authors have recently accepted that the contribution of the macroscopic (or mesoscopic) order, described by the entropy, must be considered for a rigorous description of the mechanical behavior of metals at high temperature [21].

Stochastics models have been also used to justify the appearance of fractal patterns developed during strain of metals. The necessity of a stochastic treatment, specific for fractal structures, for the characterization of mesoscopic dislocation network distributions in metals was established at the very end of the twentieth century [22]. At temperatures below $0.5 T_{\text{melt}}$ (with T_{melt} , the melting temperature), it is considered that there is no time-dependent plasticity (creep). In this range of temperatures, plasticity is explained by the avalanche-like collective behavior of dislocations [23]. The self-organized structures of dislocations generated by this phenomenon are developed at the grain size scale. This can be described as the mesoscale in metals. Natural nondeterministic fractals are self-similar in a statistical sense over a wide range of scales [1]. The localization of the plastic deformation of metals and alloys into slip bands has been investigated for a long time [24]. One example of dislocation patterns developed under uniaxial tensile tests appears with the Lüders band formation in some steels. The appearance of these patterns under uniaxial stress conditions reveals the great importance of dislocations interaction in their formation.

3. Experimental characterization of fractal structures

There are different parameters that can be used to describe a fractal. The most common one is the fractal dimension. Fractal dimensionality is considered a

measure of complexity for systems that do not present integer dimensions [25]. However, there are different fractals that have the same fractal dimension. Therefore, other parameters have been proposed to describe some characteristics of fractals. One of the most important characteristics of fractal structures of dislocations generated during metal deformation is their connectivity. Dislocations are usually arranged in grain and subgrain boundaries. These boundaries maintain their integrity, while the deformation increases due to the stress fields of dislocations that lock them in low-energy configurations. The connectivity of these structures can be described in this context by the distance to next dislocation neighbors. In particular, lacunarity is one of the most used parameters to characterize the connectivity among different parts of a fractal structure. Lacunarity is a measure of spatial heterogeneity, and it is used to differentiate images with similar fractal dimension but different appearances [26]. The local connected fractal dimension represents the connectivity of the different parts of the system by a color map that indicates the proximity of the neighbor pixels in a given image.

3.1 Image analysis

All parameters that describe a fractal structure of dislocations can be calculated from images obtained from the microstructure. In the deformation of metals, the most interesting images are related with the observation of dislocations that can be homogeneously distributed or rearranged to form subgrains or grain boundaries. These dislocation structures have been described in a very detailed manner from decades thanks to transmission electron microscopy (TEM). However, there are some aspects that must be taken into account when analyzing the fractal parameters of dislocation microstructures. The first one is the quality of the images. Normally, metallographical images have a gray range. In the case of a TEM image, moreover, there is a great contrast evolution depending on the interaction between the electrons and defects (dislocations, stacking faults, etc.), the relative orientation of the crystal, and the mode in which the images are obtained (bright field vs. dark field).

Figure 1 shows a TEM bright field image of the microstructure of a magnesium alloy which was deformed at intermediate temperatures. The presence of dislocations is clearly observed. The fractal dimension is typically measured from this kind of images using the box counting method.

The “box counting” is a method for analyzing complex patterns by splitting an image into smaller and smaller pieces, typically “box”-shaped. The fractal dimension is calculated considering the number of boxes containing part of the microstructure vs. the number of empty boxes. The ratio of full vs. empty boxes is considered for at least 4/5 box sizes for the analysis of the fractal dimension. Computer-based box counting algorithms have been applied to patterns in one-, two-, and three-dimensional spaces [27, 28]. The technique is usually implemented in specialized software to analyze patterns extracted from digital images. This software also has application in the determination of related parameters such as lacunarity and multifractal analysis [29].

The TEM photos must be treated to obtain a binary image. However, in this process, TEM images cannot be completely clean of artifacts such as shadows or dots. The presence of these artifacts introduces an overestimation of the fractal dimension.

Figure 2 shows several idealized examples presenting this situation. **Figure 2(a)** shows an idealized grain before deformation. Therefore, no dislocations or subgrain boundaries appear. After deformation, we can calculate the fractal dimension, D_F , of, as an exercise, one or two dislocations within this grain (**Figure 2(b)** and **(c)**, respectively). For these two ideal examples, the fractal dimension calculated with

Figure 1.
Bright field image of a magnesium alloy deformed around 2% in tensile mode at a temperature of 200°C and a strain rate of 10^{-3} s^{-1} . Zone axis $[11\bar{2}0]$ $g = [1\bar{1}00]$.

Figure 2.
(a) Idealized microstructure of a completely dislocation-free grain before plastic deformation. (b–d) Subgrain formation during the deformation process. (e) Subgrain coarsening during deformation process. (f) Microstructure shown in (c) with an artifact.

the plugin FracLac [30] used in FIJI software is very similar (0.96 and 0.99, respectively). Since dislocations are line defects, these values are very reasonable. As the macroscopic strain increases, new dislocations are generated, or, even, subgrains

can be developed. This increase in dislocation density results in an additional increase in the fractal dimension (**Figure 2(d)** and **(e)**). As it is shown in **Figure 2(b)–(e)**, the fractal dimension of dislocation substructures is related to the area covered by dislocations/substructure. Then, the fractal dimension of the structure shown in **Figure 2(e)** formed by thick lines is 10% greater than the structure shown in **Figure 2(b)**. However, there are some elements, mainly dots, which greatly influence the fractal dimension calculated by the box counting technique. As an example, **Figure 2(f)** shows the same structure as **Figure 2(c)**, as far as it is assumed that during the binarization process, the dusty dots are part of the image. These dusty dots are considered as an artifact. As a result of the consideration of the dusty points in the image, the fractal dimension increases around 25%.

There are other aspects that must be taken into account when the fractal dimension of a dislocation structure in a deformed metal is calculated using TEM images. The first one is related to the fact that TEM images of the dislocations can be taken under several 3D orientations of the crystal and/or using different diffraction vectors. Dislocations can be visible only when the scalar product $g \times b$ (where g is the diffraction vector of the crystal that is excited, i.e., Bragg's conditions are attained) is different from zero. **Figure 3** shows an example of a magnesium alloy deformed at room temperature. The Burgers vector in magnesium is $[11\bar{2}0]$. Dislocations are invisible when $g = [0002]$ (**a**) is excited and visible with $g = [10\bar{1}1]$ (**b**).

The second one is that TEM images can be obtained in different modes known as bright and dark fields. **Figure 4** shows the image of a deformed sample of a magnesium alloy obtained in the same area, using the same crystal orientation $B = [1120]$ and the same diffraction vector $g = [1011]$. The difference between both images is that image in **Figure 4(a)** was obtained in dark field mode (image is generated with a diffracted spot) (weak beam), while image in **Figure 4(b)** was obtained in bright field mode (image is generated with the transmitted spot).

Within the magnesium grain, different defects have been created during plastic deformation: dislocations, twins, and subgrains. Twins are clearly observed in bright field mode (**Figure 4b**), and dislocations and subgrains are clearly observed in dark field mode. Therefore, the fractal dimension of the complete deformed substructure changes depending on the TEM mode selected. In general, a combination of images must be used for a complete description of the microstructure. However, there are mechanical tests conducted under specific experimental

Figure 3.
 TEM image of a magnesium alloy deformed at 3% at room temperature and a strain rate of 10^{-3} s^{-1} . Zone axis $[11\bar{2}0]$ (**a**) $[0002]$ and (**b**) $g = [10\bar{1}1]$.

Figure 4. TEM image of a magnesium alloy deformed at 3% at room temperature and a strain rate of 10^{-3} s^{-1} . Zone axis $[11\bar{2}0]$ $g = [1\bar{1}01]$. (a) Dark field and (b) bright field.

conditions where images from one single mode, bright or dark, are enough for a complete description of the microstructure. In particular, after mechanical testing conducted at high temperature, the dislocation substructure is easily observed in bright field mode because the lattice distortion is almost eliminated.

Finally, it is interesting to point out that TEM images have a 2D nature, whereas dislocations, twins, subgrains, and, in general, the crystal structure have a 3D nature. Cui and Ghoniem [13] studied the influence of size on the fractal dimension of dislocation structure throughout dislocation dynamics simulations. The analysis showed that the D_F of the 3D structure is significantly smaller than the D_F of the 2D corresponding to the projected dislocations in all considered sizes. **Figure 5** shows two images of a 3% deformed magnesium alloy at room temperature of the same area

Figure 5. Bright field image of a magnesium alloy deformed at 3% of plastic strain at room temperature. (a) Sample fully parallel to the beam and (b) sample tilted with respect to the previous condition.

tilted at two different angles. In the first case (**Figure 5a**), a stacking fault is observed, and it is fully parallel to the beam. Therefore, atomic lines are clearly observed. When the sample is tilted (**Figure 5b**), it is possible to distinguish the stacking fault throughout the thickness of the TEM sample. Again, the fractal dimension changes.

4. Temperature effect on dislocation fractal structures

The mechanical properties of metals are highly influenced by temperature. In general, the mechanical strength decreases significantly, and the ductility and plastic deformation increases with temperature. In addition to these effects, when $T > 0.5T_{\text{melt}}$, time-dependent plasticity becomes dominant [31]. This time-dependent deformation is also called creep. In this temperature range, the dislocations increase their mobility due to the activation of the climbing mechanism, in particular when the stacking fault energy is high. The activation of dislocation climbing is revealed in the appearance of well-ordered dislocation structures generated during deformation.

In the following section, some examples of dislocation structures generated at room temperature and high temperature (573 K) will be shown. In the second case, the temperature effect on the arrangement of dislocations to form subgrain boundaries will be observed.

Figure 6.
 Bright field image of tangled dislocations in pure aluminum deformed up to 22.3% of plastic strain at room temperature at 10^{-4} s^{-1} .

4.1 Low-temperature dislocation structures

In **Figure 6**, a bright field TEM image of a commercially pure aluminum single crystal strained up to 22.3% at room temperature at 10^{-4}s^{-1} strain rate is shown. This figure corresponds to a perpendicular section of the sample with respect to the tensile axis. Dislocations are arranged in a messy way. The subgrain size developed by deformation of this pure metal at room temperature is around 1 micron.

4.2 High-temperature dislocation structures

In **Figure 7**, a detail of a subgrain boundary shows a bright field TEM image of a commercially pure polycrystalline aluminum strained around 2% at 573 K and 29 MPa. **Figure 7** corresponds also to a perpendicular section of the sample with respect to the tensile axis. The dislocations are well-ordered in the subgrain boundary. The subgrain size developed by deformation of this pure metal at room 573 K and 29 MPa is around 5 microns.

4.3 Age-hardenable alloys

A very important family of metals is that formed by age-hardenable alloys. Their properties can be tuned by means of heat treatments that usually start with a quenching process of the sample from above the solution temperature. The quenching step is followed by an isothermal annealing that typically lasts for some hours. The variation of mechanical properties is related with the precipitation state developed by the heat treatment. Therefore, deformation that takes place at high temperature is accompanied by an evolution of the precipitation state. The two phenomena are coupled. The deformation slows down due to the presence of atoms in solid solution and coherent precipitates, and the precipitation kinetics is modified

Figure 7.
Bright field image of a subgrain boundary of polycrystalline pure aluminum deformed around 2% at 573 K and 29 MPa.

Figure 8.
 Back scattered image of a AA2014 powder particle showing the precipitate structure after quenching from 800 K, followed by aging at 523 K for 10 h.

by the presence of the dislocation structure [32]. **Figure 8** shows the structure of precipitates in aged AA2014.

5. Conclusions

The formation of dislocation fractal structures with plastic deformation is, as in many others aspects of nature, a widespread phenomenon. It is, in fact, observed under different tensing conditions and metal alloys. This is because metal plasticity occurs on several temporal and spatial scales.

On the temporal scale, the dislocations can move at speeds greater than that of the sound when the dislocation density is low, leading to rapid plastic deformation events (e.g., car accident). On the contrary, deformation under creep conditions can occur in much longer time periods, as long as days, months, or even years (e.g., motion of glaciers). The same concept can be applied to the spatial scale as deformation phenomenon involves entities as small as dislocations and vacancies in the crystal lattice, and, on the other side, this phenomenon takes place at a macroscopic scale, meters in size in large components.

The behavior of dislocations during strain of metals is very rich and complex because the plasticity phenomenon covers different spatial and temporal scales and there is a very broad range of experimental conditions and alloys. Nonetheless, the existence of fractal structures of dislocations is ubiquitously found when a metal is deformed. This fact greatly encourages the use of fractals to fully describe the phenomenon of plasticity in metals.

The determination of the fractal dimension of dislocation structures, D_F , is conducted from images obtained by different microstructural characterization methods, typically, by TEM. These images must, then, be carefully treated to minimize inaccuracies in the determination of D_F . The box counting technique is revealed to be a very suitable and reliable method to determine D_F . Although the process is automated in some image analysis applications, the presence of elements different from dislocations (e.g., dots related with other microstructural features) can greatly modify the fractal dimension.

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Conflict of interest

The authors declare no conflict of interest.

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