Modification of the Hall-Petch relationship for submicron-grained fcc metals

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Abstract:

Experimental data show that the conventional Hall-Petch relationship cannot be maintained in its original form for metals having submicrometer structures. We now propose a dislocation model which modifies the Hall-Patch relationship to provide a uniform description of the grain size strengthening of submicron-structured face-centered cubic (f.c.c.) metals and solid solution alloys.

Keywords: f.c.c. metals, grain size strengthening, Hall-Petch relationship, strength of polycrystals, ultrafine-grained structures.

1. Introduction

It was a significant development in materials science when, approximately seventy years ago, in 1951 Eric Hall [1] and in 1953 Norman Petch [2] published the relationship now known as the Hall-Petch equation:

$$\sigma_y = \sigma_0 + A \cdot d^{-1/2}, \tag{1}$$

where this relationship demonstrates that the yield stress, σ_y, necessary for yielding and plastic deformation of polycrystalline materials changes linearly with the reciprocal of the square root of the grain size, *d*. In Eq. (1), σ_0 is the friction stress (or yield stress of a single crystal) and *A* is a positive material constant. This equation, which has become one of the most cited in materials science, demonstrates the basic strengthening mechanism in polycrystalline materials, as the yield stress and then the strength increase with decreasing grain size.

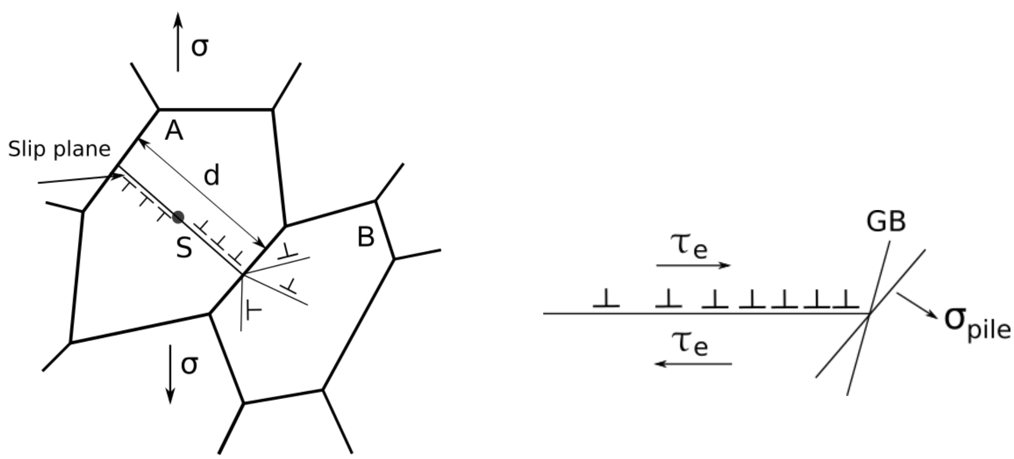
This relationship soon became a driving force of several efforts to obtain finer grain sizes in conventional materials [3-11]. Thus, through various severe plastic deformation (SPD) processes [4-11] in bulk crystalline materials, average grain sizes in the sub-micrometer ($d < 1000$ nm) range were achieved and this significantly increased the strength of these ultrafine-grained materials.

In practice it is important to note that both Hall and Petch developed Eq. (1) phenomenologically, basing their conclusions on experimental data obtained on samples of iron, copper and brass [1-3]. This relationship is well established for polycrystalline metals and alloys having average grain sizes of $d > 1$ μm but more recent studies show that the relationship no longer holds in the range of sub-micron and exceptionally small grain sizes. On the one hand the parameter *A* in Eq. (1) paradoxically decreases in the submicron range [12-14] and on the other hand at grain sizes below approximately 20 nm the Hall-Petch equation is no longer valid since, as the grain size decreases, the yield strength also decreases and the material becomes softer [11,12,14]. This latter behavior is also called the inverse Hall-Petch effect and it has been explained using several different interpretive models [15-17].

The present work is focused on experimental data obtained from submicron-structured materials having grain sizes in the range between 100 and 1000 nm. Our main motivation is to present a dislocation model that provides an appropriate modification of the Hall-Petch relation and produces a uniform description of the large numbers of results now available in the literature for submicron-structured solid solution face-centered cubic (f.c.c.) metals and alloys.

2. Modification of the original Hall-Petch relationship

a) Physical background of the original Hall-Petch equation



a) b)

Figure 1: *Effect of grain size on the yield stress: a) Schematic of the operation of a Frank-Read source (*S*) in a selected grain (*A*) and b) Effect of the corresponding set of pile-up of dislocations at the grain boundary (GB).*

When a polycrystalline specimen is subjected to a gradually increasing load, the critical shear stress is first reached in the slip plane having the most favorable orientation in any selected grain. If there is a Frank-Read source, S, belonging to this slip system, it will emit a series of dislocations as shown schematically in grain A of Fig. 1a. As a result of increasing external forces, the source emits an increasing number of dislocations which then pile up at the grain boundary (GB) as shown in Fig. 1b. In the stress field of the pile-ups, a maximum tensile stress, σ_{pile}, forms in the adjacent grain with a magnitude given by [18]:

$$\sigma_{pile} = n \cdot \tau_e \tag{2}$$

where τ_e is the effective shear stress operating in the slip plane belonging to the source and *n* is the number of dislocations in the pile-up. At a given value of τ_e, due to the back stress of the piled-up dislocations, the operation of the source is halted after the emission of *n* dislocations determined by [18]:

$$n = \frac{\pi(1-v)d}{\mu b}\tau_e \tag{3}$$

where v is Poisson's number, μ is the shear modulus, *b* is the magnitude of the Burgers vector and *d/2* is the size of the pile-up set which can be identified, in the present case, with one-half of the average grain size. From equations (2) and (3):

$$\sigma_{pile} = \frac{\pi(1-v)d}{\mu b}\tau_e^2. \tag{4}$$

This stress may activate multiple slip in grain B (Fig. 1a) after reaching a critical value. The initiation of the macroscopic deformation is related to the value of σ, the external stress, when σ_{pile} reaches a critical value necessary to initiate multiple slip. Considering also Schmid's law, the average critical shear stress acting in each grain, τ_e, is proportional to σ and the yield stress, σ_y based on Eq. (4) can be given as

$$\sigma_y = \sigma_o + K \cdot d^{-1/2}, \tag{5}$$

where *K* is the constant containing the parameters in Eq. (4) as well as the Schmid factor and σ_o is the yield strength of the single crystal. This is the physical background of the Hall-Petch relationship which is basically interpreted by the concept of dislocation pile-ups formed at grain boundaries [11,14]

b) Modification of the Hall-Petch equation

Let us consider again Eq. (4). It follows that normalizing the stresses by the shear modulus and the grain size by the magnitude of the Burgers vector leads to:

$$\frac{\sigma_{pile}}{\mu} = \pi(1-v)\left(\frac{d}{b}\right)\left(\frac{\tau_e}{\mu}\right)^2 \tag{6}$$

Given that Poisson's number is almost the same for all f.c.c metals $(\nu \approx 1/3)$, the dimensionless relationship in Eq. (6) is already appropriate for a unified description of the properties of different f.c.c metals.

For large grains, it may be assumed that macroscopic plastic deformation is observed if the normalized stress, σ_{pile}/μ, reaches a constant value K_1 to initiate multiple slip, for which the normalized effective shear stress is then given by

$$\frac{\tau_e}{\mu} = \sqrt{\frac{K_1}{\pi(1-v)}} \cdot \left(\frac{d}{b}\right)^{-\frac{1}{2}}. \tag{7}$$

This corresponds to the conventional Hall-Petch relationship for normalized stress with an exponent of $-1/2$ associated with the normalized grain size.

Alternatively, for very small grain sizes ($d < 100$ nm), the average number of dislocations in the pile-up tends to one, or to a small constant number K_2, giving pile-ups breakdown [11,14]. In the absence of a dislocation pile-up, replacing the value of n in Eq. (3) as constant K_2, the normalized stress then can be expressed as

$$\frac{\tau_e}{\mu} = \frac{K_2}{\pi(1-v)}\left(\frac{d}{b}\right)^{-1}, \tag{8}$$

where the normalized effective shear stress, $\left(\frac{\tau_e}{\mu}\right)$, is directly proportional to the reciprocal of the normalized grain size, $\left(\frac{d}{b}\right)^{-1}$. Thus, the exponent of the normalized grain size changes from -1/2 to -1 when there is a transition from large to small grain size ranges.

Since experimental experience [11,14] shows that in a wide grain size range the grain size exponent usually shows a continuously decreasing trend in the Hall-Petch relationship with decreasing grain size, it is reasonable to assume that the normalized effective shear stress is directly proportional to the power of the normalized grain size so that

$$\frac{\tau_e}{\mu} = K_3 \cdot \left(\frac{d}{b}\right)^{\beta}, \tag{9}$$

where K_3 and β are constants depending on the grain size range. Thus, for large grain sizes $d > 1000\ nm$, $K_3 = \sqrt{\frac{K_1}{\pi(1-\upsilon)}}$ and $\beta = -1/2$ (see Eq. 7), whereas for small grain sizes $d < 100\ nm$, $K_3 = \frac{K_2}{\pi(1-\upsilon)}$ and $\beta = -1$ (see Eq.8). For the intermediate submicron-size range, the value of the exponent β lies between -1/2 and -1.

Considering again Schmid's law, as well as taking into account also the yield stress, σ_o, of the single crystal, the conventional Hall-Petch relationship is now modified to the form

$$\frac{\sigma}{\mu} = \frac{\sigma_0}{\mu} + A^* \cdot \left(\frac{d}{b}\right)^{\beta}, \tag{10}$$

where A^* is a constant depending on the grain size range and the value of the exponent β is determined by using experimental data.

3. Verification of the proposed model and its application to experimental data

The application of Eq. (10) to experimental data may be examined using data from several ultrafine-grained fcc metals such as Al [5,19-22], Au [23,24], Cu [25,26] and Ni [27-30] as well as solid solution Al-1wt%Mg and Al-3wt%Mg alloys [5] having an average grain size, *d*, in the range between 100 and 1300 nm. For the purpose of the analysis, it is worthwhile examining the data in a double logarithmic representation as shown in Fig. 2. It should be noted that, for a given material, the σ_o term is usually much smaller than the yield stress of the ultrafine-grains. For example, for pure Cu σ_o is less than 30 MPa and σ is greater than 380 MPa [31]. For this reason, to a first approximation, and with an error of less than 10%, the normalized stress (σ/μ) can be considered proportional to the power of the normalized grain size (*d/b*). Thus, a straight line can be fitted approximately to the $\ln\left(\frac{\sigma}{\mu}\right) - \ln\left(\frac{d}{b}\right)$ data and the slope gives, approximately, the value of the exponent β. It is readily apparent from Fig. 2 that the experimental data fit a straight line having a slope of about $-3/4$.

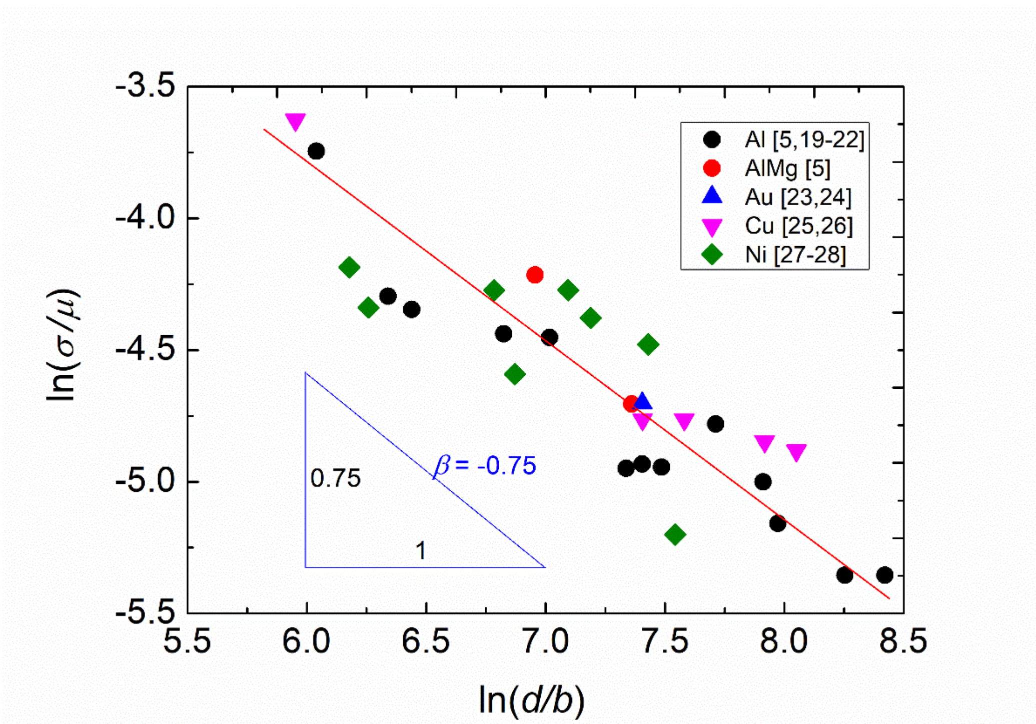


Figure 2: *Characterization of the relationship between normalized yield stress and normalized grain size in a double-logarithmic* $\left(ln\frac{\sigma}{\mu} - ln\frac{d}{b}\right)$ *representation for ultrafine-grained fcc metals and solid solution alloys.*

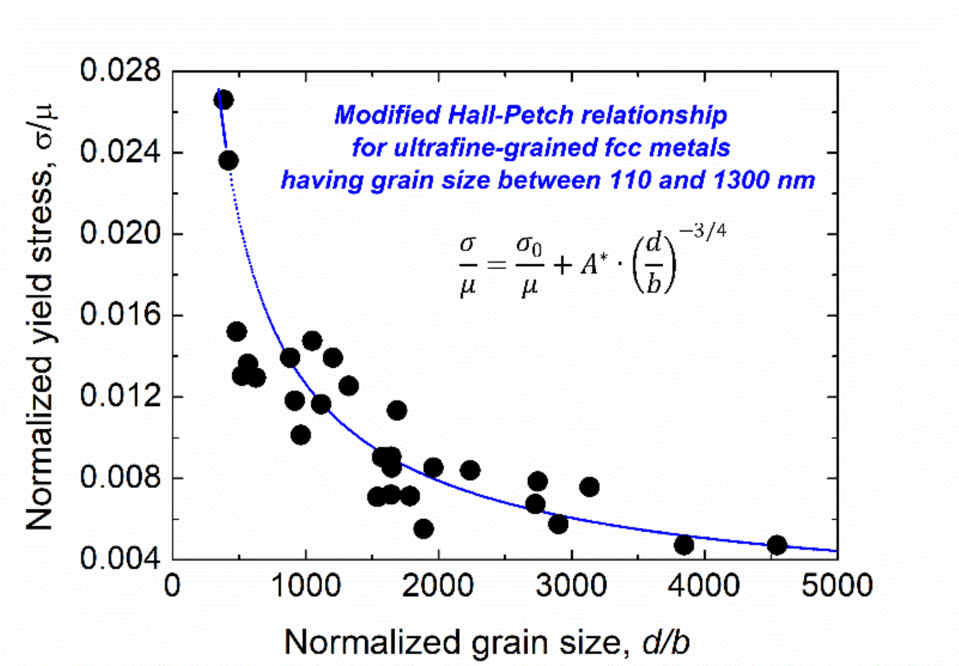


Figure 3: *Direct presentation of the normalized yield stress-normalized grain size relationship, proposing a modification of the Hall-Petch relationship for ultrafine-grained fcc metals and solid solutions.*

Based on this analysis, Fig. 3 shows directly the normalized yield stress, σ/μ, as a function of the normalized grain size, d/b, with a best-fitted line based on Eq. (10). This best fitting is obtained when $\frac{\sigma_0}{\mu} = 0.0008 \pm 0.0002$, $A^* = 2.22 \pm 0.15$ and $\beta = -0.75 \pm 0.04$. The analysis shows clearly that the strength of ultrafine-grained fcc metals and solid solutions can be uniformly described by an appropriate modification of the Hall-Petch relationship of the form

$$\frac{\sigma}{\mu} = \frac{\sigma_0}{\mu} + A^* \cdot \left(\frac{d}{b}\right)^{-3/4}. \tag{11}$$

The fact that several ultrafine-grained metals and alloys behave according to this modified Hall-Petch relationship is indicative of the robustness of the applicability of the model and the underlying physical phenomenon. Based on Eq. (11) with the parameters given above, the strength of any sub-micron structured fcc metal or solid solution may be estimated if the grain-size is known. It should be noted that the conventional Hall-Petch behavior (for $d > 1000\ nm$, $\beta = -1/2$) is the consequence of the activation of multiple slip within the individual grains. In the case of single slip ($d < 100\ nm$), $\beta = -1$. In the middle range, the value of -3/4 for β is certainly representing an intermediate state in the slip-system.

In summary, an appropriate modification of the original Hall-Petch relationship is proposed in order to maintain its applicability for submicron-structured fcc metals and solid solution alloys. It is demonstrated that the proposed relationship works well for a uniform description of a large number of results in the literature thereby demonstrating the overall robustness of the applicability of this model. In addition, and as an important consequence, the modified Hall-Petch relationship permits an estimation of the strength of any sub-micron structured fcc metal or solid solution when the grain-size is known.

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Data Availability Statement: The data that support the findings of this study are all own results of the authors, not available anywhere.

Conflict of Interest Statement: The authors declare that they have no conflict of interest.

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