Derivation of a new multiscale model: II. Deriving a modified Hall-Petch relation from the multiscale model and testing it for nano, micro, and macro materials

Sadeem Abbas Fadhil1,2, Mohsen A. Hassan3,4, Jazeel H. Azeez5 and Munaf S. Majeed1
1Al-Nahrain Nano renewable Energy Research Center (NNERC), Al-Nahrain University, 10072 Baghdad, Iraq.
2Center for Advanced Materials, Department of Mechanical Engineering, University of Malaya, 50603 Kuala Lumpur, Malaysia.
3School of Innovative Design Engineering, Department of Materials Science and Engineering, Egypt-Japan University of Science and Technology (E-JUST), New Borg El-Arab City, 21934, Alexandria, Egypt.
4Department of Mechanical Engineering, Assiut University, Assiut 71516, Egypt
5Physics Department, College of Science, Al-Nahrain University, 10072 Baghdad, Iraq.
Corresponding author email: sadeemfadhil@yahoo.com

Abstract. In the present work, a multiscale Hall-Petch relation is derived from a new multiscale model and applied for all ranges of grain sizes. The derived multiscale Hall-Petch relation has a superior over the previous models represented by its direct applicability on any material without the need for prior knowledge of grain size distribution. The new multiscale relation applicability is tested with three elements Mg, Zn, and Fe due to their importance in industrial applications. The comparison among the coefficients of different materials suggested a criterion to predict the reverse Hall-Petch relation.

1. Introduction
Quantum mechanics and classical mechanics offer a cumulative picture of the properties of the material. Within the small dimensions; quantum mechanics is dominant and offers the proper interpretation for the phenomena that occur in the atomistic and molecular scale [1]. On the other hand, classical mechanics shows enough description of large-scale materials without tremendous computation challenges [2–4]. However, there is a transition region that still stands as a heavy rock in front of both mechanics, this region is the mesoscale region [5]. The mesoscale represents a difficulty that faces the quantum mechanics, because of the extreme computation capabilities required to do the calculations, while the classical mechanics is not compatible with many experimental results within such region [6–8]. One of the relations that face such a challenge is the Hall-Petch relation [9]. This relation applies well to the classical scale of grain sizes above 100 nm while below a certain grain size, usually close to 10 nm, a relation reversal to the traditional Hall-Petch relation appears experimentally for many materials [10]. Many works were done to interpret (or predict) such a reverse relation; however, the problem is still not solved [11–16].

One of the reasons, that prevent reaching a cumulative Hall-Petch relation for all grain sizes, is the absence of a sufficient multiscale model; that is capable of describing the physical phenomena at all ranges of grain sizes [17]. In previous work, the multiscale Hall-Petch relation was applied directly without derivation following the concepts of a new theory called the time of events theory [18,19].

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new multiscale model will be used in the present work to derive a multiscale Hall-Petch relation that is capable of describing the experimental results for different materials. A criterion is suggested to predict the reverse Hall-Petch relation for any material from the parameters of the present model.

2. Derivation of a multiscale Hall-Petch relation

The time of events theory is based on assuming two spacetimes; the first is a dynamic space-time and the second is a static space-time. From a mathematical perspective, both spacetimes together form a complex space-time. The dynamic space-time represents the real space-time; in figure (1) is shown through $x'$, $t'$, and $x_r$ axes. The dynamic space-time moves at the speed of light at the direction of the real axis, $x_r$, which represents the middle axis between $x'$ and $t'$. While the static space-time represents the imaginary space-time; in figure (1) is shown through $t_i$ and $x_i$ axes. The $ct'$ and $ct_i$ are time axes, whereas $x_r$, $x'$ and $x_i$ are spatial axes. Therefore, the particle’s motion will have two velocity components; real $v_r$ and imaginary $v_i$ as shown in figure 1. According to ref. [20]; as the particle’s mass decreases the angle $\theta$, (shown in figure 1) between the particle’s velocity and the real axis, decreases. In other words, when the angle $\theta$ increases the real component decreases and the reverse is true for the imaginary component.

To derive the multiscale Hall-Petch relation, we will start from the equation of the total energy as in the first paper [20]:

$$\left(\frac{2K}{m}\right)(\cos \theta + i \sin \theta)^2 - \left(\frac{2V_{pi}}{m}\right)(\cos \theta + i \sin \theta)^2 = \left(\frac{2E_i}{m}\right)(\cos \theta + i \sin \theta)^2 \quad (1)$$

Where $m$ and $\theta$ are the particle’s mass and the angle of the particle’s velocity with the real axis respectively, $K_i$ is the kinetic energy operator, $V_{pi}$ is the potential energy operator, and $E_i$ is the eigenvalue of the total energy. For several n particles in the assembly, we can sum the energy equations as follows:

$$\sum_{i=1}^{n} \left(\frac{K_i}{m_i}\right)(\cos \theta_i + i \sin \theta_i)^2 - \sum_{i=1}^{n} \sum_{j=1}^{n} \left(\frac{V_{ij}}{m_i}\right)(\cos \theta_i + i \sin \theta_j)^2 = \sum_{i=1}^{n} \left(\frac{E_i}{m_i}\right)(\cos \theta_i + i \sin \theta_i)^2 \quad (2)$$

Where $V_{ij}$ is the potential energy between the $i^{th}$ particle and the $j^{th}$ particle. Taking the knowledge that we are targeting grains with masses heavier than the proton, therefore for the real part in equation (2),
only the $\sin^2 \theta_i$ terms will be saved, because the angle $\theta_i$ is close to 90°. For the same previous reason, the imaginary part in equation (2) will be ignored. Accordingly, equation (2) becomes:

$$
\sum_{i=1}^{n} \left( \frac{K_{ai}}{m_i} \right) - \sum_{i=1}^{n} \sum_{j=1}^{n} \left( \frac{V_{ij}}{m_j} \right) = \sum_{i=1}^{n} \left( \frac{E_i}{m_i} \right)
$$

(3)

For the grains, the right-hand side in equation (3) becomes continuous, due to large values of the principal quantum number. Therefore, it is better to replace the eigenvalue terms with a total energy term $E_i$, besides, the sign for the potential energy term will be assumed negative, because within the granular scale only the attraction forces play the major role, therefore equation (3) becomes:

$$
\sum_{i=1}^{n} \left( \frac{K_{ai}}{m_i} \right) - \sum_{i=1}^{n} \sum_{j=1}^{n} \left( \frac{V_{ij}}{m_j} \right) = E_i \sum_{i=1}^{n} \frac{1}{m_i}
$$

(4)

The number of particles included in the summation depends on the force’s type between the grains and the geometry of the studied system. Equation (4) is a general equation and can be considered the starting point for studying different materials’ properties and at different grain sizes. It is clear that the new multiscale model, unlike other models, contains the mass term in the denominator, which is crucial in deriving the general Hall-Petch relation as will be done in section (3).

3. Deriving a general Hall-Petch relation for different grain sizes:

In studying the mechanical properties, it is important to note that the fracture in any material starts at its weakest point. Usually, the weakest points for a material lie on the grain boundaries. On the grain boundaries, the atoms or molecules either connect with other atoms or molecules at the same grain or connect with atoms or molecules on the neighbouring grain. Usually, the fractures start on the grain boundaries or in the region between grains in composite materials. In general, and according to the Hall-Petch relation; the larger grains are less resistant to the fracture initiation than the smaller ones. Therefore, in deriving the right model, we need to focus on the larger grains [12]. To simplify the problem, we will separate the atoms manipulation from the grains. From equation (5), if we consider a system of grains with masses, $M_{gi}$, are connected to neighboring atoms with masses, $m_i$, then equation (5) can be rewritten in terms of grains and atoms as follows:

$$
\sum_{i=1}^{n} \left( \frac{K_{ai}}{m_i} \right) + \sum_{i=1}^{N} \left( \frac{K_{gi}}{M_{gi}} \right) - \sum_{i=1}^{n} \sum_{j=1}^{n} \left( \frac{V_{aij}}{m_j} \right) - \sum_{i=1}^{n} \sum_{j=1}^{n} \left( \frac{V_{gij}}{M_{gi}} \right)

= E_{ai} \sum_{i=1}^{n} \frac{1}{m_i} + E_{gi} \sum_{j=1}^{N} \frac{1}{M_{gi}}
$$

(5)

Where $K_{ai}$ and $K_{gi}$ are the kinetic energies for the $i^{th}$ atom and grain respectively. $n$ and $N$ are the numbers of atoms and grains respectively. $m$ is the number of atoms on the grain boundary. $V_{aij}$ and $V_{gij}$ are the potential energies from the interaction between the $i^{th}$ atom (or grain) and the $j^{th}$ atom (or grain) respectively. $V_{aij}$ is the potential energy from the interaction between the $i^{th}$ atom and the $j^{th}$ grain. $V_{gij}$ is the potential energy from the interaction between the $i^{th}$ grain and the $j^{th}$ atom. $E_{ai}$ and $E_{gi}$ are the total energies for the atoms and grains respectively. For the isotropic composition of atoms, the term can be expressed as:

$$
\sum_{i=1}^{n} \frac{1}{m_i} = \frac{n}{m_a}
$$

(5)

Substituting equation (6) in equation (5) and after neglecting the grain terms in the total energy and the kinetic energy due to relatively large grain masses, equation (5) becomes after multiplying with $m_a/n$ and simplifications:
\[ K_a = \sum_{i=1}^{n} \sum_{j=1}^{n} \left( \frac{V_{ij}}{n} \right) - \sum_{i=1}^{N} \sum_{j=1}^{N} V_{gij} \left( \frac{m_a}{nM_{gi}} \right) \]

\[ -\sum_{i=1}^{m} \sum_{j=1}^{m} V_{gij} \left( \frac{1}{n} \right) \left( 1 + \left( \frac{m_a}{M_{gi}} \right) \right) \]

\[ = E_{at} \]

Assuming the same density for the atoms and grains then Equation (7) can be used to conclude the modified Hall-Petch relation that can fit for different grain sizes from nano to macro scales. We need to focus on the weakest terms in the potential energy to conclude the right relationship for the yield stress versus the grain size. The first term in the potential energy is between atoms (or molecules) within the grain. This term is usually larger than other terms and its effect on fracture and yield stress is undetected, therefore it will be ignored here. The second term in the potential energy contains the factor \( m_a / M_{gi} \).

The factor \( m_a / M_{gi} \) is proportional to \( d_a^3 / d_g^3 \), where \( d_a \) and \( d_g \) are the atom’s and the grain’s diameter respectively. The potential energy from the interaction between grains \( V_{gij} \) is directly proportional to the surface area of the grains, i.e., \( V_{gij} \propto d_g^2 \), therefore, the second term in the potential energy \( V_{gij} \left( m_a / nM_{gi} \right) \propto (1 / d_g) \). The third term in the potential energy contains two terms; the first one is \( \sum_{i=1}^{m} \sum_{j=1}^{m} V_{gij} (1 / n) \). The number of atoms \( m \) on the boundaries which have contact with the grains is proportional to the surface area of the grain, accordingly \( \sum_{i=1}^{m} \sum_{j=1}^{m} V_{gij} (1 / n) \propto d_g^2 \). The other term is \( \sum_{i=1}^{m} \sum_{j=1}^{m} V_{gij} (1 / n) \left( m_a / M_{gi} \right) \), which contains two elements depend on the grain size; the first is proportional with \( d_g^2 \) and the other one from the grain mass is inversely proportional with \( d_g^3 \), therefore the resultant is \( \sum_{i=1}^{m} \sum_{j=1}^{m} V_{gij} (1 / n) \left( m_a / M_{gi} \right) \propto (1 / d_g) \), which is similar to the first term in the potential energy.

To have a complete description for the yield stress versus grain size relation we need to consider the Coble creep effect [21], which is inversely proportional with \( d_g^3 \), accordingly, the modified Hall-Petch relation to predict the yield stress \( \tau \) should contain the following terms:

\[ \tau \propto \frac{1}{d_g} + d_g^2 + \frac{1}{d_g^3} \]

(8)

The approximations followed to conclude equation (8) from equation (7) have considered a constant grain size, which is an ideal case. To consider the different grain sizes we need to take the square root of each of the above terms [22], therefore:

\[ \tau \propto \frac{1}{d_g^{3/2}} + d_g + \frac{1}{d_g^{3/2}} \]

(9)

There is still another case, in which all the above factors are found to work together. The term describing such a case is given through multiplying all the terms, which yields \( 1/d_g \) after averaging through taking the square root. Therefore, the final equation after adding the constants will be as follows:

\[ \tau = c_o + \frac{c_1}{d_g^{1/2}} + c_2d_g + \frac{c_3}{d_g^{3/2}} + \frac{c_4}{d_g} \]

(10)

Where \( c_o, c_1, c_2, c_3, \) and \( c_4 \) are the proportionality constants. From equation (10), it is obvious, that the first two terms represent the usual Hall-Petch relation. These terms are found inherently in equation (7). The third term is the grain’s surface area term. This term has also appeared previously in equation (7). It is related to the number of atoms connected with a certain grain. The fourth term represents the...
Coble's creep term. This term doesn't appear in equation (7), because it is related to a temperature-controlled phenomenon, whereas equation (7) is under stable temperature conditions. Therefore, we need to add the term independently. The fifth term is the cumulative term, represents all the previous terms working together. The Coble's creep term, represented by the constant $c_4$, acts for small grain sizes below 30 nm [21]. The surface area term; constant $c_2$, works in the range between the Coble creep and the classical Hall-Petch; i.e., between 30 nm and 100 nm, where the surface area for the grains has more impact on the yield stress. The first two terms $c_o$ and $c_1$ work for the grain sizes above 100 nm. The cumulative term value can give an idea about the interference between different terms, which are related to different mechanisms; acting on the value of the yield stress.

4. Results and Discussion:
To test the present model through the whole range of grain sizes; the elements Zn, Mg, and Fe will be adopted. The present model applied to the experimental data for Mg from the references [23–32], for Zn from the references [32–40], and for Fe from the references [32, 41, 42–49, 50–58]. The resulted curves are in figures 2, 3, and 4 for Mg, Zn, and Fe respectively. The fitting parameters are listed in table 1.

| The element | $c_o$ | $c_1$ | $c_2$ | $c_3$ | $c_4$ |
|-------------|-------|-------|-------|-------|-------|
| Mg          | 21.88 | 160   | 0.01719 | 1.354 | 62.66 |
| Zn          | 55.86 | 180.1 | 0.06247 | 5.272 | 59.35 |
| Fe          | 62.3  | 846.9 | 0.0009634 | 5.644 | 112.2 |

The fitting curves for both Mg and Zn show clearly the traditional and the reverse Hall-Petch manner. We chose these elements because of the reverse Hall-Petch appeared in part of their yield. The model can also fit well for the other types of elements and composites, that do not show the reverse Hall-Petch within their small grains range, as shown in figure 4 for Fe. Table 1 shows relatively large values for the classical coefficients $c_o$ and $c_1$ in addition to the cumulative coefficient $c_4$, which implicitly includes the classical pile-up effects, that usually occur between grains. The relatively large values for the classical terms are reasonable, because these coefficients are the leading ones within the classical region, while the surface and the Coble creep effects appeared only within the small grains range. The surface term represents a new addition from the present model. This term becomes more important when the surface effects are not-negligible; usually within nanoscale grain sizes. Generally, the model does not need prior knowledge of grain size distribution to be applied. Indeed, it can give a rough estimate for the grain size distribution of the studied material, but with a proper calibration procedure. The fitting results for the present elements data showed a similar manner with the Cu and Ni in previous work [19]. The classical coefficient, $c_1$, for Fe is much larger than in Mg and Zn. Unlike the classical coefficients, the $c_2$ coefficient in Fe is smaller with an order of magnitude than in Mg and Zn, which indicates that the surface term is of minor effect in Fe. This gives an interpretation of why there is no reverse Hall-Petch relation in Fe. The large classical coefficient and a relatively small surface coefficient for Fe extended the applicability of the classical Hall-Petch relation to relatively small grain sizes, below 30 nm.
Figure 2. Fitting from the modified Hall-Petch relation for the Mg specific yield stress versus the grain size. The points of the experimental data are from the references [23-32].

Figure 3. Fitting from the modified Hall-Petch relation for the Zn specific yield stress from the tensile compression tests versus the grain size. The points of the experimental data are from the references [32-40].
Figure 4. Fitting from the modified Hall-Petch relation for the specific yield stress of Fe versus grain size. The points of the experimental data are from the references [38-55, 29].

5. Conclusions:
In the present work, a new multiscale Hall-Petch relation is concluded from a new multiscale model then applied to three materials Mg, Zn, and Fe. The new multiscale relation brings with it the classical terms in the Hall-Petch relation with a new term which is the surface term; the coefficient $c_2$. This term and the Coble’s creep effect term are responsible for the reverse Hall-Petch at the small grain sizes below 30 nm. According to the present model, when the grain size reached a certain value, critical grain size, the pile-up mechanism will be overcome by the forces connecting the neighbouring grains, which is increased with increasing the number of atoms on the boundaries, i.e. at that range it is proportional with the square of the grain size. The reverse Hall-Petch relation found in Mg and Zn, whereas it didn’t appear in Fe. Comparing the coefficients $c_1$ and $c_2$; reveals the $c_1$ coefficient is larger in Fe, while $c_2$ coefficient is smaller with an order of magnitude relative to Mg and Zn. This means that the surface effects, due to the relatively increased surface area, are relatively small in Fe compared with the forces between grains. In other words, the forces between grains are mainly dominating the binding forces between grains, not the number of surface atoms connected with the neighbouring grains. The surface term value can represent the criteria to predict the reverse Hall-Petch relation. The relatively larger value for the surface term is related to the reverse Hall-Petch relation appearance.

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