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NUCLEATION AND GROWTH OF TWIN INTERFACES IN FCC METALS AND ALLOYS

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Abstract. Presence of coherent interfaces within crystallites, referred to as «annealing twins,» has many effects on the properties of several fcc metals and alloys. The mechanism of the formation of these twins have been subject of numerous studies over fifty years. Although much progress has been made in recent years, a universally accepted view on the formation of these twins has not yet emerged. This brief review will describe recent studies at Naval Research Laboratory to understand several aspects of the annealing twins including nature of these twins, the mechanism of their formation, and the compositional parameters which affect their densities. These studies have established a relation between twin density and grain size, temperature and material properties. A model of the mechanism of their formation based on the emergence of Shockley partial loops on consecutive {111} planes during grain migration has also been developed. It is argued that various experimental and theoretical results obtained over the years can be consistently and satisfactorily explained by this model. This study has analyzed two aspects of annealing twins in details viz, a) role of boron additions in reducing twin density in nickel and b) the effect of annealing twins in Hall-Petch relation.

1. INTRODUCTION

Annealing twins in various fcc (face centered cubic) metals and alloys have been observed as early as 1897 when they were seen in gold nuggets by Liversidge [1]. This was followed by the observation of Eving and Rosenthal [2] in 1901 that cast materials showed a scarcity of annealing of twins. A detailed discussion on the nature and formation of annealing twins was first provided by Carpenter and Tamura [3]. The frequency and occurrence of annealing twins has been shown by previous workers to be controlled by grain boundary energy [4,5], prior deformation [6-8], twin boundary energy (stacking fault energy) [6,9] and grain size [6,7,10] (for a review see [11]).

Twinning affects properties of a variety of material, including such technologically important material as superalloys, whose fatigue behavior at high temperature is drastically affected by the presence of twin interfaces. The role of twinning in recrystallization and grain growth is well documented. For example Gertsman *et al* [12] have recently concluded that evolving grain boundary distribution could only be due to a result of evolving multiple annealing twins. In view of their importance annealing twins have been extensively studied both theoretically and experimentally. Several attempts have been made in the past to explain the mechanism of nucleation and growth of annealing twins and a number of models have also been proposed for their formation. These models are usually classified into three different groups depending on the underlying concepts and involve (i) growth accidents [4,13-15], (ii) grain encounters [14-16] and (iii) nucleation of twins by stacking faults or fault packets [19,20]. In the growth accident model, a coherent twin boundary forms at a migrating grain boundary due to faulting on {111} facets during grain growth [14]. It is assumed in the second group of models that twin forms when suitably oriented grain meets. Finally, in the third group twins form a migrating boundary such that one of the pseudo-coherent interfaces remains attached to the boundary but there has been no single model which is as yet universally accepted. The important factors determining twinning frequency during grain growth are:

1) grain size D,
2) temperature T and time of annealing t,
3) velocity of grain boundary migration,
4) grain boundary energy,
5) twin boundary energy (or stacking fault energy).

Other factors often not considered carefully are:

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1) texture,
2) prestrain or prior deformation,
3) inclusions.

There are very few systems and studies where all these factors have been taken into account. Pande, Imam and Rath [21], have carried out a systematic study of the formation of annealing twins in nickel as a function of annealing conditions. Based on these observations they have proposed three basic rules for the development of annealing twins. These are:

1) twins nucleate at grain boundaries during their migration steps,
2) number of twins produced is proportional to the distance of grain boundary migration,
3) number of twins produced is also proportional to driving force for migration.

It was shown that these postulates lead to a simple relation between twin density, p, and grain size, D, as:

$$p = \frac{B}{D}\log\left(\frac{D}{D_0}\right), \qquad (1)$$

where p=Number of twin interfaces/unit length, B=A constant and D_0= grain size at which p=0.

This simple relation agreed well with the microstructural evolution in Ni. The constant B is material dependent and should depend strongly on stacking fault energy of the system.

2. ROLE OF BORON

One obvious question in the study of annealing twins is the role played by twin boundary energy and stacking fault energy (the two energies are usually related). Since we did not measure these parameters we do not know their exact role. They can in principle also be introduced in the theory, preferably by a microscopic theory of twin formation. A test of our mechanism would be as follows: The critical assumption in the model is that any increment in twin density is proportional to the driving force of the migrating boundary. This result can be tested by changing the driving force without significantly affecting the other pertinent parameters such as grain size. It is known [10] that the driving force of the migrating boundaries can be substantially changed by the addition of impurities. Twin densities then should be dependent on trace impurities. We have therefore studied extensively the role played by small additions of Boron in the formation of annealing twins. These results are presented in this paper. Preliminary analysis indicate the results are also consistent with our understanding of the formation of these twins.

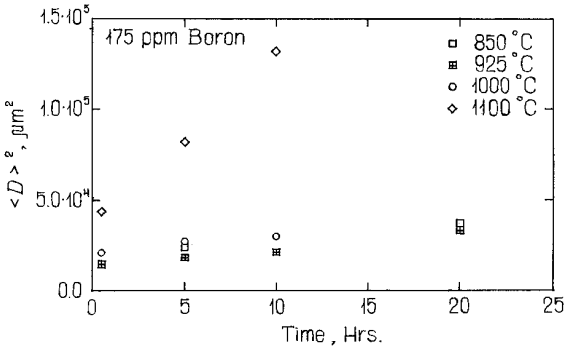

Fig. 1. A plot of square of grain size as a function of time at constant annealing temperature.

Nickel, nominally 99.9 pet pure, was used in this investigation. Boron doped nickel samples were made by arc melting. Samples containing 175, 241, 385 and 601 ppm boron were prepared. These samples were given the appropriate thermomechanical treatment sequence before final annealing These heat treatments were carried out under vacuum (10^{-5} torr or better) at temperatures ranging from 750 °C to 1200 °C and time ranging from 2 minutes to 100 hours. Metallographic parameters, such as grain size, twin density, and twin width, were evaluated using linear analysis based on Smith and Guttman's procedure [22].

Measurement of twin frequency (p) and grain size (D), was performed as follows: number of intersections of a random line of unit length with a two dimensional feature in a three dimensional structure is exactly half the surface to volume ratio, *i.e* if

L = total length traversed,
G = total number of grain boundaries intercepted,
M = number of twin boundaries intercepted, then:
grain size $D = L/G$,
twin frequency $p = M/L$, and
number of twins/grain $N = D\,M/L = L/G\,M/L = M/G$ (2)
(if grain shapes do not change).

Average grain sizes D were measured as a function of time and temperature, for both pure Nickel and also for Boron doped Nickel. These results are plotted in Fig. 1 as D^2 vs. time t. It is seen that following relation between D and t is obeyed closely:

$$D^2 - D_1^2 = Kt, \qquad (3)$$

where K and D_1^2 are constants and can be obtained from the slope and the intercept of the straight line in Fig. 1. Similar results were obtained for boron contents.

It is also seen that the slope depends on the temperature of annealing. Theory indicates that K should be related to temperature by the following relation:

$$K = K_0 \exp\left(-\frac{Q}{RT}\right), \qquad (4)$$

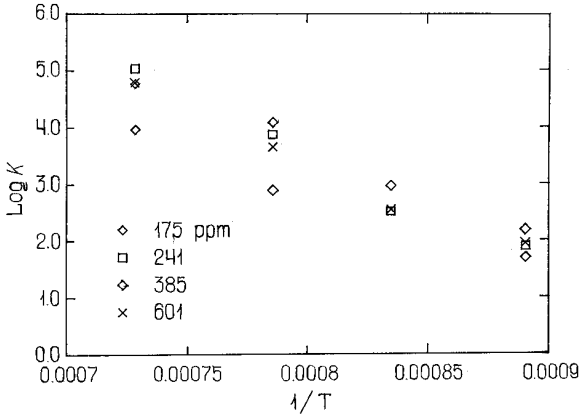

Fig. 2. A plot of log$K$ (obtained from Fig.1) as a function of inverse of temperature in Kelvin.

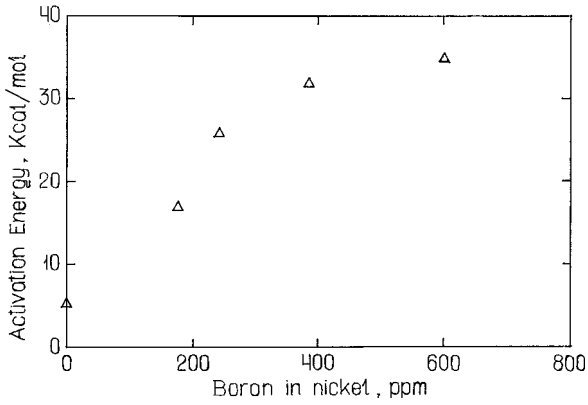

Fig. 3. A plot of activation energy as a function of boron content in nickel.

where Q is the activation of the process, R is the gas constant, and K_0 is a constant. If this relation is true, a plot of log K vs. $1/T$ should be a straight line, and the activation energy Q can be obtained from the slope of this line.

In Fig. 2 we have plotted log K as a function of $1/T$ for pure Nickel as well as for Boron doped Nickel. A straight line is obtained in all cases. From the slope we have obtained activation energy for this process for Boron doped Ni for several values of Boron content. In Fig. 3 we plot the activation energy Q as a function of the Boron content in the nickel. It is seen that activation energy increases with increasing Boron content in a smooth fashion, indicating a gradual increase in Q, and gradual slowing down of the grain boundary migration with increasing Boron content. Fig. 4 shows a plot of N, the number of twins/grain as a function of grain size for several Boron content. It is seen that N is not a monotonically increasing function of grain size. Instead N dips for a grain size of about 200 mm, and then either increases or stays roughly level. A similar situation is seen in the plot of N vs Boron content (Fig. 5), for several temperatures of annealing. N dips to a low value for a Boron content of about 200 ppm, then increases or stays level. The two curves N vs Boron content, and N vs grain size, appear to be similar. It should be noted that the Eq. 1 is still obeyed approximately as can be seen from Fig. 5.

The results reported here, provides further insight for the mechanism of growth of annealing twins, as it relates to the growth of twins with the migration of grain boundaries. The presence of Boron in Nickel slows down the migration of the boundaries, as seen by the smooth increase in activation energies with Boron content (see Fig. 3). But this is not sufficient to explain our results. An additional effect is present, as seen from the fact the N is minimum for a certain grain size or boron content (see Fig. 4 and Fig. 5) respectively. It is not yet clear what precisely this additional effect is. Transmission electron microscopy revealed the presence of some Boron-rich precipitates near the grain boundaries, for higher Boron content specimens. One way to reconcile our results present here, with our previous model is to assume that Boron not only slows down the grain boundary migration but also, prevents the migration of steps on the grain boundaries from migrating, thereby preventing the nucleation of twins. Such poisoning of twin nucleation does not need a large Boron content, and indeed it might saturate for a small Boron content, sufficient to cover a nanolayer of the grain boundary. Nucleation of a Boron rich precipitates may interfere with the poisoning of steps at higher Boron content. This explanation is necessarily tentative and needs further experimental and theoretical investigation.

In summary, we found that small additions of boron slows down grain growth, though kinetics of growth remain approximately parabolic. There is a drastic reduction in twin density for a specific Boron content of ~200 ppm. Activation energy of grain growth increases with increasing boron content. The present results are all consistent with the authors model of annealing twin formation.

3. EFFECT ON HALL-PETCH RELATION

Our studies indicate that twins multiply during grain growth and therefore could have measurable effect on mechanical properties including Hall-Petch relation. Since Hall and Petch first correlated yield strength with the inverse square root of grain diameter, D, in mild steel, the classical Hall-Petch relationship has been used for several decades to describe the effect of grain size on the yield point [23-25], or flow stress [25-29] of a polycrystalline materials, as:

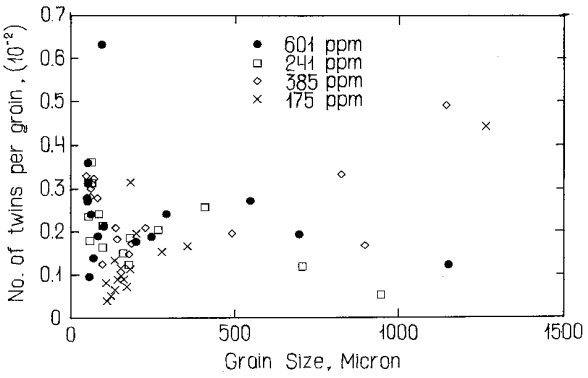

Fig. 4. A plot of number of twins per grain as a function of grain size for several boron contents in nickel.

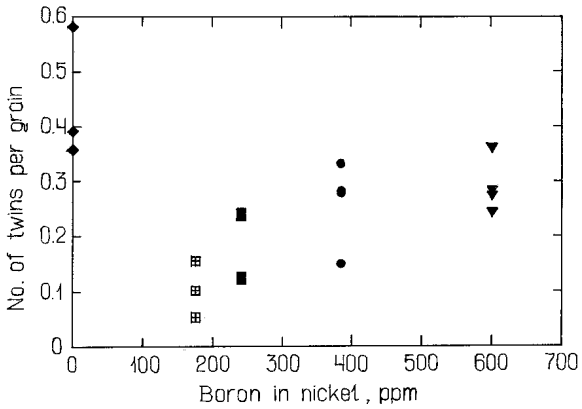

Fig. 5. A plot of number of twins as a function of boron content.

$$\sigma = \sigma_0 + kD^{-1/2}. \quad (5)$$

Where σ is the yield stress, σ_0 is often identified with "friction stress" needed to move individual dislocations during deformation, k is a constant often referred to as Hall-Petch slope and is material dependent, and *D* is average grain size. Two recent compilations [30, 31] has indicated that this relations is fairly well obeyed for large grain sizes ($D \geq 1$mm) with the grain size exponent close to -1/2 for a wide range of grain sizes and materials. On the other hand, the value of σ_0 appears to vary from study to study even for the same material. Further, in most of these studies the role of twin boundaries have been overlooked.

There are a number of models which have been proposed to account for a grain size dependence of the stress, σ, in Eq. 5; most of which can be rationalized in terms of a dislocation pile up model. These are reviewed in detail by Li and Chou [32]. In deriving Hall-Petch relation, the role of grain boundaries as a barrier to dislocation model is considered in various models. In one type of model [23,24,26] grain boundary acts as a barrier to pile up of dislocations, causing stresses to concentrate and activating dislocation sources in the neighboring grains, thus initiating slip from grain to grain. In the other type of models [27,33,34] the grain boundaries are regarded as dislocations barriers limiting the mean free path of the dislocations, thereby increasing strain hardening, resulting in a Hall-Petch type relation. In both types of models, a twin interface can act in the role of grain boundary, and thus should be included in calculating Hall-Petch type relation. Questions however remains unresolved, whether the presence of twin boundaries modify Hall-Petch relation in any Fashion. Specifically do they have any effect on the Hall-Petch slope as well as on the "friction stress" σ_0 ? This paper provides an insight to these questions. For this purpose, we utilize the results of our studies [34-36] of annealing twins in fcc materials. Specifically we utilize a relation between twin density (number of twin/grain) and grain size.

The basic assumption in our calculation is that twin boundaries act as a barrier to dislocation motion. There are several studies showing twin boundaries acting as a slip barriers. These results have been summarized by Remy [37]. Especially convincing is a remarkable electron micrograph [38] showing strain relaxation at the interaction of slip dislocations with a coherent twin boundary. Impinging matrix dislocations behind the impinging dislocations form a pile up; thus indicating that the coherent grain boundary acts as a barrier, though its strength as a barrier may not be as effective as for a grain boundary, In case of annealing twin, it has been possible to identify the dislocation reaction occurring at the twin interfaces leading to dissociation of the dislocation according to the following reaction [37]:

$$1/2[110] \rightarrow 1/2[101]_T + 1/6[\bar{2}\bar{1}\bar{1}],$$

where 1/2[110] is the Burgers vector of impinging dislocation at the twin interface and the right hand side of the reaction denotes the two dissociated dislocations. The reaction corresponds to an increase of 33% in energy [37] and is therefore not expected to be favored. Yet this has been observed experimentally using transmission electron microscopy. A similar reaction was also proposed by Pond and Smith [39] but on a [121] facet of annealing twin in aluminum.

Remy [38] in his review of interaction between slip and twinning systems has pointed out that for both deformation and annealing twins, coherent boundaries can be considered to be "fairly strong obstacles" to dislocations. The dislocations can ultimately move into an obstacle twin by dissociating, but it requires an energetically unfavorable dislocation reaction and hence need a stress from piling dislocations. These results are also borne out by the experiments of Miura and Saeki [40] on twin related bicrystals. Hence we

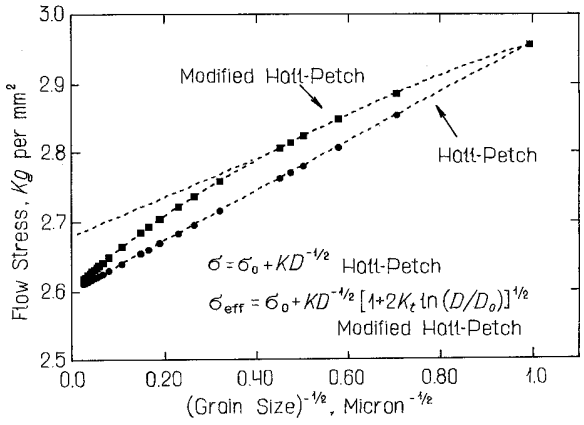

Fig. 6. A plot of effective flow stress with grain size showing departure from linearity (σ_0=2.6; K=0.36, K_t=0.2, D_0=1 micron).

assume twin interfaces acts as a barrier and calculate the effective grain size. We make use of the relation derived before [34-36] that the number of twins per grain (N) is approximately given by:

$$N = K_t \ln\left(\frac{D}{D_0}\right), \tag{6}$$

where D_0 is the grain size at which N=0; otherwise D_0 is related to initial twin density and K_t is a constant.

If there are "n" grains in a given cord length "L", then the

number of grain boundary intersections in the length $L = n+1 \sim n$
and grain size without twins, D, $= L/n$
Number of twin boundaries/grain $= 2K_t \ln(D/D_0)$,

Using Eq. 2, therefore,

total number of intersections $= n+n2K_t \ln(D/D_0)$
and effective grain size, $D_{eff} = L/[n + n2K_t \ln(D/D_0)]$
$= L/n[1 + 2K_t \ln(D/D_0)]$

Thus,

$$D_{eff} = D/[1 + 2K_t \ln(D/D_0)]. \tag{7}$$

In the above calculation we have made the assumption, that twin interface is as effective a barrier as a grain boundary. In practice it is unlikely to be so, because of the reason given before i.e. twin interface does allow partial transmission of piling dislocations. If this is accepted, the above estimate of D_{eff} might be an over estimate. If we substitute this effective grain size in the Hall-Petch relation, Eq. 5, we get

$$\sigma_{eff} = \sigma_0 + kD^{-1/2}\left[1 + 2K_t \ln\left(\frac{D}{D_0}\right)\right]^{1/2}. \tag{8}$$

This is the modified Hall-Petch relation, taking into account twin interfaces in the material. It is seen that in the traditional Hall-Petch plot of σ *vs* $D^{1/2}$, the slope is now modified to $k[1+2K_t \ln(D/D_0)]^{1/2}$ and hence the plot should no longer be linear (see Fig. 6), the departure from linearity depends on the value of grain size and K_t. In the figure the typical number for D_0 and K_t are taken from our studies in nickel [21]. As expected for grain size ~D_0, there is no departure from linearity. For very large grain sizes $\sigma \sim \sigma_0$, since $D^{-1/2}$ term is dominant, and hence the effect of twins is again small leading to the curved shape of the Hall-Petch plot as shown in Fig. 6. An interesting consequence of the curved shape is that a definitions of the σ_0 and Hall-Petch slope now becomes ambiguous, because σ_0 is usually obtained by the intercept on the s axis of the linear curve. This may explain why the quoted σ_0 even for a given material differs from researcher to researcher.

It should be pointed out that there is another model of Hall-Petch due to Li [41,42] which may not need grain boundary as barrier (for pile ups), but rather as a source of dislocations i.e. grain boundary is considered to emit dislocations. The amount of dislocations emitted by a grain is then proportional to its surface area viz $4\pi D^2$ and hence dislocation per unit volume will be proportional to $(4\pi D^2)/(4/3\pi D^3)$. In such a case the density of dislocations emitted will, therefore, depend inversely on grain size. Using the well known linear relation between dislocation density and σ^2, Hall-Petch relation follows. In Li's models grain boundaries act as dislocation source and as such our derivation may not be applicable, since twin interfaces are not known to act as dislocation source. Hence our calculation may provide a simple way to distinguish between dislocation pile ups model and dislocation source model of Hall-Petch relation.

We have recently proposed such a microscopic theory based on the idea that crystallographically annealing and deformation twins are identical [36]. They both consist of stacking fault on consecutive {111} planes. These faults are produced by the glide of Shockley partials. Based on this idea a microscopic model for the formation of annealing twins in fcc crystals was proposed. It was argued that Shockley partial loops nucleate on consecutive {111} planes by growth accidents occurring on migrating {111} steps associated with a moving grain boundary. The higher the velocity of the boundary, higher the twin density. The absence of twin in high stacking fault energy materials and influence of temperature on twin density can be rationalized in terms of the model. This model is consistent with our previous work [21]. Further evidence could in principle be obtained by precise high resolution studies of twin nucleation in fcc materials. This has as yet not been performed.

In conclusion, we have obtained a modified Hall-Petch relation that includes the effect of twin interfaces, which are usually present in fcc materials. Our estimation for nickel [35] indicates, that this modification is significant and thus amenable to experimental verification.

4. SUMMARY

Our recent studies have provided a theoretical framework to understand various aspect of annealing twins in fcc metals and alloys. A microscopic model of nucleation of these twins based on the nucleation of partial dislocations from a moving boundary have also been proposed to reconcile similar earlier models. For the first time we have a relation between twin density and grain size. This relation help us provide an analytical expression again for the first time for the Hall-Petch relation containing annealing twins. We have suggested a means to reduce twin density in metals and alloys by the boron addition. Last two aspects of annealing twins are discussed in detail in this review. Our experimental studies have been mostly confined to nickel and copper but it is expected that our conclusions should be valid for most of the fcc metals and alloys.

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