Estimation on effective modulus of polycrystalline aggregates in various geometric configurations

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**Estimation on Effective Modulus of Polycrystalline
Aggregates in Various Geometric Configurations**

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2012

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Abstract

Polycrystalline aggregates are made in a wide array of geometric configurations (thin wire, thin film, and bulk configurations, to name a few) for engineering applications. Various elastic moduli have been reported for these configurations. The inconsistency of Young's modulus has been attributed to micro-defects and microscopic texture in the existing open literature. In the present study, the modulus variation of the polycrystalline aggregates in various geometric configurations is explained from a new perspective, i.e., the constraint and interaction among the grains. A finite element scheme with statistic consideration based on the Central Limit Theory is employed to systematically investigate the effective Young's modulus of the polycrystalline copper aggregates in various geometric configurations. The correlation between the effective modulus of the polycrystalline copper and the geometric configuration is fully pictured. For the polycrystalline copper, the spread of the effective modulus due to the geometric configuration could be as large as 20%.

Keywords: effective modulus, polycrystalline aggregates, thin film, thin wire, finite element analysis, statistics.

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This thesis is dedicated to my parents, my husband and my son.

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List of Symbols

a	Width of membrane
A	Area
$A_{ijkl}^r, \mathbf{A}_r$	Strain concentration tensor
b	Width of beam
$B_{ijkl}^r, \mathbf{B}_r$	Stress concentration tensor
$C_{ijkl}, \mathbf{C}$	Elastic stiffness tensor
C_{IJ}	Elastic stiffness matrix
$C_{ijkl}^*, \mathbf{C}^*$	Effective elastic stiffness tensor
$C_{IJ}^{p,\sigma}$	Elastic stiffness matrix in the state of plane stress
d	Grain size
E	Young's modulus
E_H	Hill's average of Young's modulus
E_r	Reduced modulus
E_R	Reuss lower bound of Young's modulus
E_V	Voigt upper bound of Young's modulus
E^*	Effective Young's modulus
$\tilde{E}$	In-plane Young's modulus
$\tilde{E}_R^{p,\sigma}$	Reuss lower bound of in-plane Young's modulus
$\tilde{E}_V^{p,\sigma}$	Voigt upper bound of in-plane Young's modulus
G	Shear modulus
G_{H-a}, G_H	Hill's arithmetic average of shear modulus
G_{H-g}	Hill's geometric average of shear modulus
G_R	Reuss lower bound of shear modulus
G_V	Voigt upper bound of shear modulus
G^*	Effective shear modulus

G_{HS}^L	Hashin-Shtrikman lower bound of shear modulus
G_{HS}^U	Hashin-Shtrikman upper bound of shear modulus
$\tilde{G}$	In-plane shear modulus
$\tilde{G}_R^{p,\sigma}$	Reuss lower bound of in-plane shear modulus
$\tilde{G}_V^{p,\sigma}$	Voigt upper bound of in-plane shear modulus
h	Substrate thickness
H	Hardness
$I_{ijkl}, \mathbf{I}$	Identity tensor
K	Bulk modulus
K_H	Hill's average of bulk modulus
K_R	Reuss lower bound of bulk modulus
K_V	Voigt upper bound of bulk modulus
K^*	Effective bulk modulus
$\tilde{K}$	In-plane bulk modulus
$\tilde{K}_R^{p,\sigma}$	Reuss lower bound of in-plane bulk modulus
$\tilde{K}_V^{p,\sigma}$	Voigt upper bound of in-plane bulk modulus
L	Length
m, M	Element number
n	Sample size
N	Grain number
p	Pressure
P	Force
P_N	Population
R	Radius of curvature
s	Standard deviation of sample
S_b	Stiffness of beam
$S_{ijkl}, \mathbf{S}$	Elastic compliance tensor
S_{IJ}	Elastic compliance matrix
$S_{ijkl}^*, \mathbf{S}^*$	Effective elastic compliance tensor
$(S_r^*)_{ijkl}, \mathbf{S}_r^*$	Eshelby's tensor
$S_{IJ}^{p,\sigma}$	Elastic compliance matrix in the state of plane stress
t	Film thickness

t_{ij}	Transformation matrix
T_i	Traction
$u, u_x, u_y, u_z,$	Displacement
U	Strain energy
$\bar{u}$	Average displacement
w	Deflection
$\bar{x}$	Mean value of sample
Δ	Prescribed displacement
α, β, γ	Euler angle
δ	Dimensionless quantity
δ_{ij}	Kronecker delta
ϵ, ϵ_{ij}	Strain tensor
$\bar{\epsilon}_{ij}$	Volume average of strain tensor
ϵ_I	Strain matrix
ϵ_{ij}^0	Uniform strain tensor
ϵ_{ij}^r	Strain tensor of grain r
μ	Mean value of population
ν	Poisson's ratio
ν_H	Hill's average of Poisson's ratio
ν^*	Effective Poisson's ratio
σ	Standard deviation of population
σ_0	Residual stress
σ, σ_{ij}	Stress tensor
σ_I	Stress matrix
$\bar{\sigma}_{ij}$	Volume average of stress tensor
σ_{ij}^0	Uniform stress tensor
σ_{ij}^r	Stress tensor of grain r

Chapter 1 Introduction

1.1 Thin film applications

Polycrystalline aggregates, known as single phase material and yet heterogeneous material due to the anisotropy of each grain, are widely used in modern industries. Traditionally, the polycrystalline aggregates are made in three-dimensional bulk configuration. In the past decades, the micro- and nano-scale components have been broadly introduced to meet the requirement of ongoing miniaturization in the fields of microelectronics, optics, magnetics and micro-electro-mechanical system (MEMS) (Nix, 1989; Freund and Suresh, 2003; Connolley *et al.*, 2005). Consequently, the polycrystalline aggregates are often made in micrometers or even nanometers in the form of thin films.

The thin films, as the basic geometric configuration in the construction of small-scale devices and systems, have a very wide application. In a state-of-the-art integrated circuit microprocessor, hundreds of millions of thin-film transistors are included and connected by numerous thin wires or interconnects. The thin films are also the basic components of many MEMS devices (e.g., accelerometers, rate gyroscopes, ink-jet printer head as well as digital micro-mirror device) which are the integration of mechanical elements, sensors, actuators and electronics (Spearing, 2000; Romig Jr. *et al.* 2003). In addition, the thin films as surface coating are used as wear resistant protection on cutting tools and hard disks, to name a few.

1.2 Motivation

During fabrication and application, the thin film materials are often subjected to mechanical stresses. For example, the lattice mismatch between the substrate and the thin film causes misfit strains which result in stresses in the thin film. The thermal stress is introduced by the difference in the thermal expansion coefficient between the film and its surrounding material (Nix, 1989; Freund and Suresh, 2003). Large stresses may lead to unfavorable failures, such as excessive deformation, delamination and fracture. Hence, understanding the mechanical properties, e.g., Young's modulus and yield strength etc., of the thin film materials is important to ensure the reliability and integrity of these devices. Also, these mechanical properties, especially the Young's modulus, are indispensable in the stage of design and evaluation.

It has been reported that the mechanical properties of thin film materials are drastically different from those of bulk materials (Hardwick, 1987; Nix, 1989; Vinci and Vlassak, 1996; Arzt, 1998). Direct extrapolation from the properties of the bulk materials is highly questionable to characterize the thin film materials. Thus, many researchers have been studying the mechanical behavior and properties of the thin film materials by means of experimental measurement and theoretical analysis.

Various experimental methods have been developed to describe the mechanical properties of the thin film materials (Huang and Spaepen, 2000; Kalkman *et al.*, 2001; Volinsky *et al.*, 2001; Son *et al.*, 2003; Emery and Povirk, 2003; Espinosa *et al.*, 2004; Hong *et al.*, 2005; Xiang *et al.*, 2006; Faurie *et al.*, 2010). There are two common thin film configurations used in the experiments, i.e., the free-standing thin film and the film attached to a substrate. Inconsistent experimental results have been reported. Let

us examine the copper thin film as an example. On the one hand, some of the experimental measurements showed that the Young's modulus of the thin film dramatically deviates from that of bulk counterpart. This difference could be as large as 20% to 30% (Huang and Spaepen, 2000; Volinsky *et al.*, 2001). On the other hand, some researchers reported that there is no change of modulus between the thin film configuration and the bulk configuration (Espinosa *et al.*, 2004; Xiang *et al.*, 2006).

Figure 1.1 Classification of heterogeneous material.

From theoretical point of view, various analytical models have been developed for predicting the effective elastic properties of heterogeneous materials. A classification of heterogeneous material is shown in Fig. 1.1. The multiphase material with distinct matrix phase is commonly referred to as matrix/particulate material; while for the multi-continuous material, all the constituents or phases are on the equal status, that is, the matrix and particulate cannot be clearly distinguished. In the case of single phase material such as the polycrystalline aggregates, heterogeneity comes from the anisotropic behavior of each grain having different crystallographic orientation, shape and size.

With reference to Fig. 1.1, Table 1.1 presents the analytical approaches developed for different type of materials. It is seen that the Mori-Tanaka method and three-phase model may not be applicable for studying the polycrystalline aggregates. This may be attributed to the assumption introduced in these models, that is, there is one phase acting as the matrix and the rest phases are particulates embedded in the matrix. However, for the polycrystalline aggregates, each of the grains has the same significance in determining the effective properties. Hence, the concept of a matrix with embedded particulates no longer makes sense.

Table 1.1 Analytical approaches for heterogeneous material.

Analytical approach		Heterogeneous material		
		Multiphase material		Single phase material
		Matrix/particulate material	Multi-continuous material	
Bound methods	Voigt-Reuss bounds ^a	Yes	Yes	Yes
	Hashin-Shtrikman bounds ^b	Yes	Yes	Yes
	Hill's average ^c	Yes	Yes	Yes
Approximate methods	Self-consistent model ^d	Yes	Yes	Yes
	Mori-Tanaka model ^e	Yes	No	No
	Three-phase model ^f	Yes	No	No
^a Hill (1952), Hill (1963). ^b Hashin and Shtrikman (1962 a, b; 1963), Walpole (1966a, b), Willis (1977, 1981). ^c Hill (1952), Mura (1987). ^d Hill (1965), Budiansky (1965), Kröner <i>et al.</i> (1966), Wu (1966), Walpole (1969), Morris (1970, 971), Willis (1977, 1981). ^e Mori and Tanaka (1973), Weng (1984, 1990), Benveniste (1987). ^f Christensen and Lo (1979), Christensen (1990).				

Bound methods, such as Voigt-Reuss bounds (Hill, 1952) and Hashin-Shtrikman bounds (Hashin and Shtrikman, 1962a, b), provide the upper and lower values for the effective properties and these values sometimes deviate far away from each other. Considering the fact that Hill's average (Hill, 1952) is based on the upper and lower bounds, here, we categorize it into the group of bound methods in Table 1.1. Approximate methods like self-consistent scheme (Morris, 1970, 1971; Willis, 1977, 1981), give an explicit value for the effective properties which should fall within the range between the lower and upper values. It is worthwhile to mention that all these methods were originally proposed for the estimation of the effective elastic properties of the bulk material from the elastic constants of their constituents (each grain in a polycrystalline aggregate, for instance). Hence, these methods may not be accurate when we study the thin film materials. Recently, for the polycrystalline free-standing thin film, some analytical results for the effective properties such as biaxial modulus, Young's modulus and Poisson's ratio were reported (Baral *et al.*, 1982; Anatassakis and Siakavellas, 2001; Huang and Weaver, 2005). These results, however, were generally derived for the thin films with textures. In the case of the substrate-attached thin film, the coupling deformation between the film and the substrate makes it almost impossible to analytically evaluate the effective Young's modulus of the film material.

With the rapid progress of computational speed and memory capacity, numerical simulation, such as finite element method (FEM), has become an effective method in studying the heterogeneous material. The numerical simulation has certain advantages over both the theoretical method and experimental test. In comparison with the theoretical method, the numerical simulation is able to deal with complex microstructural information and mutual interaction among the grains. In contrast with the experimental test, the numerical simulation is able to model the specimen with

different microstructures and isolate the essential factors affecting the mechanical behavior. Although a great endeavor has been given to numerically study the mechanical properties (e.g., the effective Young's modulus and Poisson's ratio) of the polycrystalline thin films, most studies focused on the free-standing thin films which have been further simplified into the state of plane stress or plane strain (Mullen *et al.*, 1997; den Toonder *et al.*, 1999; Choi, 2004; Zhang and Sun, 2006). The study on the mechanical behavior of the substrate-attached thin films has gradually received attention in recent years (Wikström and Nygård, 2002; Šiška *et al.* 2007; Shen, 2008, 2009). More study on the effective modulus of the substrate-attached thin film is needed.

From the foregoing review, it is seen that systematically studying the modulus difference of the polycrystalline aggregates in different geometric configurations is absent. We do not introduce both micro-defects (e.g., micro-cracks, micro-voids and compliant grain boundary) and preferred texture. In the present work, a new perspective is proposed via finite element simulation. Rather, we look into the constraint among the grains.

1.3 Description of geometric configurations

Let us consider various geometric configurations of the polycrystalline aggregates in micro and macro scale shown in Fig. 1.2. Figure 1.2 (a) is the configuration that there is only one row of the grains serially connected. This is the so-called bamboo-like structure observed in the polycrystalline interconnects (Besser *et al.*, 2001; Paik *et al.*, 2006; Zhang *et al.*, 2009; Brandstetter *et al.*, 2010). By adding more rows of the grains in y -direction (Fig. 1.2(b)), one has a thin film (Fig. 1.2(c)). By aggregating

more layers of the grains (Fig. 1.2(d)), one obtains a three-dimensional block (Fig. 1.2(e)). Alternatively, by adding the grains in the cross section, say, y - z plane (Fig. 1.2 (f)), one obtains the other wire configuration different from Fig. 1.2 (a). When more and more grains are added in the y - z plane, one also reaches a three-dimensional block (Fig. 1.2(e)).

Figure 1.2 Polycrystalline aggregates in various geometric configurations.

Furthermore, depending on the degree of constraint on deformation imposed by the surrounding materials, it is considered to be free-standing or unconfined if the boundaries associated with its thin dimensions (i.e., the planes perpendicular to the film thickness or the planes parallel to the wire length) are free to deform, shown in Figs 1.2(a)-(d) and (f). Otherwise, it is said to be confined when these boundaries are constrained against deformation. In Figs 1.2 (g) and (h), part of these boundaries are shared with the surrounding material; while Figs. 1.2 (i) and (j) show that these

boundaries are completely confined by the surrounding materials (Freund and Suresh, 2003). The cases shown in Figs. 1.2 (h) and (j) are, respectively, named as the substrate-attached film and sandwiched film.

1.4 Objective and outline

In the present work, a new perspective is proposed to interpret the modulus variation of the polycrystalline aggregates in various geometric configurations. A systematic study on the effective Young's modulus of the polycrystalline aggregates in different macro-scale geometric configurations, as shown in Fig. 1.2, is performed through the finite element method within the scope of linear elastic continuum mechanics. Polycrystalline copper is selected for illustration. Such selection is made based on the following considerations. Firstly, polycrystalline copper is preferred as the most promising interconnect material in the advanced integrated circuits, due to lower electrical resistivity, higher electro-migration resistance and higher melting temperature in contrast to aluminum. Secondly, from the mechanical perspective, copper single crystal exhibits a medium degree of elastic anisotropy so that the polycrystalline copper has the Young's modulus in the range from 109 GPa (the Reuss lower bound) to 144GPa (the Voigt upper bound). Therefore, it is necessary to have a clear picture correlating the effective modulus with the geometric configuration. The objective of the present work is presented as follows.

- A finite element scheme in combination with statistic concept is employed to evaluate the effective elastic constants of the polycrystalline aggregates in bulk configuration. The accuracy of the proposed finite element scheme is validated against the existing analytical solutions.

- Polycrystalline copper in thin film configuration, including the free-standing, the substrate-attached and the sandwiched cases, is simulated to investigate the modulus variation via the verified finite element scheme.
- Simulation of the free-standing polycrystalline copper thin wire is carried out to evaluate the effective Young's modulus. The influence of the grain structure in the wire's cross section is considered.
- The micro-morphological influence on the effective elastic modulus of the polycrystalline aggregates is discussed.

In our simulation, no defects (i.e., micro-cracks, micro-voids, grain boundary sliding as well as compliant grain boundary) and size effect are involved. The numerical results clearly indicate the influence of the geometric configuration on the effective modulus of the polycrystalline aggregates.

The thesis is organized in seven chapters including this introductory chapter.

Chapter 2 reviews the current status of research on the effective mechanical properties of the polycrystalline aggregates.

Chapter 3 summarizes necessary fundamental concepts related to the effective elastic constants of the polycrystalline aggregates. The derivation of the bounds on the effective Young's modulus of the polycrystalline aggregates in the state of plane stress is included in this chapter.

In Chapter 4, a finite element scheme in combination with statistic concept is described. The proposed scheme is then used to model the polycrystalline aggregates in the three-dimensional bulk configuration. Key issues in the finite element

simulation are discussed, including mesh refinement, the effects of the number of grains aggregating the polycrystal and the grain shape on the effective elastic constants. Comparison is made among the numerical results and the analytical solutions.

Chapter 5 focuses on studying the polycrystalline copper in thin film configuration. For the free-standing thin film, the effect of film thickness and grain structure along the thickness direction is considered. Planar Voronoi tessellation is employed to investigate the effect of microstructural morphology (the variation of the grain shape and size). For the substrate-attached thin film, our emphasis is placed on the influence of the substrate stiffness and substrate thickness. The simulation of the sandwiched copper thin film is also performed in this chapter.

Chapter 6 estimates the effective modulus of the free-standing polycrystalline copper wire. The modulus sensitivity to the grain structure in the wire's cross-section is studied.

In Chapter 7, concluding remarks and key findings of this work are summarized.

Chapter 2 Literature Review

In this chapter, the existing methods for determining the effective mechanical properties of polycrystalline aggregates in various geometric configurations are reviewed. These methods may be classified into three groups: experimental measurement, analytical modeling and numerical simulation. To begin with, we review the mechanical testing techniques of thin film materials. Some experimental data for the Young's modulus of copper thin films are summarized for reference. The analytical approaches including bound methods and approximate methods are then presented. Lastly, we review the numerical methods.

2.1 Thin film experimental techniques

Due to the small dimension of thin film materials, the traditional mechanical testing devices for bulk materials cannot be directly applied to the thin film measurement. Various novel experimental techniques have been developed since 1980s. In general, these experimental techniques can be classified into two groups according to the film configuration: methods for the free-standing thin films and methods for the substrate-attached thin films. Hereby, we briefly review the most common experimental approaches in the literature. Readers are encouraged to refer to the work by Nix (1989), Yi and Kim (1999), Kraft and Volkert (2001), and Kamat (2009) for more information.

2.1.1 Experimental techniques for free-standing thin film

The advantage of this type of testing techniques is that the intrinsic mechanical behavior of thin film material can be directly determined. However, preparing and handling the specimen are tricky.

Microtensile testing

It is a tensile test performed on a thin film. A stretching load is applied uni-axially on the specimen to produce uniform stress and strain. The measured stress-strain behavior provides the straightforward data to determine the elastic and plastic properties (e.g., the Young's modulus and yield strength). A few improved tensile methods have been proposed to overcome the difficulties stemming from the specimen preparation and handling, as well as aligning between the loading and the specimen directions (Read *et al.*, 2001; Haque and Saif, 2002b; Espinosa *et al.*, 2003). For example, Haque and Saif (2002b) fabricated a free-standing thin film specimen with the force sensor beam at one end and the support beam assembly at the other end. By using their improved tensile testing equipment, even 30-50nm thick films can be measured. Espinosa and his colleagues (Espinosa *et al.*, 2003; Peng *et al.*, 2006) developed a so-called membrane deflection experiment (MDE). In this technique, a double “dog bone-shaped” free-standing film membrane in a fixed-fixed configuration is fabricated with an enlarged contact area at its center. Under a line loading imposed by a nanoindenter tip on this contact area, the gauge section of the free-standing film undergoes a pure stretch. Simultaneously, an interferometer on the bottom side of the membrane records its deflection.

An alternative technique is to deform a film that is still attached on a compliant substrate like polyimide by stretching the film/substrate composite structure (Kang and Ho, 1997; Yu and Spaepen, 2004). In this case, tensile tests on the substrate with and without the film are performed, respectively. The stress in the film is obtained by subtracting the substrate contribution from the force-displacement curve of the film/substrate system.

The tensile test has been widely used to characterize the mechanical properties of the thin film materials including copper (Huang and Spaepen, 2000; Read *et al.*, 2004; and Lin *et al.*, 2010), gold (Emery and Povirk, 2003) and aluminum (Read *et al.*, 2001; Haque and Saif, 2002a, b), to name a few.

Plane-strain bulge test

The bulge test, as a technique to measure the in-plane mechanical properties of the thin films, was originally proposed by Beams in 1959. It became popular after the rapid development of silicon micromachining technology (Vlassak and Nix, 1992). In the bulge test, a free-standing thin film is deflected by applying a uniform pressure p to one side of the film, shown in Fig. 2.1. Vlassak and Nix (1992) used a rectangular membrane (with aspect ratio larger than 4) which deforms in an in-plane strain state. The pressure p can be related to the deflection w at the center of a membrane by

$$p = 2 \frac{\sigma_0 t}{a^2} w + \frac{4}{3} \frac{Et}{(1-\nu^2)a^4} w^3, \quad (2.1)$$

where $2a$ is the membrane width, t is the membrane thickness, σ_0 is the in-plane residual stress, E and ν are Young's modulus and Poisson's ratio of the film.

Moreover, the in-plane stress and strain can be calculated from the measured pressure (p)-deflection (w) curve through the following formula (Kraft and Volkert, 2001),

$$\varepsilon = \frac{2w^2}{3a^2}, \quad (2.2)$$

$$\sigma = \sigma_0 + \frac{E}{1-\nu^2} \varepsilon = \frac{a^2}{2wt} p. \quad (2.3)$$

Hence, from the measured p - w curve, the residual stress σ_0 and the plane-strain modulus $E/(1-\nu^2)$ can be determined. As suggested by Vlassak and Nix (1992), the Young's modulus E and Poisson's ratio ν can be separately determined by comparing the bulge test result on rectangular membrane with that on square membrane. This technique has been applied to a wide range of materials, for example, Jayaraman *et al.* (1999) for polysilicon film, Kalkman *et al.* (2001) for aluminum, gold and tungsten films, Xiang *et al.* (2005, 2006) and Wei *et al.* (2007) for copper films, and Huang *et al.* (2007) for polymer film.

Figure 2.1 Schematic illustration of bulge test on a rectangular thin film.

Microbeam bending test

This technique, introduced by Weihs *et al.* (1988), involves the fabrication of cantilever beam specimen by micromachining. The deflection of the beam is induced

by a nanoindenter at the free end. A schematic geometry of the microbeam bending test is presented in Fig. 2.2.

Figure 2.2 Schematic illustration of microbeam before (solid line) and after (dashed line) loading imposed by an indenter.

Based on the elastic beam bending theory, the deflection w of a cantilever beam with a rectangular cross section can be determined by

$$w = \frac{4PL^3(1-\nu^2)}{Ebt^3}, \quad (2.4)$$

where P is the applied force; L , b and t are shown on Fig. 2.2; E and ν are Young's modulus and Poisson's ratio of the beam material, respectively. Furthermore, the stiffness S_b of the beam can be obtained as

$$S_b = \frac{dP}{dw} = \frac{E}{1-\nu^2} \frac{bt^3}{4L^3}. \quad (2.5)$$

By analyzing the P - w curve, the mechanical properties of the beam material can be characterized. This method was used by Baker and Nix (1994) for modulated Au-Ni films, Son *et al.* (2003) for gold and aluminum films, Nilsson *et al.* (2004) for chromium film and Hong *et al.* (2005) for copper film.

2.1.2 Experimental techniques for substrate-attached thin film

In comparison with the free-standing thin films, the substrate-attached films are easier to prepare. However, the presence of the substrate makes the interpretation of experimental data troublesome.

Nanoindentation test

Nanoindentation test has been used to characterize the hardness and Young's modulus of thin film material which is attached on a substrate (Baker and Nix, 1994; Doerner and Nix, 1986; Suresh *et al.*, 1999; Volinsky *et al.*, 2001; Han *et al.*, 2006). By pushing the rigid indenter tip into a specimen and then withdrawing it, the load (P)-indentation (w) behavior is recorded during the nanoindentation test, shown in Fig. 2.3.

Figure 2.3 (a) Schematic illustration of nanoindentation measurement; (b) load (P) - indentation (w) relation.

If friction and finite compliance of the measuring system and the indenter tip are neglected, the hardness H and reduced modulus E_r can be extracted using the following equations

$$H = P / A , \quad (2.6)$$

$$S = \beta \frac{2}{\sqrt{\pi}} E_r \sqrt{A}, \quad (2.7)$$

where the hardness H is defined as the ratio between the indentation load P and the projected contact area A under the indenter; the contact stiffness $S = dP/dw$ is calculated as the slope of the initial portion of the elastic unloading curve, with reference to Fig. 2.3 (b); β is a dimensionless parameter that depends on the indenter shape, for instance, $\beta \approx 1.08$ for the commonly used Berkovich tip which is three-sided pyramid (Chen and Vlassak, 2001).

The reduced modulus E_r is expressed in terms of the elastic properties of the indenter (i) and film (f) as

$$\frac{1}{E_r} = \frac{1 - \nu_i^2}{E_i} + \frac{1 - \nu_f^2}{E_f}. \quad (2.8)$$

Obviously, the reduced modulus of the film, $E_f / (1 - \nu_f^2)$, can be determined from the above equation if the elastic properties of the indenter tip are known.

Note that the accuracy of estimating the Young's modulus and hardness depends strongly on the accurate measurement of the contact area. Because material may pile up or sink in around the indenter, one may underestimate or overestimate the contact area. Moreover, due to the presence of the substrate, the load-indentation relation is the contribution from both the film and the substrate. Hence, the characterization of the mechanical properties of the film material from experimental data should be very careful. In order to avoid the substrate influence, Oliver and Pharr (1992) suggested that the indentation depth should be limited to a small fraction of the film thickness, typically about 10%, for the measurement of thin film properties.

Substrate curvature method

The substrate curvature measurement is a simple method to evaluate the stress and strain in a thin film attached to a substrate. In general, stresses arise in the film if there is misfit between the film and substrate. For example, a change in temperature produces residual stresses when the film and the substrate have different coefficients of thermal expansion. The stress in the film can be related to the substrate curvature through the well-known Stoney's equation (Freund and Suresh, 2003)

$$\sigma = \frac{E_s}{1 - \nu_s} \frac{t_s^2}{6t_f} \frac{1}{R}, \quad (2.9)$$

in which σ is the film stress, E_s and ν_s are the Young's modulus and Poisson's ratio of the substrate material, t_s is the thickness of the substrate, t_f is the thickness of the film and R is the radius of curvature of the substrate. This expression is valid if the substrate deforms elastically and the film has a uniform thickness with $t_f \ll t_s$. It is seen from the above equation that only the film thickness is needed, while the knowledge of the mechanical properties of film material is implicitly reflected in R .

2.1.3 Summary for experimental observations

The mechanical properties of material play an important role in accurately predicting and determining the mechanical response of devices in the stage of design and application. Of our interest, we summarize the available Young's modulus of copper thin film in Table 2.1 in chronological order. For other metallic films, such as aluminum, gold and nickel films, readers are referred to Connolley *et al.* (2005) and references cited here.

Table 2.1 The Young's modulus of polycrystalline copper films from some published experimental tests.

reference	Test method	Deposition method	Film thickness (μm)	Average grain size (μm)	$E(\text{GPa})$	remarks
Jämtning <i>et al.</i> (1997)	Biaxial bending	Sputter	0.45 $\pm$ 0.02	0.12	116 $\pm$ 8	Porosity, delamination
Suresh <i>et al.</i> (1999)	nanoindentation	E-beam evaporation	1.0	0.5 (1-2 grains through thickness)	126	60-80% <111> texture, with the rest exhibiting random orientation
Huang and Spaepen (2000)	Tensile test	E-beam evaporation	$\sim$ 3	on the order of thickness	102 $\pm$ 7	strong <111> texture
Volinsky <i>et al.</i> (2001)	Nanoindentation	Electroplating	0.2	$\sim$ 0.35	133	Porosity; surface roughness
	Nanoindentation	Electroplating	2	NA	110	
Fang and Chang (2003)	Nanoindentation	Sputter	0.25	NA	135	
Read <i>et al.</i> (2004)	Tensile test	Electrodeposited	2.59	1.5	73 $\pm$ 24	Strong <111> texture
Espinosa <i>et al.</i> (2004)	Membrane deflection experiment	E-beam evaporation	0.2-1.0	NA	125-129	
Zhou <i>et al.</i> (2004)	Microbridge	Electroplating	9.4	NA	115.2	Plane-strain modulus $E/(1-\nu^2)$
	Nanoindentation	Electroplating	9.4	NA	110 $\pm$ 1.67	
Xiang <i>et al.</i> (2006)	Plane-strain bulge test	Electroplating	3.0	NA	135 $\pm$ 5	
			1.8	NA	143 $\pm$ 4	
			0.9	NA	154 $\pm$ 5	
Wei <i>et al.</i> (2007)	Plane-strain bulge test	Evaporation	0.17-1		111-132	
Lin <i>et al.</i> (2010)	Tensile test	Sputter	0.5	NA	113	
			0.9	NA	117	

It is noticed that the experimental data listed in Table 2.1 are scattering in a wide range. Direct comparison among the values from different sources is difficult and meaningless because the material processing parameters, device fabrication parameters and test procedures vary from laboratory to laboratory. For instance, some tested films exhibit strong texture, while others have random orientation. Some films have only one grain through the film thickness, while others are composed of multiple grains in the direction of thickness. It is clear that the film microstructure does affect the mechanical properties. A systematic characterization of microstructure is necessary to correlate the microstructure-property relation.

Huang and Spaepen (2000) carried out a microtensile testing to measure the Young's modulus of the free-standing Cu, Ag and Al thin films and Au/Cu multilayers. The observation by X-ray diffraction indicated that all the thin films and multilayers were highly $\langle 111 \rangle$ textured. The grain size of the pure films was in the order of the thickness. The measured Young's moduli were on average 10-20% below the value estimated by averaging the anisotropic elastic constants. In their work, the influence of the texture, voids, dislocation microplasticity, reversible microplasticity, compliant grain boundaries and microcracking on the modulus variation was discussed. In view of their observed microstructural features, Huang and Spaepen (2000) concluded that compliant grain boundaries lead to the modulus reduction. Haque and Saif (2002b) also attributed the observed modulus deficit to compliant grain boundary in 30-50nm-thick freestanding gold and aluminum films.

Espinosa *et al.* (2004) used membrane deflection experiment to evaluate the elastic modulus of the free-standing polycrystalline Al, Au and Cu thin films. The measured elastic moduli were in the range of 53-55GPa for Au, 125-129GPa for Cu and 65-

70GPa for Al. It is interesting to notice that both the Cu and Al thin films have elastic modulus close to their bulk value, while the Au thin film has lower modulus than its bulk value. The modulus reduction was explained as the result of grain orientation distribution.

Yu and Spaepen (2004) studied the mechanical behavior of copper film with thickness between 0.1 and 3 μm by stretching a film/polyimide structure. About 20% modulus reduction was observed in contrast with the value of its bulk counterpart. The grain boundary cracks and dislocations were believed to be the source of the modulus reduction. Fang and Chang (2003) performed nanoindentation technique on 0.25 μm -thick copper attached on different substrates including Si, SiO₂ and LiNbO₃. They reported that the Young's modulus of the copper film on LiNbO₃ is the highest and that of the copper film on Si is the lowest.

Son *et al.* (2003) performed cantilever bending tests on Au films with thickness equal to 0.56, 0.99 and 1.26 μm , and measured the corresponding Young's modulus equal to 130.3 ± 6.1 , 108.7 ± 3.9 and $107.2 \pm 4.4\text{GPa}$, respectively. It was claimed by Son *et al.* (2003) that the Young's modulus increased with decreasing film thickness. They did not discuss the reason for the modulus difference. However, from their data of the grain size, we find that the 1.26 μm -thick film has columnar grain structure, while the thinner film has about 6 grains with the assumption of equiaxed grains. The modulus difference may be attributed to the grain interaction.

To understand the influence of the grain size, Haque and Saif (2004) performed tensile tests on 50nm-, 100nm-, 150nm- and 480nm-thick aluminum films with corresponding average grain sizes of 22, 50, 65, and 212nm, respectively. The measured elastic modulus was 63GPa for the smallest-grained (22nm) sample, 68GPa for 50-nm-

grained sample, and 70GPa for the larger-grained sample. Based on their experimental data, they suggested that the size effect on the elastic modulus only appears when the grain size is 50nm or less.

2.2 Analytical modeling

The effort of estimating the effective elastic properties of polycrystalline aggregates may be traced back at least sixty years ago (Hill, 1952). The polycrystalline aggregates are made up of grains with different crystallographic orientation, shape and size. Hence, the effective properties of the polycrystalline aggregates are significantly dependent on their microstructure. Unfortunately, it is almost impossible to set up an analytical model including all the microstructural information. To tackle this problem, various analytical models have been developed by introducing certain simplifications.

In the literature, the analytical approaches can be classified into two groups. The first group is called as “bound method” which provides the lower and upper bounds on the effective properties. The second group is “approximate method” which gives an approximate value for the effective properties. In the following subsections, we briefly review these two types of analytical methods. For much more detailed review, readers are referred to papers by Hashin (1983), Hirsekorn (1990), Adams and Olson (1998), and books by Mura (1987), Nemat-Nasser and Hori (1999), to mention a few.

2.2.1 Bound methods

There are two prominent pairs of lower and upper bounds, namely Voigt-Reuss bounds (VR bounds) and Hashin-Shtrikman bounds (HS bounds). They are often employed to validate the accuracy of the other analytical or numerical models.

Voigt-Reuss bounds

In the pioneering work of Hill (1952), the Voigt value and Reuss value for the effective elastic moduli, i.e., the bulk modulus and shear modulus, have been proved to be the upper and lower bounds on the basis of the classic principle of potential and complementary energies, respectively.

In the Voigt model, the strain is assumed to be identical for each grain in an aggregate. Such assumption leads to an upper bound for the effective elastic constants by averaging the elastic stiffness constants of each grain over all the possible crystallographic orientations. The effective bulk modulus and shear modulus, denoted by K_V and G_V respectively, are (Hill, 1952)

$$K_V = \frac{1}{9}[(C_{11} + C_{22} + C_{33}) + 2(C_{12} + C_{23} + C_{31})], \quad (2.10)$$

$$G_V = \frac{1}{15}[(C_{11} + C_{22} + C_{33}) - (C_{12} + C_{23} + C_{31}) + 3(C_{44} + C_{55} + C_{66})], \quad (2.11)$$

where C_{ij} are the elastic stiffness constants of single crystal in Voigt contracted notation. Note that the assumption of uniform strain in the Voigt model satisfies the compatibility of deformation, but breaks down the equilibrium condition among the grains.

In the Reuss model, each grain is assumed to be in the uniform stress state. A lower bound on the effective elastic constants is, thus, determined by averaging the elastic compliance constants of each grain over all the possible crystallographic orientations. The results from the Reuss model are (Hill, 1952)

$$1/K_R = [(S_{11} + S_{22} + S_{33}) + 2(S_{12} + S_{23} + S_{31})], \quad (2.12)$$

$$1 / G_R = \frac{1}{15} [4(S_{11} + S_{22} + S_{33}) - 4(S_{12} + S_{23} + S_{31}) + 3(S_{44} + S_{55} + S_{66})], \quad (2.13)$$

in which K_R and G_R denote the effective bulk modulus and shear modulus from the Reuss model, S_{ij} represent the elastic compliance constants of single crystal in Voigt contracted notation. It should be pointed out that the compatibility of deformation among the grains is violated, while the mechanical equilibrium condition is satisfied in the Reuss model.

It is seen that only nine elastic constants are included in the Voigt and Reuss results, whereas twenty-one independent elastic constants are required for the most general crystal structure. In the case of face-centered cubic (FCC) crystal or body-centered cubic (BCC) crystal, there are only three independent elastic constants. Thus, the corresponding effective bulk modulus and shear modulus can be recast into

$$K_V = K_R = \frac{1}{3} (C_{11} + 2C_{12}), \quad (2.14)$$

$$G_V = \frac{1}{5} [(C_{11} - C_{12}) + 3C_{44}], \quad (2.15)$$

$$1 / G_R = \frac{1}{5} [4(S_{11} - S_{12}) + 3S_{44}]. \quad (2.16)$$

Equation (2.14) indicates that the bulk modulus for a polycrystalline aggregate with cubic symmetry is uniquely determined from the elastic constants of single crystal. The difference in the shear modulus obtained from the Voigt model and Reuss model is a second order quantity as

$$G_V - G_R = \frac{3 [2C_{44} - (C_{11} - C_{12})]^2}{5 [4C_{44} + 3(C_{11} - C_{12})]}. \quad (2.17)$$

Clearly, the gap between the upper and lower bounds increases with increasing the grain anisotropy (see Eq. (3.8) for definition). Since no assumption of the grain morphology (i.e., the grain shape, size and distribution) is made in the Voigt and Reuss models, the Voigt and Reuss values must be the bounds for all the exact elastic constants.

Hashin-Shtrikman bounds

Hashin and Shtrikman (1962a) developed a pair of variational principles which can be used to determine the upper and lower bounds on the effective elastic properties of heterogeneous materials. Taking advantage of these principles, they derived a pair of improved upper and lower bounds by assuming that each grain has piecewise constant polarization strain or polarization stress (Hashin and Shtrikman, 1962b). For the polycrystalline aggregates with cubic symmetry, the effective bulk modulus is unique since the equivalent Voigt value coincides with the Reuss value. Therefore, only an explicitly improved pair of bounds on the effective shear modulus is determined by

$$G_{HS}^L = G_1 + 3 \left[\frac{5}{G_2 - G_1} + \frac{12(K + 2G_1)}{5G_1(3K + 4G_1)} \right]^{-1}, \quad (2.18)$$

$$G_{HS}^U = G_2 + 2 \left[\frac{5}{G_1 - G_2} + \frac{18(K + 2G_2)}{5G_2(3K + 4G_2)} \right]^{-1}, \quad (2.19)$$

where

$$K = \frac{1}{3}(C_{11} + 2C_{12}), G_1 = \frac{1}{2}(C_{11} - C_{12}), G_2 = C_{44}.$$

The symbols, G_{HS}^L and G_{HS}^U , denote the HS lower and upper bounds, respectively.

Although HS bounds are narrower than VR bounds, there is an implicit assumption of

“equiaxial crystals” (i.e. the grain shapes have no preferred direction and the crystallographic orientations are randomly distributed throughout the grains) which is aroused from the assumption that each grain has piecewise uniform stress or strain field.

Incorporating the assumption of piecewise constant polarization stress or strain into the classical variational principles, Walpole (1966b) derived identical bounds with those of Hashin and Shtrikman (1962b) for the effective elastic constants of polycrystals with cubic symmetry. Willis (1977, 1981) combined the Green’s function with variational principle of Hashin and Shtrikman (1962a, b) to obtained the lower and upper bounds of anisotropic composites. Willis’ results covered the solutions obtained by Hashin and Shtrikman (1962a, b; 1963) and Walpole (1966a, b; 1969) as special cases.

Moreover, Hashin and Shtrikman’s variational approach (1962a, b) has been extended to investigate the effective elastic properties of polycrystalline aggregates with hexagonal and trigonal crystal structure by Peselnick and Meister (1965), and tetragonal symmetry by Meister and Peselnick (1966), with corrections made later by Watt and Peselnick (1980). For the orthorhombic and monoclinic symmetry, the explicit lower and upper bounds were developed by Watt (1979, 1980).

2.2.2 Approximate methods

Hill’s average

The exact deformation in a polycrystalline aggregate is in between the uniform stress of the Reuss assumption and the uniform strain of the Voigt assumption. Hence, for

the polycrystalline aggregates with cubic symmetry, Hill (1952) suggested that the arithmetic or geometric mean of the Voigt and Reuss values is an approximation for the exact shear modulus. They are

$$G_{H-a} = \frac{1}{2}(G_V + G_R), \quad (2.20)$$

$$G_{H-g} = \sqrt{G_V G_R}, \quad (2.21)$$

where G_{H-a} and G_{H-g} represent the arithmetic and geometric mean value of the Voigt and Reuss values, respectively. This is the so-called Hill's average in the literature. For many cubic polycrystals, Hill's averages are in good agreement with the experimental values (Watt *et al.*, 1976).

Self-consistent scheme

The original self-consistent scheme (SCS) was independently proposed by Hershey and Kröner for predicting the effective elastic constants of polycrystalline aggregates (Christensen, 1979; Mura, 1987). Subsequently, Hill (1965) and Budiansky (1965) extended SCS to study the effective properties of multiphase material. The essential point of SCS is to treat each phase as an inhomogeneity embedded in a homogenized material with yet to be determined effective properties.

In the application of SCS to the polycrystalline aggregates, each anisotropic grain with a specific crystallographic orientation is regarded as a spherical or ellipsoidal inhomogeneity which is then imbedded in an equivalently homogenized polycrystalline aggregate with unknown effective isotropic properties. When a uniform strain is loaded at far-field, the strain and stress fields within the ellipsoidal grain are uniform (Eshelby, 1957). Averaging over all the possible orientations of a

single ellipsoidal grain produces the self-consistent expression for the macroscopic effective modulus.

For the polycrystalline aggregates with cubic symmetry, Kröner *et al.* (1966) presented a cubic equation (having only one positive root) for the effective shear modulus as

$$8G^{*3} + (5C_{11} + 4C_{12})G^{*2} - C_{44}(7C_{11} - 4C_{12})G^* - C_{44}(C_{11} - C_{12})(C_{11} + 2C_{12}) = 0, \quad (2.22)$$

where G^* is the effective shear modulus, C_{ij} are the elastic constants of single crystal. The solution was verified to be within the range of HS upper and lower bounds (Kröner *et al.*, 1966).

Based on Eshelby's inclusion solution (Eshelby, 1957) and the idea of SCS, Morris (1970, 1971) evaluated the effective modulus of the polycrystalline aggregates by considering the distribution of crystallographic orientation which is described by orientation distribution function (ODF). As an illustration, comparison was made between the results from his procedure and Hill's average. Morris' results were only a couple of percents different from Hill's average solution which is much easier to be derived.

In the work of Willis (1977, 1981), the essence of SCS and Hashin and Shtrikman's variational principle (Hashin and Shtrikman, 1962a, b) were integrated to investigate the effective elastic properties of the polycrystalline aggregates. For the polycrystalline aggregates with cubic symmetry, the estimated effective bulk modulus, denoted by K^* , is equal to the solution of Hashin and Shtrikman (1962b), i.e. $K^* = (C_{11} + 2C_{12})/3$, and a cubic equation containing the effective shear modulus is obtained as

$$8G^{*3} + (9K + 4G_1)G^{*2} - 3G_2(K + 4G_1)G^* - 6KG_1G_2 = 0, \quad (2.23)$$

where

$$K = \frac{1}{3}(C_{11} + 2C_{12}), G_1 = \frac{1}{2}(C_{11} - C_{12}), G_2 = C_{44}.$$

After some manipulations, Eq. (2.23) is equivalent to Eq. (2.22). Willis (1981) stressed that his solution directly followed the variational approach without reference to the grain shape, while Kröner (1966) restricted the grain shape to sphere. The identical estimation of the effective shear modulus for the polycrystals with cubic symmetry was claimed by Walpole (1981).

Recently, Berryman (2005) derived self-consistent approximation for the polycrystalline aggregates with hexagonal, trigonal, and tetragonal symmetries, based on his established compact formulae for the effective properties bounds in all the foregoing cases. His idea is similar to Willis' work (1977, 1981).

2.3 Numerical simulation

With the progress of computational techniques and capabilities, numerical simulations have been increasingly used in engineering and material science. Among these numerical methods, finite element method (FEM) is employed widely to investigate the mechanical behavior and properties of the polycrystalline aggregates. In the finite element simulation, the basic steps include: establishing a finite element model to capture the main microstructural feature of the polycrystalline aggregates, describing constitutive properties of the grains, and solving boundary value problem. A comprehensive discussion on the numerical simulation has been given by Cailletaud *et*

al. (2003) and Zohdi and Wriggers (2008). The following review is mainly concerned about the first step, i.e. to simulate the polycrystalline aggregates in the grain structure.

In general, there are two ways of representing the microstructural information in the finite element simulation. The first approach is to reconstruct the microstructure from the information gathered in the experimental measurement. The second one is to automatically generate a computational model to characterize the grain structure.

2.3.1 Reconstruction technique

To reconstruct the real microstructure in the finite element simulation, a systematic description of the material's microstructure is the prerequisite. For a polycrystalline aggregate, the microstructure is characterized by the grain shape, size, crystallographic orientation and spatial distribution, etc. Real grain microstructure can be measured using advanced material characterization techniques, such as serial sectioning, X-ray tomography, X-ray diffraction, and electron backscatter diffraction (EBSD) (Caillaud *et al.*, 2003; Sundararaghavan and Zabaras, 2005). A representation of the micro-morphology of polycrystal, i.e. the grain size, shape and distribution, can be provided by the conventional optical microscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), as well as X-ray tomography. The grain orientations can be accomplished using EBSD methods. These techniques are adopted by many investigators to completely characterize three-dimensional (3D) microstructure of a material (Groeber *et al.*, 2006; Lewis *et al.*, 2006; Lewis and Geltmacher; 2006; Bhandari *et al.*, 2007; Musienko *et al.*, 2007; Siddiq Qidwai *et al.*, 2009). In the absence of experimental instruments or a sufficiently detailed description of the microstructural information, an effective alternative is to employ limited statistical properties extracted from two-dimensional (2D) micrograph or

image. This procedure is referred to as “statistical reconstruction” because the statistical properties of the model are matched with those of a 2D image (Yeong and Torquato, 1998; Roberts and Garboczi, 1999; Groeber *et al.*, 2008a, b).

A series of contributions by Lewis and co-workers (Lewis *et al.*, 2006; Lewis and Geltmacher; 2006; Siddiq Qidwai *et al.*, 2009) addressed the application of EBSD-based orientation image microscopy (OIM) technique along with optical microscopy and serial sectioning to create a 3D image-based finite element model. Such model presents the detailed morphology and crystallographic orientation distribution. As an example, Lewis and Geltmacher (2006) collected the microstructural information by performing 10-20 sections per grain. The established finite element model contained 138 individual grains and twins with identified crystallographic orientation. The mechanical response in the grain level was examined to further identify the critical microstructural features in failure and fracture. Recently, Musienko *et al.* (2007) obtained a full 3D morphology and crystallographic orientation, which were further incorporated into the finite element model of the polycrystalline copper specimen, through a series of micropolishing operation and orientation image microscopy. Their numerical simulation was in good agreement with the experimental measurement of the global stress-strain behavior.

For the polycrystalline aggregates with columnar grain structure, the microstructural feature can be obtained by the real image in the film plane. Following this idea, den Toonder *et al.* (1999) digitized and reconstructed an actual microstructural image containing 56 grains into a finite element model for further study. Based on the established model, they predicted the effective Young’s modulus and Poisson’s ratio of polycrystalline BaTiO₃, In and ZrSiO₄ in the states of plane strain and plane stress,

respectively. Choi (2004) developed a lattice-based microstructure simulation scheme to model the process of microstructure evolution and eventually obtained the finite element model of a real material. This model was extended by Kim *et al.* (2007) to examine the effective elastic constants of polycrystalline silicon thin films at micro-scale. Kim and coworkers (2007) also used molecular dynamics simulation to study the mechanical behavior of nano-crystalline silicon. They suggested that the size effect should be taken into account only when the grain size is below 60Å.

According to the two-dimensional microstructural image of silver-tungsten composite, Roberts and Garboczi (1999) established a three-dimensional model whose statistical properties were in accordance with that from the real microstructural image. A good estimation of the effective modulus was reported in comparison with the experimental data. Subsequently, Sundararaghavan and Zabarar (2005) developed a statistical reconstruction technique to obtain 3D morphology by matching the microstructural information with machine learning technique. In their series of papers, Groeber *et al.* (2008a, b) extensively studied the statistical features of the three-dimensional grain-level microstructure of polycrystalline materials measured by the serial sectioning experiments. A framework to construct a finite element model with equivalently statistical characterization of the real microstructure was developed.

Obviously, the success in reconstructing the real microstructure into a finite element model depends on the application of various experimental methods mentioned above. However, these measurements are usually intricate and expensive. Moreover, complicated post-processing is needed to extract the useful data from the huge amount of the measured data. A further complication is the fact that one quantitative analysis of the structure only leads to one set of specific result.

2.3.2 Computer-generated microstructure

In order to avoid the drawbacks in the reconstruction technique, the method to automatically generate a computational model of material's microstructure is in favor for many researchers. In this case, some researchers assume all the grains have identical shape like cube, rhombic dodecahedron and truncated octahedron, and repeatedly tessellate the volume space with the selected shape (Mika and Dawson, 1998; Delannay *et al.*, 2006; Ranganathan and Ostoj-Starzewski, 2008; Ritz and Dawson, 2009). Noticeably, Voronoi tessellation is one of the most popular methods due to its topological similarity between Voronoi tessellation and the real microstructure of polycrystalline aggregates. It is a process of dividing a n-dimensional space into space-filling Voronoi cells with various sizes and shapes. Examples can be found in the works by Kumar (1992), Mullen *et al.* (1997), Barbe *et al.* (2001a, b), Weyer *et al.* (2002), Mahadevan and Zhao (2002), Zhao and Tryon (2004), Fritzen *et al.* (2009), to name a few.

Kumar (1992) discretized a unit cube into many Voronoi cells using Poisson-Voronoi tessellation. Each grain was simulated as a Voronoi cell. The effective moduli of the polycrystalline aggregates with different symmetry (i.e., cubic, hexagonal, trigonal and tetragonal symmetry) were calculated and compared with analytical solutions. By modeling the unit cube with different shapes of grains, he noticed that the discrepancy among the predicted elastic moduli is about one percent. Later, Kumar *et al.* (1996) applied the Voronoi model to investigate the stress distribution within the polycrystalline aggregates having 200 grains. Barbe *et al.* (2001a, b) characterized the intergranular and intragranular behavior of the polycrystalline aggregates represented by an assemblage of about 200 Voronoi cells.

In view of the microstructural feature of the polycrystalline thin film with columnar grain structure, many researchers simplify the film from three-dimensional case to two-dimensional case in the state of plane stress or plane strain. The planar Voronoi tessellation is then employed to simulate the grain structure of film. Mullen *et al.* (1997) simulated various (100) polycrystalline thin films with two-dimensional Voronoi tessellation. Under the assumption of plane strain condition, the authors related the crystal anisotropy and the number of grains to the effective Young's modulus and Poisson's ratio. With the help of planar Voronoi tessellation, Weyer and colleagues (2002) examined the influence of boundary conditions on the Young's modulus of polycrystalline Al_2O_3 in the state of plane strain and concluded that the predicted value from different boundary conditions converged to a single value with increasing the number of grains.

Although the substrate-attached thin film is the most common configuration in the microelectronic and MEMS devices, the study using the numerical simulation is obviously lagged behind the experimental study. Most attention has been given to characterize the local stress distribution and plastic behavior of the attached film (Wikström and Nygåards, 2002; Šiška *et al.*, 2007; Shen, 2008, 2009). Wikström and Nygåards (2002) elaborated the distribution of stress in the grains with different orientation by simulating a textured Cu thin film deposited on the polyimide and silicon. Similar research has been reported by Šiška *et al.* (2007). It is noted that the elastic modulus for the substrate-attached thin film has received little attention in the literature.

Chapter 3 Analytical Considerations

Analytical considerations in predicting the effective elastic properties of polycrystalline aggregates are reviewed in this chapter. They are Voigt and Reuss bounds, Hill's average and self-consistent scheme. In view of the unique geometry of columnar grained thin film, we present the derivation of Voigt upper and Reuss lower bounds for the effective elastic constants of polycrystalline aggregates in the state of plane stress.

3.1 Elastic anisotropy

For linear elastic materials, the relationship between the local stress and the local strain is described by the generalized Hooke's law in tensor notation as (Nye, 1985)

$$\boldsymbol{\sigma} = \mathbf{C}\boldsymbol{\varepsilon} \text{ or } \boldsymbol{\varepsilon} = \mathbf{S}\boldsymbol{\sigma}, \quad (3.1)$$

or in index notation as

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl} \text{ or } \varepsilon_{ij} = S_{ijkl}\sigma_{kl} \quad (i, j, k, l = 1, 2, 3), \quad (3.2)$$

where $\mathbf{C}$ (or C_{ijkl}) and $\mathbf{S}$ (or S_{ijkl}) are the fourth-order elastic stiffness and compliance tensors, $\boldsymbol{\sigma}$ (or σ_{ij}) and $\boldsymbol{\varepsilon}$ (or ε_{ij}) are the second-order stress and strain tensors, respectively. Here and from now on, the lowercase Latin subscripts have the range of 1, 2, 3 and the summation convention on repeated indices is implicitly applied.

Alternatively, it is sometimes convenient to express the generalized Hooke's law in matrix form (i.e. in Voigt convention) as

$$\sigma_I = C_{IJ} \varepsilon_J \quad \text{or} \quad \varepsilon_I = S_{IJ} \sigma_J \quad (I, J = 1, 2, \dots, 6), \quad (3.3)$$

where σ_I and ε_I are 6×1 column matrices, C_{IJ} and S_{IJ} are 6×6 matrices called the elastic stiffness and compliance matrices, respectively. Note the uppercase Latin indices take the value from 1 to 6. The rules listed in Table 3.1 are used to transform the tensor components to the matrix components and vice versa (Nye, 1985).

Table 3.1 Transformation between tensor indices and Voigt matrix indices.

Tensor notation	11	22	33	23,32	31,13	12,21
Matrix notation	1	2	3	4	5	6

Hence, the components of tensors σ_{ij} and ε_{ij} are related to those of matrices σ_I and ε_I by

$$\begin{bmatrix} \sigma_{11} & \sigma_{12} & \sigma_{13} \\ \sigma_{21} & \sigma_{22} & \sigma_{23} \\ \sigma_{31} & \sigma_{32} & \sigma_{33} \end{bmatrix} \rightarrow \begin{bmatrix} \sigma_1 & \sigma_6 & \sigma_5 \\ \sigma_6 & \sigma_2 & \sigma_4 \\ \sigma_5 & \sigma_4 & \sigma_3 \end{bmatrix}, \begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{bmatrix} \rightarrow \begin{bmatrix} \varepsilon_1 & \frac{1}{2}\varepsilon_6 & \frac{1}{2}\varepsilon_5 \\ \frac{1}{2}\varepsilon_6 & \varepsilon_2 & \frac{1}{2}\varepsilon_4 \\ \frac{1}{2}\varepsilon_5 & \frac{1}{2}\varepsilon_4 & \varepsilon_3 \end{bmatrix}. \quad (3.4)$$

The transformation between the stiffness tensor C_{ijkl} and matrix C_{IJ} can also be performed according to the aforementioned rules. But it is not the case when transforming the compliance tensor S_{ijkl} into matrix S_{IJ} . Some numerical factors have to be introduced, i.e.,

$$S_{ijkl} = S_{IJ} \quad \text{when } I \text{ and } J \text{ are 1, 2, or 3 ,}$$

$$S_{ijkl} = S_{IJ} / 2 \quad \text{when } I \text{ either } J \text{ are 4, 5, or 6 ,}$$

$$S_{ijkl} = S_{IJ} / 4 \quad \text{when both } I \text{ and } J \text{ are 4, 5, or 6 .}$$

Considering the symmetry of the stress and strain tensors and the existence of strain energy, one has the fourth-order tensors $\mathbf{C}$ (or C_{ijkl}) and $\mathbf{S}$ (or S_{ijkl}) with up to 21 independent elastic constants. This is the case of triclinic crystal, the most general anisotropic material. The existence of a higher degree of crystal symmetry can further reduce the number of independent elastic constants to 3~13. For a face-centered cubic (FCC) crystal (e.g., copper and aluminum), only three independent elastic constants are needed, namely C_{11} , C_{12} and C_{44} (or S_{11} , S_{12} and S_{44}). The corresponding elastic stiffness and compliance matrices for the cubic crystal are given by

$$[C_{IJ}] = \begin{bmatrix} C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{11} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{12} & C_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{44} \end{bmatrix}, \quad (\text{a})$$

(3.5)

$$[S_{IJ}] = \begin{bmatrix} S_{11} & S_{12} & S_{12} & 0 & 0 & 0 \\ S_{12} & S_{11} & S_{12} & 0 & 0 & 0 \\ S_{12} & S_{12} & S_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & S_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & S_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & S_{44} \end{bmatrix}, \quad (\text{b})$$

where

$$S_{11} = \frac{C_{11} + C_{12}}{(C_{11} - C_{12})(C_{11} + 2C_{12})}, S_{12} = \frac{-C_{12}}{(C_{11} - C_{12})(C_{11} + 2C_{12})}, S_{44} = \frac{1}{C_{44}}.$$

Note that the foregoing elastic constants are referred to the crystal coordinate system, whose coordinate axes coincide with the orthogonal crystallographic directions of the single crystal.

Figure 3.1 The relation between global coordinate system $O-XYZ$ and crystal coordinate system $O-xyz$. The line N is the intercept line of planes xy and XY . Euler angles (α, β, γ) describe the relation between these two coordinate systems.

When the crystal coordinate system $O-xyz$ does not coincide with a global coordinate system $O-XYZ$, shown in Fig. 3.1, the elastic constants for the crystal in terms of the global coordinate system may be obtained through a transformation, i.e.,

$$C_{ijkl}^g = t_{im}t_{jn}t_{kp}t_{lq}C_{mnpq}, \quad (\text{a}) \quad (3.6)$$

$$S_{ijkl}^g = t_{im}t_{jn}t_{kp}t_{lq}S_{mnpq}, \quad (\text{b})$$

where the superscript g is referred to the global coordinate system, t_{ij} are the components of the transformation matrix. Here, Euler angles (α, β, γ) are used to describe the relation between these two sets of coordinate system, as illustrated in Fig. 3.1. Then, the coordinate transformation matrix can be calculated by

$$(t_{ij}) = \begin{bmatrix} \cos \alpha \cos \beta \cos \gamma - \sin \alpha \sin \gamma & -\cos \alpha \cos \beta \sin \gamma - \sin \alpha \cos \gamma & \cos \alpha \sin \beta \\ \sin \alpha \cos \beta \cos \gamma + \cos \alpha \sin \gamma & -\sin \alpha \cos \beta \sin \gamma + \cos \alpha \cos \gamma & \sin \alpha \sin \beta \\ -\sin \beta \cos \gamma & \sin \beta \sin \gamma & \cos \beta \end{bmatrix}, \quad (3.7)$$

where $\alpha \in [0, 2\pi]$, $\beta \in [0, \pi]$ and $\gamma \in [0, 2\pi]$.

In general, most crystalline materials exhibit anisotropic properties. For the cubic crystals, the degree of anisotropy may be described by the so-called anisotropic ratio (AR) which is given by (Nye, 1985)

$$AR = \frac{2C_{44}}{C_{11} - C_{12}} = \frac{2(S_{11} - S_{12})}{S_{44}}. \quad (3.8)$$

If the AR is equal to 1, the mechanical behavior of such material is isotropic and the associated elastic properties are independent of the crystallographic orientation, such as tungsten. The higher the AR is, the stronger the anisotropy is.

3.2 Effective elastic properties of polycrystalline aggregates

For statistically homogeneous heterogeneous materials like the composite materials and polycrystalline aggregates, the associated effective elastic properties are defined by (Hill, 1963; Hashin, 1983; Mura, 1987)

$$\bar{\sigma}_{ij} = C_{ijkl}^* \bar{\varepsilon}_{kl} \text{ or } \bar{\varepsilon}_{ij} = S_{ijkl}^* \bar{\sigma}_{kl}, \quad (3.9)$$

where C_{ijkl}^* and S_{ijkl}^* are the fourth-order effective elastic stiffness and compliance tensors, $\bar{\sigma}_{ij}$ and $\bar{\varepsilon}_{ij}$ are the volume averages of the stress and strain tensors over all the constituent phases, such as each grain in a polycrystalline aggregate. Denoting the volume average by angle bracket, we have

$$\bar{\sigma}_{ij} = \langle \sigma_{ij} \rangle \quad \text{and} \quad \bar{\varepsilon}_{ij} = \langle \varepsilon_{ij} \rangle .$$

Under a homogeneous displacement boundary condition given by

$$u_i(S) = \varepsilon_{ij}^0 x_j, \quad (3.10)$$

or a homogeneous traction boundary condition given by

$$T_i(S) = \sigma_{ij}^0 n_j, \quad (3.11)$$

the corresponding volume averages of the stress and strain tensors can be determined from the average theorem as (Mura, 1987; Nemat-Nasser and Hori, 1999)

$$\bar{\sigma}_{ij} = \sigma_{ij}^0 \quad \text{or} \quad \bar{\varepsilon}_{ij} = \varepsilon_{ij}^0 . \quad (3.12)$$

In the foregoing equations, σ_{ij}^0 and ε_{ij}^0 are the uniform stress and strain tensors prescribed on the boundary S of the volume. Obviously, when the displacement boundary condition, i.e. Eq. (3.10), is prescribed, to determine the effective stiffness tensor C_{ijkl}^* , the average stresses $\bar{\sigma}_{ij}$ should be computed. On the other hand, with the imposed traction boundary condition, i.e. Eq. (3.11), to determine the effective compliance tensor S_{ijkl}^* , the average strains $\bar{\varepsilon}_{ij}$ should be computed.

Due to the fact that different grains in a polycrystalline aggregate have different crystallographic orientations, the response to the applied loading differs from grain to grain. In the case of displacement boundary condition, i.e. Eq. (3.10), the induced strains in the grain r , denoted by ε_{ij}^r , can be related to the average strains $\bar{\varepsilon}_{ij}$ by strain concentration tensor A_{ijkl}^r as

$$\boldsymbol{\varepsilon}_{ij}^r = A_{ijkl}^r \bar{\boldsymbol{\varepsilon}}_{kl} = A_{ijkl}^r \boldsymbol{\varepsilon}_{kl}^0 . \quad (3.13)$$

Similarly, in the case of traction boundary condition, i.e. Eq. (3.11), the induced stresses in the grain r , denoted by σ_{ij}^r , can be related to the average stresses $\bar{\sigma}_{ij}$ by stress concentration tensor B_{ijkl}^r as

$$\sigma_{ij}^r = B_{ijkl}^r \bar{\sigma}_{kl} = B_{ijkl}^r \sigma_{kl}^0 . \quad (3.14)$$

It follows directly from Eqs. (3.13) and (3.14) that the volume averages of the strain and stress concentration tensors correspond to the fourth-order identity tensor $\mathbf{I}$. They are

$$\langle A_{ijkl}^r \rangle = \langle B_{ijkl}^r \rangle = I_{ijkl} , \quad (3.15)$$

where $I_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$. δ_{ij} denote the Kronecker symbol.

In combination with the constitutive relation associated with the grain r , which is dependent on its crystallographic orientation and represented by the generalized Hooke's law as

$$\sigma_{ij}^r = C_{ijkl}^r \varepsilon_{kl}^r \quad \text{or} \quad \varepsilon_{ij}^r = S_{ijkl}^r \sigma_{kl}^r ,$$

it is readily determined the fourth-order effective elastic stiffness tensor C_{ijkl}^* and compliance tensor S_{ijkl}^* by the following averages in tensor notation

$$\mathbf{C}^* = \langle \mathbf{C}^r \mathbf{A}_r \rangle , \quad (3.16)$$

$$\mathbf{S}^* = \langle \mathbf{S}^r \mathbf{B}_r \rangle . \quad (3.17)$$

Note that Eqs. (3.16) and (3.17) are the exact solutions for the effective elastic properties of the polycrystalline aggregates. Because the concentration tensors, $\mathbf{A}_r$ and $\mathbf{B}_r$, depend on the anisotropic behavior of the grains and the microstructure of the polycrystalline aggregates, it is very difficult to find out their explicit expressions. To overcome such difficulties, various simplifications and assumptions are introduced. Once these concentration tensors are determined, the effective stiffness tensor $\mathbf{C}_{ijkl}^*$ and compliance tensor $\mathbf{S}_{ijkl}^*$ can be computed from Eqs. (3.16) and (3.17).

3.2.1 Revisiting Voigt and Reuss approximations

The Voigt approximation is determined under the assumption that all the grains aggregating the polycrystal have a uniform strain. From Eq. (3.13), it is readily seen that the strain concentration tensor, $\mathbf{A}_r$, is equal to the identity tensor $\mathbf{I}$ for all the grains. And following Eq. (3.16), the Voigt value for the effective elastic stiffness tensor $\mathbf{C}^*$ is

$$\mathbf{C}^* = \langle \mathbf{C}^r \rangle. \quad (3.18)$$

For a polycrystalline aggregate with macroscopically isotropic properties, two independent elastic constants, such as a pair of bulk modulus and shear modulus or a pair of Young's modulus and Poisson's ratio, are needed. Taking the bulk and shear moduli as example and from Eq. (3.18), the Voigt approximation for the effective bulk modulus K_V is obtained by assuming all the grains have the same dilatation strain ε_{ii}^0 as (Mura, 1987)

$$K_V = \frac{C_{iii}}{9}. \quad (3.19)$$

The Voigt approximation for the effective shear modulus G_V is obtained by assuming all the grains have the same shear strain ε_{12}^0 as (Mura, 1987)

$$G_V = \frac{1}{8\pi^2} \int_0^{2\pi} d\alpha \int_0^\pi \sin \beta d\beta \int_0^{2\pi} C_{1212}^g d\gamma, \quad (3.20)$$

where C_{1212}^g is the elastic constant of the crystal with respect to the global coordinate system O -XYZ, as shown in Fig. 3.1. It is a function of Euler angles (α, β, γ) and related to the single crystal elastic constants C_{ijkl} through Eq. (3.6a).

Correspondingly, from the uniform stress assumption in the Reuss model, the effective elastic compliance tensor $\mathbf{S}^*$ is

$$\mathbf{S}^* = \langle \mathbf{S}^r \rangle \quad (3.21)$$

with the stress concentration tensor $\mathbf{B}_r = \mathbf{I}$ for each of the grains.

The Reuss approximations for the effective bulk modulus K_R and shear modulus G_R are calculated, respectively, by

$$K_R = \frac{1}{S_{ijij}} = \frac{C_{ijij}}{9}, \quad (3.22)$$

$$G_R = \left[\frac{1}{2\pi^2} \int_0^{2\pi} d\alpha \int_0^\pi \sin \beta d\beta \int_0^{2\pi} S_{1212}^g d\gamma \right]^{-1}, \quad (3.23)$$

where S_{1212}^g is referred to the global coordinate system O -XYZ. It is computed from Eq. (3.6b) in terms of Euler angles (α, β, γ) .

3.2.2 Hill's average

Hill (1952) proved that the Voigt and Reuss values are the upper and lower bounds of the effective elastic constants, respectively. They are

$$K_R \leq K \leq K_V, \quad (3.24)$$

$$G_R \leq G \leq G_V, \quad (3.25)$$

where K_R , K_V , G_R and G_V are given, respectively, in Eqs.(2.14)-(2.16) for the polycrystalline aggregates with cubic symmetry.

Based on the relationship between the elastic constants, the Young's modulus E can be expressed in terms of the bulk modulus K and the shear modulus G by

$$\frac{1}{E} = \frac{1}{3G} + \frac{1}{9K}. \quad (3.26)$$

Hill (1952) pointed out that the effective Young's modulus also has lower bound E_R and upper bound E_V , expressed as

$$E_R \leq E \leq E_V, \quad (3.27)$$

where

$$\frac{1}{E_R} = \frac{1}{3G_R} + \frac{1}{9K_R} \quad \text{and} \quad \frac{1}{E_V} = \frac{1}{3G_V} + \frac{1}{9K_V}.$$

Following by Hill's suggestion that the arithmetic average of the Voigt value and Reuss values is a good approximation for the effective elastic constants, we have

$$K_H = (K_R + K_V) / 2, \quad (3.28)$$

$$G_H = (G_R + G_V) / 2, \quad (3.29)$$

where K_H and G_H correspond to the Hill's average of the effective bulk modulus and shear modulus, respectively. Furthermore, the Hill's averages of Young's modulus E_H and Poisson's ratio ν_H are given by

$$\frac{1}{E_H} = \frac{1}{3G_H} + \frac{1}{9K_H}, \quad (3.30)$$

$$\nu_H = \frac{1}{2} \left(\frac{3K_H - 2G_H}{3K_H + G_H} \right). \quad (3.31)$$

3.2.3 Self-consistent scheme

The self-consistent scheme has been used to study the effective mechanical properties for both the composite materials (Hill, 1965; Budiansky, 1965) and polycrystalline aggregates (Kröner *et al.*, 1966; Morris, 1970, 1971; Willis, 1977, 1981). To deal with the statistically homogeneous and isotropic polycrystalline aggregates, each grain is viewed as an inhomogeneity embedded in an homogeneous medium (consisting of all the other grains) with unknown effective stiffness tensor $\mathbf{C}^*$ (or compliance tensor $\mathbf{S}^*$). This treatment means that every grain in the aggregate is on the equal footing.

Clearly, once the strain concentration tensor $\mathbf{A}_r$ is known as a function of the effective stiffness tensor $\mathbf{C}^*$, we can solve Eq. (3.16) for the effective stiffness tensor $\mathbf{C}^*$. Similarly, if the stress concentration tensor $\mathbf{B}_r$ is known as a function of the effective compliance tensor $\mathbf{S}^*$, we can solve Eq. (3.17) for the effective compliance tensor $\mathbf{S}^*$. To determine the concentration tensors $\mathbf{A}_r$ or $\mathbf{B}_r$ associated with the grain r , one convenient way is to assume each grain as a spherical or ellipsoidal inhomogeneity embedded in medium with the yet unknown effective stiffness tensor $\mathbf{C}^*$ or compliance tensor $\mathbf{S}^*$.

When a homogenous displacement boundary condition given by Eq. (3.10) is imposed on a polycrystal, following the framework of Eshelby's equivalent inclusion method (Eshelby, 1957) and after some manipulations, the induced strain $\boldsymbol{\varepsilon}^r$ in the inhomogeneity (corresponding to the grain r) with stiffness $\mathbf{C}^r$ can be related to $\boldsymbol{\varepsilon}^0$ by Eq. (3.13). The strain concentration tensor $\mathbf{A}_r$ is

$$\mathbf{A}_r = [\mathbf{I} + \mathbf{S}_r^* \mathbf{C}^{*-1} (\mathbf{C}^r - \mathbf{C}^*)]^{-1}. \quad (3.32)$$

The superscript '-1' denotes the inverse operation. The fourth-order tensor $\mathbf{S}_r^*$ is the Eshelby's tensor computed based on $\mathbf{C}^*$. For a spherical inhomogeneity, the Eshelby's tensor $\mathbf{S}_r^*$ is given by

$$(\mathbf{S}_r^*)_{ijkl} = \frac{5\nu^* - 1}{15(1 - \nu^*)} \delta_{ij} \delta_{kl} + \frac{4 - 5\nu^*}{15(1 - \nu^*)} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \quad (3.33)$$

where ν^* is the effective Poisson's ratio of the polycrystalline material. By substituting Eq. (3.32) into Eq. (3.16), we can obtain the effective stiffness tensor $\mathbf{C}^*$ by

$$\mathbf{C}^* = \langle \mathbf{C}^r \mathbf{A}_r \rangle = \langle \mathbf{C}^r [\mathbf{I} + \mathbf{S}_r^* \mathbf{C}^{*-1} (\mathbf{C}^r - \mathbf{C}^*)]^{-1} \rangle. \quad (3.34)$$

Similarly, if a homogenous traction boundary condition given by Eq. (3.11) is imposed on a polycrystal, after the identical procedure, we may determine the stress concentration tensor $\mathbf{B}_r$ as

$$\mathbf{B}_r = [\mathbf{S}_r - \mathbf{S}_r^* (\mathbf{S}_r - \mathbf{S}^*)]^{-1} \mathbf{S}^*. \quad (3.35)$$

Thus, we can obtain the effective compliance tensor $\mathbf{S}^*$ by substituting Eq. (3.35) into Eq. (3.17) which is recast as

$$\mathbf{S}^* = \langle \mathbf{S}^r [\mathbf{S}^r - \mathbf{S}_r^* (\mathbf{S}^r - \mathbf{S}^*)]^{-1} \mathbf{S}^* \rangle. \quad (3.36)$$

Apparently, an iterative procedure is needed to evaluate the effective stiffness tensor $\mathbf{C}^*$ from Eq. (3.34) or the effective compliance tensor $\mathbf{S}^*$ from Eq. (3.36). As an example, the following procedure can be implemented to approximate the effective stiffness tensor $\mathbf{C}^*$.

Assume that the distribution of crystallographic orientation in the polycrystal is uniformly distributed in the Euler angle space. Thus, the volume average value at the right hand of Eq. (3.34) can be calculated by

$$\langle \mathbf{C}^r \mathbf{A}_r \rangle = \frac{1}{8\pi^2} \int_0^{2\pi} d\alpha \int_0^\pi \sin \beta d\beta \int_0^{2\pi} \{ \mathbf{C}^r [I + \mathbf{S}_r^* \mathbf{C}^{*-1} (\mathbf{C}^r - \mathbf{C}^*)]^{-1} \} d\gamma, \quad (3.37)$$

where (α, β, γ) are the Euler angles defining the crystallographic orientation of the grain r with respect to the global coordinate system. The Hill's arithmetic average (Hill, 1952) is taken as the first trial value of $\mathbf{C}^*$. Then, the average value $\langle \mathbf{C}^r \mathbf{A}_r \rangle$, given in Eq. (3.37), may be estimated through 32-point Gaussian quadrature over each variable in (α, β, γ) . The obtained value corresponds to the new approximation of the effective stiffness tensor $\mathbf{C}^*$. In the iterative process, if the relative error for each component between the $(n+1)^{\text{th}}$ estimation $(\mathbf{C}^*)^{n+1}$ and the n^{th} estimation $(\mathbf{C}^*)^n$, namely $|((\mathbf{C}_{ijkl}^*)^{n+1} - (\mathbf{C}_{ijkl}^*)^n) / (\mathbf{C}_{ijkl}^*)^n|$, is less than the prescribed tolerance (i.e., 10^{-4}), the $(n+1)^{\text{th}}$ estimation $(\mathbf{C}^*)^{n+1}$ is the required effective stiffness tensor $\mathbf{C}^*$.

3.3 Voigt and Reuss bounds in the state of plane stress

The analytical derivations reviewed in the previous section are originally developed for studying the polycrystalline aggregates in bulk form. They may not be applicable to characterize the mechanical properties of the polycrystalline aggregates in other geometrical configurations, such as the thin film and thin wire. The film may have a few or even one grain in the film thickness, while the grains spanning the cross-section of the thin wire are much less than those along the wire length. Although the Voigt-Reuss bounds cover all the possibilities of the effective moduli of the polycrystalline aggregates, the gap could be sufficiently large if the crystallites exhibit high anisotropic properties.

Hereby, the Voigt and Reuss models will be extended to evaluate the upper and lower bounds on the effective elastic constants of the polycrystalline aggregates in the state of plane stress. It is motivated when studying the mechanical behavior of polycrystalline thin film with columnar grain structure. When the ratio between the film thickness t and the grain size d in the film plane vanishes, the film can be approximated as in the state of plane stress.

Voigt approximation

For each grain in the film, the stress components perpendicular to the surface of the film vanish, that is

$$\sigma_3 = \sigma_4 = \sigma_5 = 0, \quad (3.38)$$

where $\sigma_3 = \sigma_{33}$, $\sigma_4 = \sigma_{23}$ or σ_{32} , and $\sigma_5 = \sigma_{13}$ or σ_{31} .

Hence, the nonzero stress components, namely $\sigma_1(=\sigma_{11})$, $\sigma_2(=\sigma_{22})$ and $\sigma_6(=\sigma_{12}$ or $\sigma_{21})$, can be related to the strain components in the film plane, namely $\varepsilon_1(=\varepsilon_{11})$, $\varepsilon_2(=\varepsilon_{22})$ and $\varepsilon_6(=2\varepsilon_{12}$ or $2\varepsilon_{21})$, in the matrix notation by

$$\begin{Bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_6 \end{Bmatrix} = \begin{bmatrix} C_{11}^{p,\sigma} & C_{12}^{p,\sigma} & C_{16}^{p,\sigma} \\ C_{21}^{p,\sigma} & C_{22}^{p,\sigma} & C_{26}^{p,\sigma} \\ C_{61}^{p,\sigma} & C_{62}^{p,\sigma} & C_{66}^{p,\sigma} \end{bmatrix} \begin{Bmatrix} \varepsilon_1 \\ \varepsilon_2 \\ \varepsilon_6 \end{Bmatrix}. \quad (3.39)$$

Note that the components in the elastic stiffness matrix, $C_{IJ}^{p,\sigma}$ ($I, J=1, 2$ and 6), is determined by using Eq. (3.6) and taking into account of the plane stress conditions given in Eq. (3.38). Each component is the function of Euler angles (α, β, γ). The superscript, p,σ , denotes the plane stress condition.

According to the Voigt assumption, the upper bound for the effective in-plane bulk modulus is determined by imposing

$$\varepsilon_1 = \varepsilon_2 = \varepsilon; \varepsilon_6 = 0. \quad (3.40)$$

Thus, the average stress components, $\langle \sigma_1 \rangle$ and $\langle \sigma_2 \rangle$, over all the grains can be determined by

$$\begin{aligned} \langle \sigma_1 \rangle &= \frac{\varepsilon}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^{2\pi} (C_{11}^{p,\sigma} + C_{12}^{p,\sigma}) \sin \beta d\alpha d\beta d\gamma, & (a) \\ \langle \sigma_2 \rangle &= \frac{\varepsilon}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^{2\pi} (C_{21}^{p,\sigma} + C_{22}^{p,\sigma}) \sin \beta d\alpha d\beta d\gamma. & (b) \end{aligned} \quad (3.41)$$

Considering the definition of the in-plane bulk modulus $\tilde{K}$, which is (Nemat-Nasser and Hori, 1999)

$$\tilde{K} = \frac{(\sigma_1 + \sigma_2)/2}{\varepsilon_1 + \varepsilon_2}, \quad (3.42)$$

we may obtain the upper bound for the effective in-plane bulk modulus $\tilde{K}_V^{p,\sigma}$ by the following equation. It is

$$\tilde{K}_V^{p,\sigma} = \frac{(\langle \sigma_1 \rangle + \langle \sigma_2 \rangle)/2}{\varepsilon_1 + \varepsilon_2}. \quad (3.43)$$

To determine the upper bound for the effective in-plane shear modulus, denoted by $\tilde{G}_V^{p,\sigma}$, the following conditions are imposed,

$$\varepsilon_1 = \varepsilon_2 = 0; \varepsilon_6 = \varepsilon. \quad (3.44)$$

According to Eqs. (3.39) and (3.44), the average shear stress over all the grains is then determined by

$$\langle \sigma_6 \rangle = \frac{\varepsilon}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^\pi C_{66}^{p,\sigma} \sin \beta d\alpha d\beta d\gamma. \quad (3.45)$$

Hence, the upper bound for the effective in-plane shear modulus, defined as

$$\tilde{G}_V^{p,\sigma} = \frac{\langle \sigma_6 \rangle}{\varepsilon_6}, \quad (3.46)$$

can be obtained by substituting Eqs. (3.44) and (3.45) into Eq. (3.46).

Reuss approximation

Alternatively, in the state of plane stress, the in-plane stress-strain relation may be recast in terms of the elastic compliance matrix as

$$\begin{Bmatrix} \varepsilon_1 \\ \varepsilon_2 \\ \varepsilon_6 \end{Bmatrix} = \begin{bmatrix} S_{11}^{p,\sigma} & S_{12}^{p,\sigma} & S_{16}^{p,\sigma} \\ S_{21}^{p,\sigma} & S_{22}^{p,\sigma} & S_{26}^{p,\sigma} \\ S_{61}^{p,\sigma} & S_{62}^{p,\sigma} & S_{66}^{p,\sigma} \end{bmatrix} \begin{Bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_6 \end{Bmatrix}. \quad (3.47)$$

Note that the components in the elastic compliance matrix, $S_{IJ}^{p,\sigma}$ ($I, J=1, 2$ and 6), are determined by Eq. (3.6b) in terms of Euler angles (α, β, γ) .

According to the assumption of the Reuss model, the following conditions are prescribed to each grain when determining the lower bound for the effective in-plane bulk modulus denoted by $\tilde{K}_R^{p,\sigma}$,

$$\sigma_1 = \sigma_2 = \sigma; \sigma_6 = 0. \quad (3.48)$$

Then, we have

$$\tilde{K}_R^{p,\sigma} = \frac{(\sigma_1 + \sigma_2)/2}{\langle \varepsilon_1 \rangle + \langle \varepsilon_2 \rangle}, \quad (3.49)$$

in which

$$\begin{aligned} \langle \varepsilon_1 \rangle &= \frac{\sigma}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^{2\pi} (S_{11}^{p,\sigma} + S_{12}^{p,\sigma}) \sin \beta d\alpha d\beta d\gamma, & (a) \\ \langle \varepsilon_2 \rangle &= \frac{\sigma}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^{2\pi} (S_{21}^{p,\sigma} + S_{22}^{p,\sigma}) \sin \beta d\alpha d\beta d\gamma. & (b) \end{aligned} \quad (3.50)$$

To determine the lower bound for the effective in-plane shear modulus designated by $\tilde{G}_R^{p,\sigma}$, we suppose each grain has identical stress state as

$$\sigma_1 = \sigma_2 = 0; \sigma_6 = \sigma. \quad (3.51)$$

Thus, we have

$$\tilde{G}_R^{p,\sigma} = \frac{\sigma_6}{\langle \varepsilon_6 \rangle}, \quad (3.52)$$

in which

$$\langle \varepsilon_6 \rangle = \frac{\sigma}{8\pi^2} \int_0^{2\pi} \int_0^\pi \int_0^{2\pi} S_{66}^{p,\sigma} \sin \beta d\alpha d\beta d\gamma. \quad (3.53)$$

Clearly, to evaluate the lower and upper bounds from the foregoing equations, numerical integration is needed. Furthermore, we may determine the lower and upper bounds for the effective in-plane Young's modulus $\tilde{E}$, which can be expressed in terms of the in-plane bulk modulus $\tilde{K}$ and in-plane shear modulus $\tilde{G}$ by

$$\tilde{E} = \frac{4\tilde{K}\tilde{G}}{\tilde{K} + \tilde{G}}. \quad (3.54)$$

The lower and upper bounds are

$$\tilde{E}_R^{p,\sigma} \leq \tilde{E}^{p,\sigma} \leq \tilde{E}_V^{p,\sigma}, \quad (3.55)$$

where

$$\tilde{E}_R^{p,\sigma} = \frac{4\tilde{K}_R^{p,\sigma} \tilde{G}_R^{p,\sigma}}{\tilde{K}_R^{p,\sigma} + \tilde{G}_R^{p,\sigma}}, \quad \tilde{E}_V^{p,\sigma} = \frac{4\tilde{K}_V^{p,\sigma} \tilde{G}_V^{p,\sigma}}{\tilde{K}_V^{p,\sigma} + \tilde{G}_V^{p,\sigma}}. \quad (3.56)$$

As an example, we consider polycrystalline copper in the state of plane stress. The corresponding elastic constants of single crystal are given in Table 4.1. Based on the above expressions and after some manipulations, the upper and lower bounds for the effective in-plane Young's modulus are

$$109.4 \text{ GPa} \leq \tilde{E}^{p,\sigma} \leq 131.3 \text{ GPa}.$$

Chapter 4 Key Issues in the Numerical Simulation

Finite element simulation with commercial software ANSYS10.0 is employed to investigate the effective Young's modulus of polycrystalline aggregates in various geometric configurations. In this chapter, a framework of the finite element scheme is described. To determine the effective elastic constants of the polycrystalline aggregates, statistics concept is introduced. Moreover, some key issues in the numerical simulation are discussed. They are mesh refinement analysis, the effect of the number of grains aggregating a polycrystal, and the effect of grain shape. As a benchmarking step, the effective elastic constants of the polycrystalline aggregates in bulk configuration are numerically estimated by the proposed finite element scheme with statistic consideration. Comparison is made between the numerical results and the existing analytical solutions.

In our finite element simulation, the following assumptions are considered:

- 1) The crystallographic orientation for each grain is randomly distributed within the polycrystalline aggregates. The preferred texture can be considered without any difficulties.
- 2) The interfaces between the grains are perfect so that the deformation and stress on the interface are continuous.

3) The theory of linear elasticity is applied. No special mechanisms, such as dislocation, micro-voids, micro-cracks and residual stress, are considered.

4.1 Finite element scheme with statistic consideration

4.1.1 Considerations in modeling polycrystalline aggregates

At the first glance, the primary point in studying the mechanical properties of polycrystalline aggregates is to set up a finite element model capturing the major microstructural features. The microstructure of a polycrystalline aggregate is mainly characterized by the microstructural morphology, i.e., the grain shape, size and spatial distribution, and the distribution of crystallographic orientation.

In our simulation, the grain size is selected as a characteristic dimension. All the other dimensions, such as the thickness of thin film and the length of thin wire, are normalized by the grain size. No absolute size is introduced. It has been noticed by both numerical study (Mill and Shenoy, 2000; Dingreville *et al.*, 2005; Zhang and Sun, 2006) and experimental measurement (Haque and Saif, 2004; Tan *et al.*, 2007) that the elastic behavior may be size-dependent when the surface energy overtakes the volume energy in the deformation. This is the so-called “size effect”. Such size effect, however, takes place only when the structure dimension is below tens of nano-meters. The present work does not include this “nano” size effect.

As far as the grain shape is concerned, it is possible to reconstruct the microstructural morphology of a real polycrystal with the advanced computational technique and experimental instrument with high precision and resolution. However, it is a tedious task, especially for the three-dimensional bulk configuration. Since the focus of our

work is to investigate the dependency of the effective modulus of the polycrystalline aggregates on macro geometric configuration, we use uniform grain shape to characterize the micro-morphology of the polycrystalline aggregates. To shed some light on the influence of the grain shape, the grain will be modeled with different shape in Section 4.3. It will be shown that the effective modulus is insensitive to the grain shape, no matter what degree of anisotropy the crystallite has.

For the crystallographic orientation, in general, different grains have different crystallographic orientations. This leads to a strong mechanical interaction and a large influence on the effective mechanical behavior of the polycrystalline aggregates. In the following simulation, we use random distribution of crystallographic orientation, while preferred orientation can be easily implemented along our scheme. To describe the crystallographic orientation of a grain in terms of the global coordinate system, a set of Euler angles (α, β, γ) is employed.

4.1.2 Finite element scheme

As the first step of our modeling, a simple scheme is proposed to automatically generate the microstructural model of the polycrystalline aggregates. In our model, all the grains have identical size and shape. The focus is the random distribution of crystallographic orientation. The effect of variation on the grain shape and size will be discussed later. This finite element model is, hereafter, called as uniform model. The framework of the proposed finite element scheme is presented as follows.

A prism specimen with macroscopic dimension $L_x \times L_y \times L_z$ is used to model a polycrystalline aggregate. The edges are parallel to the global coordinate axes, shown in Fig. 4.1(a). We first tessellate the specimen into N cubic domains with dimension d

$\times d \times d$, where d is the grain size. Each domain is treated as one grain. Obviously, there are N ($N = (L_x/d) \times (L_y/d) \times (L_z/d)$) grains in the specimen. Because different grain exhibits different anisotropic properties defined by a set of Euler angles (α, β, γ) , N -sets of Euler angles are randomly selected from Euler angle space with uniform distribution. Then, each set of Euler angles is randomly assigned to one grain. The finite element model of a polycrystalline specimen containing N grains has thus been established.

Figure 4.1(a) shows one realization (i.e., the generated finite element model of the specimen in the grain structure) of the specimen containing 512 grains. Different grain is represented by different grey level. It is seen that all the grains have identical cubic shape but different crystallographic orientation. By changing the crystallographic orientation of each grain, different realization can be constructed. Apparently, different realization exhibits different mechanical behavior and response under the identical loading condition.

Figure 4.1 Schematic illustration of a specimen containing 512 cubic grains. (a) Before mesh; (b) after mesh. The grain represented by different grey level is meshed into 27 hexahedral elements.

Since the crystallographic orientation varies from grain to grain, there is material property mismatch among the grains. High numerical error would be induced if there is only one element per grain. Hence, a process of mesh refinement, which subdivides each grain into multiple elements, is necessary. Detailed discussion will be presented in Subsection 4.2.2.

4.1.3 Statistic consideration

Due to the random distribution of crystallographic orientation, there is infinite number of specimens containing N grains. All these specimens form a “**population**” P_N . Different specimens exhibit different macroscopically mechanical properties such as the effective Young’s modulus. Because it is not possible to obtain the values of all the specimens in the population P_N , we need statistic concept to infer the population’s properties, on the basis of the information obtained from a representative sample randomly selected from the population.

For a given population P_N , the mean value and standard deviation of the studied property are denoted by μ and σ , respectively. From a sample consisting of n specimens, we can obtain the sample mean value $\bar{x}$ and the sample standard deviation s , i.e.,

$$\bar{x} = \frac{1}{n} \sum_{i=1}^n x_i, \quad (4.1)$$

$$s = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (x_i - \bar{x})^2}, \quad (4.2)$$

where x_i is the studied property (e.g., the effective modulus) of the specimen i ($i = 1, 2, \dots, n$). Furthermore, the coefficient of variation (COV), which describes the

fluctuation of the studied property relative to the corresponding mean value, can be obtained by

$$COV = \frac{s}{\bar{x}} \times 100\% . \quad (4.3)$$

According to the Central Limit Theorem (Mendenhall *et al.*, 2006), the mean value of the population μ can be inferred with a 99% confidence interval as

$$\bar{x} - 2.58 \frac{s}{\sqrt{n}} \leq \mu \leq \bar{x} + 2.58 \frac{s}{\sqrt{n}} , \quad (4.4)$$

when the sample size n is large. Note that the population standard deviation σ has been approximated by the sample standard deviation s . Apparently, the interval depends on the sample standard deviation s and the sample size n . It will be shown that the magnitude of the sample standard deviation s varies with the specimen size (or the number of the grains aggregating the specimen). Selecting the specimen size is a process of compromise between the computational cost and accuracy.

4.2 Estimation of the effective elastic constants

As an illustration, four polycrystals with cubic symmetry are examined in our numerical simulation. They are aluminum, copper, β -brass and lithium. Table 4.1 lists their elastic constants and anisotropic ratio (defined by Eq. (3.8)) of single crystal. It is seen that aluminum is almost isotropic, copper is medium anisotropic, β -brass is highly anisotropic and lithium is extremely anisotropic.

For the purpose of comparison and reference, the calculated effective Young's modulus and Poisson's ratio based on the analytical models described in Chapter 3 are

summarized in Table 4.2. It is observed that the spread between the lower bound and upper bound of the effective Young's modulus of a polycrystal positively correlates with its single crystal anisotropic ratio. Comparing the Hill's average value with the self-consistent solution, less than 3% discrepancy is observed for these four materials. In the rest chapters, our numerical results are validated against the Hill's average.

Table 4.1 Elastic constants of single crystal with cubic symmetry.

Material	C_{11} (GPa)	C_{12} (GPa)	C_{44} (GPa)	Anisotropic ratio (AR)
Aluminum ^a	107.30	60.90	28.30	1.22
Copper ^a	168.40	121.40	75.40	3.21
β -brass ^b	130.00	102.00	74.00	5.30
Lithium ^a	13.50	11.44	8.78	8.52

^a Freund and Suresh (2003).
^b Grimvall (1999).

Table 4.2 Analytical solutions for the effective elastic constants of polycrystalline aggregates.

Material	Effective Young's modulus (GPa)				Effective Poisson's ratio	
	Reuss value	Voigt value	Hill's average	Self-consistent solution	Hill's average	Self-consistent solution
Aluminum	70.08	70.68	70.38	70.41	0.346	0.346
Copper	109.44	144.69	127.36	129.36	0.345	0.343
β -brass	75.62	130.47	103.88	106.42	0.344	0.341
Lithium	6.20	14.74	10.65	10.93	0.354	0.350

4.2.1 Boundary condition and calculation of the elastic constants

In predicting the effective elastic constants of heterogeneous materials from their micro-constituents properties and microstructural aspects by using numerical simulation, there are various types of boundary conditions. They are homogeneous traction boundary condition, homogeneous displacement boundary condition, periodic boundary condition and mixed boundary condition. If the specimen contains sufficient number of micro-constituents, for example, grains in the polycrystalline aggregates or inclusions in the composites, the predicted effective properties are independent of the type of boundary conditions (Hill, 1963; Hazanov and Huet, 1994; Jiang *et al.*, 2001; Ostoja-Starzewski, 2006). Such specimen is called as “Representative Volume Element (RVE)”. In the literature, the first two types of boundary conditions (i.e., homogeneous traction boundary condition and homogeneous displacement boundary condition) have widely been used in the analytical research. It has been verified that the effective elastic constants obtained under the homogeneous traction boundary condition and the homogeneous displacement boundary condition are corresponding to the lower and upper bounds, respectively (Hill, 1963; Hashin, 1983; Hazanov and Huet, 1994; to mention a few).

To estimate the effective elastic constants, we prescribe the mixed boundary condition on the specimen with reference to Fig. 4.1(a), i.e.

$$\begin{aligned} u_x &= 0 & \text{at } x = 0, \\ u_x &= \Delta & \text{at } x = L_x, \\ u_y &= 0 & \text{along } x = 0, y = 0, \\ u_z &= 0 & \text{along } x = 0, z = 0, \end{aligned} \tag{4.5}$$

where Δ is the prescribed displacement. The symbols, u_x , u_y and u_z , denote the displacement along the direction x , y and z in the global coordinate system, respectively. In addition to the aforementioned boundary conditions, the non-specified boundary conditions are “traction-free”.

After numerical analysis, the data of stress, strain, displacement and strain energy can be obtained. According to the principle of energy balance, the effective Young’s modulus can be determined by

$$E = \frac{2L_x U}{\Delta^2 A}, \quad (4.6)$$

where L_x is the length of edge along the x direction, illustrated in Fig. 4.1(a); A is the cross-sectional area of the plane perpendicular to axis x ; U is the total strain energy collected from all the elements in the specimen.

For the effective Poisson’s ratio, we have

$$\begin{aligned} \nu_{xy} &= -\frac{\varepsilon_y}{\varepsilon_x} = -\frac{[\bar{u}_y(y=L_y) - \bar{u}_y(y=0)] / L_y}{\Delta / L_x}, \\ \nu_{xz} &= -\frac{\varepsilon_z}{\varepsilon_x} = -\frac{[\bar{u}_z(z=L_z) - \bar{u}_z(z=0)] / L_z}{\Delta / L_x}, \end{aligned} \quad (4.7)$$

where $\bar{u}_y(y=L_y)$ and $\bar{u}_y(y=0)$ are the averaged nodal displacement in the y direction over the specified surfaces $y=L_y$ and $y=0$ of the specimen, respectively; $\bar{u}_z(z=L_z)$ and $\bar{u}_z(z=0)$ are the averaged nodal displacement in the z direction over the specified surfaces $z=L_z$ and $z=0$ of the specimen, respectively.

4.2.2 Mesh refinement analysis

In order to reduce the computational error and guarantee the convergence of the numerical results, it is necessary to perform mesh refinement analysis. Hereafter, twenty-node hexahedral element, Solid186 provided by ANSYS, is used. This element is quadratic element and has 14 Gauss integration points. For a specific specimen, after the distribution of crystallographic orientation, each grain is meshed into 1, $2^3(=8)$, $3^3(=27)$, $4^3(=64)$ and $5^3(=125)$ elements, respectively. We refer them as the zeroth (R0), first (R1), second (R2), third (R3) and fourth (R4) level of mesh refinement, respectively. In Fig. 4.1(b), each grain in the specimen is meshed into 27 hexahedral elements (at the R2 mesh level).

To examine the influence of mesh refinement on the effective Young's modulus, a polycrystalline copper specimen containing 512 grains is considered. One realization is illustrated in Fig. 4.1 (a). Here, thirty different realizations (i.e., $n = 30$) are sampled. Since each realization is somewhat different in the distribution of crystallographic orientation, each realization corresponds to one pair of Young's modulus and Poisson's ratio after solving the boundary value problem described in Eq. (4.5). Thus, n pairs of Young's modulus and Poisson's ratio are obtained. From Eqs. (4.1) - (4.3), it is readily to obtain the sample statistic data (i.e., mean value, standard deviation and coefficient of variation) of the predicted property.

Table 4.3 summarizes the numerical results for the effective Young's modulus and Poisson's ratio obtained at various levels of mesh refinement. Figure 4.2 schematically presents the predicted effective Young's modulus versus the level of mesh refinement, together with the Reuss lower bound, Voigt upper bound and Hill's average. The error bar represents the discrepancy between the minimum and

maximum values of the numerical results. It is positively related to the sample standard deviation.

Table 4.3 Summary of finite element simulation of polycrystalline copper aggregates at various levels of mesh refinement. The specimen consists of 512 cubic grains. Twenty-node hexahedral element is used.

Mesh level (elements per grain)	Number of nodes	Number of elements	Effective Young's modulus (GPa)		Effective Poisson's ratio			
					ν_{xy}		ν_{xz}	
			Mean value	Standard deviation	Mean value	Standard deviation	Mean value	Standard deviation
R0 (1)	2673	512	129.04	1.61	0.344	0.006	0.346	0.004
R1(8)	18,785	4,096	127.40	1.60	0.345	0.006	0.346	0.004
R2(27)	60,625	13,824	127.08	1.60	0.345	0.006	0.347	0.004
R3(64)	140,481	32,768	126.96	1.60	0.345	0.006	0.347	0.004
R4(125)	270,641	64,000	126.90	1.60	0.345	0.006	0.347	0.005

It is noted that the predicted mean value decreases with increasing the mesh level or the number of elements per grain. This is in line with the finite element theory for the displacement-based formulation. Moreover, the predicted mean value converges with increasing the mesh level. Comparing the predicted mean value at the second mesh level (R2) with that obtained at the fourth mesh level (R4), there is less than 0.6% difference when the number of elements in one grain increases from $3^3(=27)$ to $5^3(=125)$. Based on these data, a mesh of 27 elements per grain is adopted for the following simulations. On the other hand, it is observed that the standard deviation is insensitive to the level of mesh refinement.

Figure 4.2 Scatter of the effective Young's modulus of polycrystalline copper aggregates containing 512 grains from the zeroth (R0) to fourth (R4) level of mesh refinement. The number of elements per grain is denoted by m for each level of mesh refinement. The sample size is $n = 30$. The error bar presents the discrepancy between the minimum and maximum values of the evaluated effective modulus.

4.2.3 Effect of the number of grains

In order to estimate the effective properties of polycrystalline aggregates by numerical simulation, the specimen should be a RVE which is a statistical representative of the studied material. Such specimen consists of certain number of grains (equivalently appropriate specimen size) which is a reasonable trade-off between the computational cost and accuracy of the estimation of the effective properties.

Investigation on the RVE size has been an active research topic in recent decade, due to the wide application of MEMS devices (Ren and Zheng , 2002, 2004; Nygård, 2003; Cho and Chasiotis, 2007; Houdaigui *et al.*, 2007; Ranganathan and Ostoj-Starzewski, 2008; Böhlke *et al.*, 2010). It is known that the size of RVE is material-property dependent. For example, the bulk modulus of a cubic material is a constant

value determined from the elastic constants of single crystal (Hill, 1952). This means that the RVE can be small. However, it is not the case for the shear modulus. Furthermore, the mechanical behavior and properties of a RVE must be independent of the type of boundary conditions (Hill, 1963).

Hereby, we estimate the effective Young's modulus in terms of specimen containing different number of grains, under the boundary conditions given in Eq. (4.5). Simulations are carried out for polycrystalline aluminum, copper, β -brass and lithium. The numerical results indicate the number of grains needed in a RVE for different polycrystalline aggregates.

For the sake of simplification, the specimen is modeled as a cubic volume with edge length equal to L . A dimensionless parameter δ , which is defined as the ratio of the edge length L relative to the grain size d , is introduced. Clearly, the number of grains aggregating the specimen is $N = (L/d)^3 = \delta^3$. The value of δ is in the range from 2 to 13. In other words, the grain number takes the value from $2^3 (=8)$ to $13^3 (=2197)$.

A sample of n different realizations is simulated for each specimen with given size δ (or equivalently containing $N = \delta^3$ grains). Note that the value of n is dependent on the specimen size δ and determined by the Central Limit Theorem. Let us recall Eq. (4.4). To ensure the estimation of the mean value μ of the population P_N to be within a specified error ε_{err} , namely, $|(\mu - \bar{x}) / \bar{x}| \leq \varepsilon_{err}$, we may obtain the sample size n from Eq. (4.4)

$$n \geq (2.58 \frac{s}{\bar{x} \varepsilon_{err}})^2. \quad (4.8)$$

This equation can serve as an indicator to select the sample size n so that the mean value μ of the population P_N can be inferred from the sample mean value $\bar{x}$ within the specific error.

Tables 4.4 - 4.7 summarize the numerical simulations for polycrystalline aluminum, copper, β -brass and lithium, respectively. Also included is the sample size n or the number of realizations associated with each studied specimen, which is calculated from Eq. (4.8) by setting $\varepsilon_{\text{err}} = 1\%$. For a given material, it is noted that the coefficient of variation $s / \bar{x}$ is inversely related to the number of the grains or the specimen size. Hence, for a specimen containing few grains, a large number of simulations should be conducted to satisfy the given error ε_{err} . Take polycrystalline copper as an example. With reference to Table 4.5, the value $s / \bar{x}$ reduces from 9.60% to 0.44% when the specimen size increases from $2 \times 2 \times 2$ to $13 \times 13 \times 13$, and 600 and 30 simulations are, respectively, needed for the specimen $2 \times 2 \times 2$ and $13 \times 13 \times 13$. Furthermore, for a given specimen size, the value $s / \bar{x}$ is positively correlated with the anisotropic ratio of the single crystal. For instance, when the specimen size is fixed at $5 \times 5 \times 5$, the sample size is 30, 50, 70 and 100 for polycrystalline aluminum, copper, β -brass and lithium, respectively. This implies that more realizations should be sampled to infer the population's mean value within the prescribed tolerance.

The determined mean value and the extreme values (the maximum and minimum values obtained in the simulations) of the predicted effective Young's modulus in terms of the number of grains $N (= \delta^3)$ are presented in Figs. 4.3(a) - (d) for polycrystalline aluminum, copper, β -brass and lithium. Moreover, the analytical results for the effective Young's modulus, i.e. the Reuss lower bound, Voigt upper bound and Hill's average, are included in Figs. 4.3 (a)-(d) for comparison.

Table 4.4 Statistic results for the effective Young's modulus of polycrystalline aluminum in bulk configuration (AR = 1.22). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
$2 \times 2 \times 2$	8	30	70.15	1.21	1.72
$3 \times 3 \times 3$	27	30	70.30	0.68	0.97
$4 \times 4 \times 4$	64	30	70.34	0.39	0.56
$5 \times 5 \times 5$	125	30	70.39	0.25	0.36
$6 \times 6 \times 6$	196	30	70.41	0.25	0.36
$7 \times 7 \times 7$	343	30	70.40	0.16	0.23
$8 \times 8 \times 8$	512	30	70.38	0.16	0.23
$9 \times 9 \times 9$	729	30	70.39	0.15	0.21
$10 \times 10 \times 10$	1,000	30	70.38	0.08	0.11
$11 \times 11 \times 11$	1,331	30	70.40	0.08	0.12
$12 \times 12 \times 12$	1,728	30	70.39	0.07	0.10
$13 \times 13 \times 13$	2,197	30	70.38	0.07	0.10

Table 4.5 Statistic results for the effective Young's modulus of polycrystalline copper in bulk configuration (AR = 3.21). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
$2 \times 2 \times 2$	8	600	121.12	11.63	9.60
$3 \times 3 \times 3$	27	200	123.36	6.34	5.14
$4 \times 4 \times 4$	64	80	124.81	4.26	3.41
$5 \times 5 \times 5$	125	50	125.78	2.97	2.36
$6 \times 6 \times 6$	196	30	126.71	2.27	1.79
$7 \times 7 \times 7$	343	30	127.20	1.69	1.33
$8 \times 8 \times 8$	512	30	127.13	1.34	1.05
$9 \times 9 \times 9$	729	30	127.74	1.15	0.90
$10 \times 10 \times 10$	1,000	30	128.00	0.83	0.65
$11 \times 11 \times 11$	1,331	30	127.85	0.63	0.49
$12 \times 12 \times 12$	1,728	30	127.75	0.6	0.47
$13 \times 13 \times 13$	2,197	30	127.89	0.56	0.44

Table 4.6 Statistic results for the effective Young's modulus of polycrystalline β -brass in bulk configuration (AR = 5.30). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
$2 \times 2 \times 2$	8	1250	93.39	12.61	13.51
$3 \times 3 \times 3$	27	300	97.03	6.32	6.52
$4 \times 4 \times 4$	64	180	99.62	4.85	4.87
$5 \times 5 \times 5$	125	70	101.37	3.30	3.25
$6 \times 6 \times 6$	196	50	101.01	2.55	2.52
$7 \times 7 \times 7$	343	30	102.27	2.14	2.09
$8 \times 8 \times 8$	512	30	102.85	1.98	1.92
$9 \times 9 \times 9$	729	30	103.11	1.23	1.19
$10 \times 10 \times 10$	1,000	30	103.93	1.16	1.12
$11 \times 11 \times 11$	1,331	30	104.20	0.89	0.86
$12 \times 12 \times 12$	1,728	30	104.28	0.84	0.81
$13 \times 13 \times 13$	2,197	30	104.35	0.70	0.67

Table 4.7 Statistic results for the effective Young's modulus of polycrystalline lithium in bulk configuration (AR = 8.52). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
$2 \times 2 \times 2$	8	2100	8.85	1.57	17.73
$3 \times 3 \times 3$	27	600	9.37	0.87	9.30
$4 \times 4 \times 4$	64	210	9.80	0.55	5.61
$5 \times 5 \times 5$	125	100	10.06	0.39	3.82
$6 \times 6 \times 6$	196	70	10.24	0.33	3.21
$7 \times 7 \times 7$	343	40	10.26	0.24	2.31
$8 \times 8 \times 8$	512	30	10.41	0.22	2.08
$9 \times 9 \times 9$	729	30	10.43	0.18	1.72
$10 \times 10 \times 10$	1,000	30	10.52	0.15	1.46
$11 \times 11 \times 11$	1,331	30	10.55	0.10	0.95
$12 \times 12 \times 12$	1,728	30	10.60	0.10	0.93
$13 \times 13 \times 13$	2,197	30	10.64	0.08	0.72

(a)

(b)

(c)

(d)

Figure 4.3 Scatter of the effective Young's modulus of polycrystalline aggregates in terms of the number of grains. (a) Aluminum; (b) copper; (c) β -brass; (d) lithium. The data are obtained at R2 mesh level, in which each grain is meshed into 27 twenty-node hexahedral elements.

From Figs. 4.3 (a)-(d), two basic trends can be clearly identified. Firstly, the scatter of the predicted Young's modulus decreases with increasing the number of the grains or the specimen size. Secondly, the mean value of the predicted Young's modulus depends on the number of the grains aggregating the specimen, and tends to be stable when the number of the grains increases.

For the first trend, it is in agreement with common sense in statistics. The scattered data is due to the heterogeneity of material, especially for the specimen with small number of the grains. It is known that the anisotropy of a cubic crystallite increases as its anisotropic ratio increases. As a result, the dispersion of the predicted Young's modulus of the specimen is bigger when the crystallite is more anisotropic. This can be verified by the coefficient of variation listed in the Tables 4.4 - 4.7. For a given specimen size, the higher the anisotropic ratio of single crystal is, the larger the coefficient of variation is.

For the second trend, it is in line with the homogenization theory. The results plotted in Figs. 4.3 (a) – (d) indicate that the mean value of the effective Young's modulus converges to the Hill's average in our cases as the number of the grains increases. It is known that the Hill's average is a good approximation to the experimental values for many polycrystalline aggregates (Watt *et al.*, 1976). It is again verified by our finite element simulation.

In our simulations, if we take the error $\varepsilon_{err} = |(\mu - \bar{x}) / \bar{x}| = 1\%$ and the sample size $n = 30$, we can approximately estimate that 8, 196, 343 and 512 grains are needed in the RVE for polycrystalline aluminum, copper, β -brass and lithium, respectively. The greater the anisotropic ratio is, the more the grains are needed in the RVE. Similar

results have been reported by other investigators. Houdaigui *et al.* (2007) claimed that the RVE size remains in the order of 40 to 400 grains for a given precision of 1% in the estimation of the effective elastic constants, and the sample size n varies from 10 to 100. According to their formula, about 220 grains are needed in the RVE for polycrystalline copper by taking $n = 20$. Ren and Zheng (2004) claimed that a minimum RVE size is 16 grains for most of materials.

Moreover, with a close inspection on Fig. 4.3, it is found that the specimen containing few grains appears anisotropic properties and its corresponding elastic modulus falls outside the range between the Reuss lower bound and Voigt upper bound. However, such anisotropic properties may be smeared out by an average procedure over a large sample. The sample mean value of the Young's modulus, thus, lies somewhere within the range of Reuss-Voigt bounds, no matter how many grains are enclosed in the specimen. This fact has been proved by Hill from theoretical point of view (Hill, 1952). Huet (1990) also concluded that the mean value of the predicted moduli should be in the range between the Reuss value and the Voigt value, no matter the specimen is larger or smaller than the RVE.

To make the numerical data complete, the statistic data associated with the estimated effective Poisson's ratio are summarized in Tables 4.8-4.11 for polycrystalline aluminum, copper, β -brass and lithium.

4.3 Effect of the grain shape

In the above, the randomness of crystallographic orientation is fully incorporated into the finite element model, while the grain shape and size remain identical for all the grains. To shed some light on the influence of the grain morphology (the description

of the variation of the grain size and grain shape), the effective Young's modulus of the polycrystalline aggregates are compared in three micro-morphologies.

Table 4.8 Statistic results for the effective Poisson's ratio of polycrystalline aluminum in bulk configuration (AR = 1.22). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Sample size (n)	ν_{xy}			ν_{xz}		
		Mean value	Standard deviation	Coefficient of variation (%)	Mean value	Standard deviation	Coefficient of variation (%)
$2 \times 2 \times 2$	30	0.3488	0.0074	2.13	0.3452	0.0078	2.27
$3 \times 3 \times 3$	30	0.3474	0.0052	1.49	0.3460	0.0052	1.51
$4 \times 4 \times 4$	30	0.3468	0.0034	0.98	0.3463	0.0030	0.86
$5 \times 5 \times 5$	30	0.3459	0.0024	0.69	0.3469	0.0024	0.68
$6 \times 6 \times 6$	30	0.3459	0.0021	0.60	0.3468	0.0020	0.57
$7 \times 7 \times 7$	30	0.3463	0.0010	0.29	0.3464	0.0011	0.32
$8 \times 8 \times 8$	30	0.3465	0.0011	0.32	0.3463	0.0009	0.27
$9 \times 9 \times 9$	30	0.3462	0.0011	0.31	0.3465	0.0009	0.25
$10 \times 10 \times 10$	30	0.3463	0.0009	0.27	0.3465	0.0009	0.26
$11 \times 11 \times 11$	30	0.3464	0.0007	0.20	0.3463	0.0006	0.18
$12 \times 12 \times 12$	30	0.3465	0.0005	0.15	0.3463	0.0005	0.15
$13 \times 13 \times 13$	30	0.3465	0.0005	0.14	0.3464	0.0005	0.15

Table 4.9 Statistic results for the effective Poisson's ratio of polycrystalline copper in bulk configuration (AR = 3.21). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Sample size (n)	ν_{xy}			ν_{xz}		
		Mean value	Standard deviation	Coefficient of variation (%)	Mean value	Standard deviation	Coefficient of variation (%)
$2 \times 2 \times 2$	600	0.3563	0.0574	16.12	0.3541	0.0589	16.63
$3 \times 3 \times 3$	200	0.3488	0.0350	10.05	0.3539	0.0357	10.08
$4 \times 4 \times 4$	80	0.3511	0.0196	5.57	0.3476	0.0188	5.42
$5 \times 5 \times 5$	50	0.3465	0.0134	3.87	0.3490	0.0143	4.11
$6 \times 6 \times 6$	30	0.3457	0.0109	3.17	0.3475	0.0107	3.09
$7 \times 7 \times 7$	30	0.3447	0.0090	2.61	0.3469	0.0090	2.58
$8 \times 8 \times 8$	30	0.3480	0.0073	2.09	0.3440	0.0084	2.46
$9 \times 9 \times 9$	30	0.3448	0.0048	1.38	0.3451	0.0047	1.29
$10 \times 10 \times 10$	30	0.3448	0.0043	1.24	0.3445	0.0047	1.35
$11 \times 11 \times 11$	30	0.3450	0.0035	1.00	0.3447	0.0032	0.93
$12 \times 12 \times 12$	30	0.3446	0.0033	0.97	0.3451	0.0032	0.93
$13 \times 13 \times 13$	30	0.3446	0.0025	0.74	0.3448	0.0026	0.76

Table 4.10 Statistic results for the effective Poisson's ratio of polycrystalline β -brass in bulk configuration (AR = 5.30). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Sample size (n)	ν_{xy}			ν_{xz}		
		Mean value	Standard deviation	Coefficient of variation (%)	Mean value	Standard deviation	Coefficient of variation (%)
$2 \times 2 \times 2$	1250	0.3653	0.0879	24.07	0.3631	0.0889	24.48
$3 \times 3 \times 3$	300	0.3589	0.0441	12.30	0.3559	0.0470	13.20
$4 \times 4 \times 4$	180	0.3519	0.0307	8.73	0.3524	0.0301	8.55
$5 \times 5 \times 5$	70	0.3511	0.0185	5.28	0.3478	0.0175	5.02
$6 \times 6 \times 6$	50	0.3500	0.0152	4.33	0.3496	0.0155	4.42
$7 \times 7 \times 7$	30	0.3512	0.0150	4.26	0.3445	0.0136	3.94
$8 \times 8 \times 8$	30	0.3449	0.0112	3.25	0.3484	0.0119	3.41
$9 \times 9 \times 9$	30	0.3436	0.0077	2.25	0.3488	0.0071	2.04
$10 \times 10 \times 10$	30	0.3451	0.0095	2.74	0.3447	0.0089	2.59
$11 \times 11 \times 11$	30	0.3443	0.0055	1.61	0.3448	0.0052	1.52
$12 \times 12 \times 12$	30	0.3437	0.0050	1.44	0.3449	0.0048	1.40
$13 \times 13 \times 13$	30	0.3435	0.0036	1.05	0.3447	0.0037	1.08

Table 4.11 Statistic results for the effective Poisson's ratio of polycrystalline lithium in bulk configuration (AR = 8.52). Each grain is meshed into 27 twenty-node hexahedral elements.

Specimen (L/d) $\times$ (L/d) $\times$ (L/d)	Sample size (n)	ν_{xy}			ν_{xz}		
		Mean value	Standard deviation	Coefficient of variation (%)	Mean value	Standard deviation	Coefficient of variation (%)
$2 \times 2 \times 2$	2100	0.3844	0.1172	30.50	0.3851	0.1173	30.45
$3 \times 3 \times 3$	600	0.3763	0.0611	16.23	0.3734	0.0604	16.18
$4 \times 4 \times 4$	210	0.3723	0.0343	9.20	0.3641	0.0340	9.33
$5 \times 5 \times 5$	100	0.3602	0.0257	7.15	0.3669	0.0259	7.05
$6 \times 6 \times 6$	70	0.3610	0.0178	4.94	0.3614	0.0174	4.74
$7 \times 7 \times 7$	40	0.3592	0.0173	4.83	0.3615	0.0163	4.53
$8 \times 8 \times 8$	30	0.3559	0.0081	2.27	0.3604	0.0071	1.96
$9 \times 9 \times 9$	30	0.3575	0.0084	2.34	0.3576	0.0085	2.39
$10 \times 10 \times 10$	30	0.3563	0.0081	2.28	0.3560	0.0098	2.75
$11 \times 11 \times 11$	30	0.3540	0.0058	1.64	0.3574	0.0062	1.74
$12 \times 12 \times 12$	30	0.3538	0.0062	1.74	0.3562	0.0068	1.92
$13 \times 13 \times 13$	30	0.3537	0.0060	1.70	0.3538	0.0061	1.73

In the previous section, we model the polycrystalline aggregates with uniform hexahedron-shaped grains (shown in Fig. 4.1). Hereby, we use another uniform grain shape, i.e. tetrahedron, to make comparison with the previous result. To set up such model, we first discretize the specimen into N tetrahedron-shaped domains (i.e., Solid187 provided by ANSYS). Each domain is treated as one grain. Each grain is then randomly assigned with a set of Euler angles (α, β, γ) to characterize its material properties. Figure 4.4 (a) shows one realization of a specimen containing 536 tetrahedron-shaped grains. The grains are represented by different grey levels. The mesh refinement analysis is carried out for polycrystalline copper aggregates containing 536 grains. Table 4.12 lists the corresponding numerical results.

Figure 4.4 Schematic illustration of a specimen modeling in different grain structures. (a) Specimen containing 536 tetrahedron-shaped grains; (b) specimen containing 512 grains with various shapes and sizes.

In the third model of the grain morphology, we incorporate the process of regular crystallization into the model. Firstly, the specimen is meshed into a larger number of elements M . Secondly, N points, which position is defined by (x, y, z) with respect to the global coordinate system, are randomly generated within the specimen. Each point is corresponding to the nucleus of one grain. Each nucleus is allocated a set of Euler

angles defining its material properties. Thirdly, the distances between the center of each element and each nucleus are calculated. According to the nearest-neighbor rule, for the nucleus i , the elements which are nearer to it than to any other nuclei are identified. All these elements form the grain with the nucleus i and thus have identical material properties with this nucleus. In this model, the grain shape and size varies from grain to grain. A realization for a specimen containing 512 grains is presented in Fig. 4.4 (b). In this case, the cubic specimen is discretized into $30 \times 30 \times 30$ twenty-node hexahedral elements. Hence, one grain is made up of about 50 elements on average.

Table 4.12 Summary of finite element simulations for polycrystalline copper aggregates in terms of the level of mesh refinement. The specimen is composed of 536 tetrahedron-shaped grains. Ten-node tetrahedral element is used. The sample size is $n = 30$.

Mesh level (elements per grain)	Number of nodes	Number of elements	Effective Young's modulus (GPa)		Effective Poisson's ratio			
					ν_{xy}		ν_{xz}	
			Mean value	Standard deviation	Mean value	Standard deviation	Mean value	Standard deviation
R0(1)	985	536	134.54	1.40	0.338	0.006	0.336	0.007
R1(8)	6,736	4,287	130.11	1.40	0.343	0.006	0.341	0.007
R2(27)	21,537	14,464	128.92	1.28	0.344	0.007	0.343	0.007
R3(64)	49,599	34,226	128.16	1.51	0.345	0.007	0.343	0.007
R4(216)	163,135	115,734	127.68	1.48	0.346	0.007	0.344	0.007
R5(512)	381,345	274,272	127.78	1.38	0.346	0.006	0.344	0.007
R6(729)	540,478	390,525	127.73	1.37	0.346	0.006	0.344	0.007

To examine the effect of grain shape, a specimen is modeled with 512 cube-shaped, 536 tetrahedron-shaped, and 512 randomly shaped grains and the corresponding effective Young's modulus is evaluated. All simulations are performed for polycrystalline aluminum, copper, β -brass and lithium aggregates. The numerical results are summarized in Table 4.13. It is noticed that the mean values of the predicted effective Young's modulus obtained from different micro-morphology are very close, even for lithium crystal with high anisotropic ratio ($AR = 8.52$). Therefore, we may conclude that the effective modulus of a polycrystalline aggregate is slightly sensitive to the grain shape. This is consistent with the observations of Kumar (1992), Elvin (1996), Mika and Dawson (1998), and Lebensohn *et al.* (2005). With this knowledge in mind, we adopt cube-shaped grain to model the polycrystalline aggregates in the following analysis.

Table 4.13 The mean value of the effective Young's modulus for polycrystalline specimen modeled with different micro-morphology. Each data is based on thirty simulations.

Grain shape	Number of grains (elements per grain)	Mean value of the effective Young's modulus (GPa)			
		Aluminum	Copper	β -brass	Lithium
Cube	512 (27)	70.35	127.08	103.63	10.37
Tetrahedron	536 (216)	70.41	127.68	103.00	10.50
Randomness	512 (average 50)	70.35	127.43	103.49	10.49

4.4 Summary

In this chapter, a finite element scheme in combination with statistic consideration is developed to evaluate the effective elastic constants of polycrystalline aggregates. The

polycrystalline aggregates are modeled with cubic grains and random distribution of crystallographic orientation. Four materials with cubic symmetry, i.e., aluminum, copper, β -brass and lithium, are simulated to predict the effective elastic constants in bulk configuration by the proposed finite element scheme with statistic consideration. The accuracy of the numerical results is validated against the analytical solutions.

The study on the effect of the number of grains indicates that the mean values of the effective elastic constants (Young's modulus and Poisson's ratio) become stable as the number of the grains or the specimen size increases, while the corresponding standard deviations inversely reduce. Meanwhile, the number of the grains in a RVE is positively correlated with the anisotropic ratio of the crystallite. The numerical results show that 8, 196, 343 and 512 grains are needed in the RVE for aluminum, copper, β -brass and lithium, respectively.

The influence of the micro-morphology (the grain shape, size and distribution) is studied by using hexahedron-shaped grain, tetrahedron-shaped grain and randomly shaped grain. It is seen that the effective elastic constants of the polycrystalline aggregates poorly depend on the microstructural morphology, even for the extremely anisotropic lithium crystallite ($AR = 8.52$). Hence, it is reasonable to use the simple uniform model to simulate the real microstructure of the polycrystalline aggregates.

Chapter 5 Copper Thin Films

In this chapter, polycrystalline copper thin films in different configuration are simulated for estimating their effective Young's modulus by the proposed finite element scheme. The configurations are the free-standing film, the substrate-attached film and the sandwiched film. For the free-standing film, we look into the effect of the film thickness and the grain structure. For the substrate-attached and the sandwiched films, we focus on the influence of the surrounding material. The influence of the microstructural morphology on the effective modulus is examined by comparing Voronoi model with uniform model.

5.1 The definition of thin film

In the present study, a film is referred to as “thin film” if it is made of a single layer of grains. This columnar structure is commonly found in the polycrystalline thin film (Nix, 1989; Freund and Suresh, 2003). Otherwise, the film is called as multiple-layered film. In the Fig. 5.1, the geometry of a thin film is illustrated in micro- and macro-structural dimension, in which the grain shape is assumed to be cubic. The parameter L is the macroscopic dimension of the thin film in x - y plane, t is the film thickness, and d is the grain size of the film in the x - y plane. The grain size d is taken as the characteristic dimension of the film. And all the other geometrical parameters, i.e., t , L and h (the thickness of the substrate used later), are normalized by the grain size as t/d , L/d and h/d respectively.

Figure 5.1 A free-standing thin film with columnar grain structure. The specimen is composed of $(L/d) \times (L/d)$ grains with equal size $d \times d \times t$. Grains are represented by different grey levels.

5.2 Simulation of the free-standing films

To evaluate the effective Young's modulus of the free-standing thin film, with reference to Fig. 5.1, the following uni-axial loading is prescribed.

$$\begin{aligned}
 u_x &= 0 && \text{at } x = 0, \\
 u_x &= \Delta && \text{at } x = L, \\
 u_y &= 0 && \text{along } x = 0, y = 0, \\
 u_z &= 0 && \text{along } x = 0, z = 0,
 \end{aligned} \tag{5.1}$$

where Δ is the prescribed displacement. The symbols, u_x , u_y and u_z , denote the displacement along the direction x , y and z in the global coordinate system, respectively. The non-specified boundary conditions are “traction-free”.

Based on the principle of energy balance, the effective Young's modulus can be calculated from

$$E = \frac{2LU}{\Delta^2 A}, \quad (5.2)$$

where A is the cross-sectional area of the specimen at $x = L$ and U is the total strain energy collected from all the grains in the thin film.

5.2.1 Sensitivity of the effective Young's modulus to the film thickness

The specimen with different ratio t/d between the film thickness t and the grain size d is simulated to investigate the variation of the effective modulus. The ratio t/d takes the value from 0.1 to 1. As pointed out in Chapter 4, for polycrystalline copper, a specimen can be treated as a RVE for 196 or more grains formed an aggregate. Hereby, all the specimens assemble 400 grains with identical size $d \times d \times t$. Each grain is randomly assigned a crystallographic orientation with uniform distribution. Hence, no preferred texture is included and the film material is macroscopically homogeneous and isotropic. The twenty-node hexahedral element with 14 Gauss integration points (Solid186) is used to do mesh. After performing mesh refinement, the specimen is discretized into 30,000 elements and 152,207 nodes. Each grain contains 75 elements and has more than 1,000 Gauss points.

Figure 5.2 plots the mean value and dispersion of the effective Young's modulus versus the ratio t/d . For a given ratio t/d , thirty numerical simulations are performed. It is noted that the mean value of the effective Young's modulus decreases with reducing the film thickness. Such feature may be attributed to the weakening constraint among the grains when the film is thinned.

Figure 5.2 Scatter of the effective Young's modulus of polycrystalline copper free-standing thin film versus the ratio t/d between the film thickness t and the grain size d . Each specimen consists of 400 grains. Each grain is discretized into 75 twenty-node hexahedral elements. The sample size is $n = 30$.

5.2.2 Plane stress approximation for the free-standing thin film

When the film thickness is much less than the grain size, the free-standing thin film may be simplified from three-dimensional case to two-dimensional case in the state of plane stress. Such plane stress approximation has been used to study the effective elastic properties of polycrystalline thin film by some researchers. However, the film was always assumed to have preferred crystallographic orientation. Examples can be found in the work by den Toonder *et al.* (1999) and Choi (2004).

To verify this point, a square specimen containing 400 grains with size $d \times d$ is numerically analyzed via the plane stress approximation. The crystallographic orientation of each grain is randomly assigned. Each grain is discretized into 81 elements with eight-node quadrilateral element (Plane 183 provided by ANSYS) after

mesh refinement. The square specimen is thus meshed into 32,400 elements and 97,921 nodes.

Table 5.1 presents the statistic data for the effective Young's modulus via the simulation of the polycrystalline copper free-standing thin film with $t/d = 0.1$ and the plane stress approximation. Negligible discrepancy is observed. It is, therefore, reasonable to believe that the effective modulus obtained from the plane stress approximation is the limit value of the free-standing thin film as the ratio t/d approaches zero.

Table 5.1 Statistic data for the effective Young's modulus via three-dimensional simulation of polycrystalline copper free-standing thin film with $t/d = 0.1$ and plane stress approximation. The sample size is $n = 30$.

Simulation model	Specimen $(L/d) \times (L/d) \times (t/d)$	Number of grains	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
Three-dimensional simulation	$20 \times 20 \times 0.1$	400	116.69	1.30	1.11
Plane stress approximation	20×20	400	116.46	1.71	1.47

The plane stress approximation is further performed for the specimen consisting of different number of the grains. Table 5.2 summarizes the numerical results of various copper specimens via the plane stress approximation. Also included is the sample size n or the number of realizations associated with each studied specimen, which is calculated from Eq. (4.8) by setting $\varepsilon_{\text{err}} = 1\%$. It is again indicated that 400 grains is sufficient for a copper specimen to be a RVE.

Table 5.2 Statistic data for the effective Young's modulus of polycrystalline copper via plane stress approximation. Each grain is meshed into 81 eight-node quadrilateral elements.

Specimen (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
3×3	9	600	115.01	10.73	9.33
4×4	16	300	115.94	7.66	6.60
5×5	25	250	116.07	6.30	5.42
7×7	49	100	115.97	4.22	3.63
10×10	100	50	115.89	3.01	2.60
15×15	225	30	116.29	2.28	1.96
20×20	400	30	116.46	1.71	1.47
30×30	900	30	116.53	1.06	0.91
40×40	1600	30	116.98	0.69	0.59
50×50	2500	30	117.09	0.55	0.47
60×60	3600	30	116.84	0.43	0.36
80×80	6400	30	116.82	0.20	0.17

Figure 5.3 presents the corresponding scatter and mean value of the effective modulus of polycrystalline copper via plane stress approximation in terms of the number of grains. When the number of grains increases, the mean value has negligible variation, while the corresponding standard deviation reduces dramatically. This feature is consistent with the observed phenomenon in Chapter 4. For a complete reference, the numerical results for polycrystalline aluminum, β -brass and lithium in the state of plane stress are summarized in Tables 5.3 - 5.5.

Figure 5.3 Scatter of the effective Young's modulus of polycrystalline copper versus the number of grains via plane stress approximation. Each grain is meshed into 81 eight-node quadrilateral elements.

5.2.3 Sensitivity of the effective modulus to the film grain structure

It is known that the grain structure of a polycrystalline film is affected by processing technique and condition (Thompson, 2000). Besides the aforementioned thin film with columnar grain structure, the film with multiple layers of grains in the thickness

direction is also existed in the application (i.e., Fig. 1.2(d)). It is expected such film would exhibit different modulus.

Table 5.3 Statistic data for the effective Young's modulus of polycrystalline aluminum via plane stress approximation. Each grain is meshed into 81 eight-node quadrilateral elements.

Specimen (L/d) $\times$ (L/d)	Number of grain (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
3×3	9	30	70.21	0.99	1.41
4×4	16	30	70.33	0.73	1.03
5×5	25	30	70.29	0.61	0.87
7×7	49	30	70.29	0.45	0.65
10×10	100	30	70.23	0.38	0.54
15×15	225	30	70.17	0.22	0.31
20×20	400	30	70.18	0.16	0.23
30×30	900	30	70.21	0.13	0.18
40×40	1600	30	70.18	0.07	0.10
50×50	2500	30	70.20	0.05	0.07
60×60	3600	30	70.19	0.04	0.06
80×80	6400	30	70.19	0.03	0.04

Table 5.4 Statistic data for the effective Young's modulus of polycrystalline β -brass via plane stress approximation. Each grain is meshed into 81 eight-node quadrilateral elements.

Specimen (L/d) $\times$ (L/d)	Number of grains (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
3×3	9	1100	83.69	10.73	12.82
4×4	16	600	84.30	7.71	9.14
5×5	25	400	84.24	6.24	7.41
7×7	49	200	83.95	4.38	5.22
10×10	100	100	84.56	3.24	3.83
15×15	225	60	84.59	2.27	2.69
20×20	400	40	85.12	1.96	2.30
30×30	900	30	85.23	1.05	1.23
40×40	1600	30	85.06	0.79	0.93
50×50	2500	30	85.14	0.66	0.78
60×60	3600	30	85.14	0.51	0.60
80×80	6400	30	85.22	0.22	0.26

Table 5.5 Statistic data for the effective Young's modulus of polycrystalline lithium via plane stress approximation. Each grain is meshed into 81 eight-node quadrilateral elements.

Specimen (L/d) $\times$ (L/d)	Number of grain (N)	Sample size (n)	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
3×3	9	1800	7.30	1.21	16.55
4×4	16	1000	7.34	0.90	12.30
5×5	25	700	7.34	0.72	9.87
7×7	49	400	7.34	0.51	6.99
10×10	100	200	7.39	0.38	5.13
15×15	225	100	7.44	0.26	3.54
20×20	400	60	7.48	0.21	2.75
30×30	900	30	7.47	0.13	1.78
40×40	1600	30	7.45	0.08	1.09
50×50	2500	30	7.47	0.07	0.91
60×60	3600	30	7.49	0.05	0.68
80×80	6400	30	7.49	0.03	0.43

Free-standing film containing multiple grains in the film thickness

The films with one, two, three and five layers of the grains in the thickness direction (z direction) are simulated. Each layer consists of 400 grains with identical size $d \times d \times d$ and randomly assigned crystallographic orientation. Figure 5.4 depicts the microstructure of the films consisting of one, two and three layers of the grains. Grains are represented by different grey levels. In estimating the effective modulus, all the grains are meshed into 27 twenty-node hexahedral elements. Hence, the specimen with two layers of the grains plotted in Fig. 5.4(b), for example, is discretized into 21,600 elements and 99,613 nodes.

Figure 5.4 The films consisting of various layers of grains along the thickness direction (z -direction). Each layer is composed of 400 grains with equal size $d \times d \times d$. Grains are represented by different grey levels. (a) One layer; (b) two layers; (c) three layers.

Figure 5.5 shows the mean values marked by ‘♦’ of the effective modulus associated with different number of the grain layers in the z direction, together with Hill’s average (127.36GPa), Reuss lower bound (109.44GPa) and Voigt upper bound (144.69GPa). Each mean value is based on a sample of thirty simulations.

It is seen that the effective modulus of the film approaches to the value of its bulk counterpart (which can be approximated by Hill's average) as the number of the grain layers along the film thickness increases. When multiple layers of the grains are added in the direction of the film thickness, the interior grains increase. These grains behave in a similar manner as the interior grains in the bulk polycrystal. As expected, the effective modulus approaches to the value of bulk polycrystalline aggregates from the below. In the case of the specimen built up with five layers of the grains, the predicted effective modulus is 127.04GPa.

Figure 5.5 The mean value of the effective Young's modulus of polycrystalline copper film versus the number of the grain layers along the thickness direction. The symbol '◆' is the mean value based on a sample of thirty simulations. Each layer is composed of 400 grains with equal size $d \times d \times d$ and randomly assigned crystallographic orientation.

Free-standing film containing different rows of grains

Consider a single layered thin film with limited number of rows of the grains in, say, y -direction. With the knowledge obtained in the previous section, we now

approximate this three-dimensional free-standing thin film as a two-dimensional film in the state of plane stress. Four films with 2, 5, 10 and 20 rows of the grains are constructed. All the films consist of 400 grains with identical size $d \times d$ and randomly assigned crystallographic orientation. Figure 5.6 (a) represents a realization of the specimen made of 20 rows of the grains; while Fig. 5.6 (b) is a realization of the specimen composed of 10 rows of the grains. After performing mesh refinement, each of the grains in the specimen is meshed into 81 eight-node quadrilateral elements (Plane 183). As an example, the specimen shown in Fig. 5.6 (a) is discretized into 32,400 elements and 97,921 nodes.

Figure 5.6 The single layered films with different rows of grains in y -direction. Each film consists of 400 grains with equal size $d \times d$. Grains are represented by different grey levels. (a) Twenty rows; (b) ten rows.

The mean values of the effective Young's modulus in terms of the number of the grain rows in the y direction are presented in Fig. 5.7, together with Reuss lower bound (109.44GPa) and Voigt upper bound (144.69GPa). The numerical results marked by symbol '♦' are the corresponding mean values based on thirty different simulations. It is observed that the effective modulus decreases and comes close to the Reuss lower bound as the rows of the grains reduce from 20 to 2. When the number of the grains in

the y direction is comparable with that in the x direction, the effective modulus approaches to 116.46GPa, the predicted modulus of the free-standing thin film.

The trend showing in Fig. 5.7 is due to the reducing constraints among the grains. As a result, each of the grains has more and more uniform stress under the uni-axially tensile loading. For the specimen with two rows of the grains, the constraint on the deformation of a grain mostly comes from the grains in the same row and thus the state of each grain is very similar to the assumption in the Reuss model. Therefore, the corresponding predicted effective modulus is very close to the Reuss lower bound.

Figure 5.7 The mean value of the effective Young's modulus of polycrystalline copper thin film via plane stress approximation versus the number of the grain rows in y -direction. The symbol '♦' is the mean value based on a sample of thirty simulations. All the specimens consist of 400 grains with equal size $d \times d$ and randomly assigned crystallographic orientation.

5.3 Simulation of the substrate-attached thin films

Figure 5.8 schematically illustrates the geometric configuration and corresponding geometric parameters (t , d , L and h) of a substrate-attached thin film with columnar

grain structure, in which t is the film thickness, d is the grain size in the x - y plane perpendicular to the film thickness, L is the edge length of the film along the x and y directions, h is the thickness of the substrate. The thickness of the substrate is much larger than the thickness of the attached film.

Figure 5.8 A substrate-attached thin film with columnar grain structure. The thin film is composed of $(L/d) \times (L/d)$ grains with equal size $d \times d \times t$. Grains are represented by different grey levels.

5.3.1 Sensitivity of the effective Young's modulus to the substrate

The influence of the substrate is studied from two aspects, namely, the substrate stiffness and the substrate thickness. Four types of substrate material are selected to examine the effect of the substrate stiffness. They are polyimide, silicon, silicon carbide and polycrystalline diamond. This problem was noticed in measuring the mechanical properties of the thin films attached to different substrate materials (Saha and Nix, 2002; Fang and Chang, 2003). The material constants associated with the substrate are listed in Table 5.6. Polyimide is the softest substrate in our study and widely used in the flexible electronic devices (Lu *et al.*, 2010), while polycrystalline diamond has the highest stiffness. Silicon substrate, which is the most common

substrate material in the microelectronics and MEMS, is a piece of single crystal Si wafer with (100) orientation. Silicon carbide (SiC) is widely used in the semiconductor electronics. For the study of the substrate thickness, the ratio h/d between the substrate thickness h and the grain size d varies from 0.1 to 5.

Table 5.6 Elastic constants of substrate material.

Material type	Young's modulus (GPa)	Poisson's ratio
Polyimide ^a	2.5	0.34
Silicon carbide ^b	445	0.22
Polycrystalline diamond ^b	974	0.10
Silicon ^b	$C_{11} = 165.7\text{GPa}$, $C_{12} = 63.9\text{GPa}$, $C_{44} = 79.6\text{GPa}$	

^a Wikström and Nygård (2002).

^b Freund and Suresh (2003).

For the substrate-attached film, we cannot directly apply Eq. (5.2) to evaluate the effective Young's modulus due to the coupled deformation between the film and the substrate. We apply homogenization concept to determine the film modulus through comparing the strain energy obtained from the system of a real film and substrate (Figs. 5.9 (a) and (c)) with that from the system of an equivalently homogenized film and substrate (Figs. 5.9 (b) and (d)). Since the attached film is macroscopically isotropic material, which effective elastic properties can be characterized by an independent pair of Young's modulus and Poisson's ratio (E , ν) or of bulk modulus and shear modulus (K , G), two independent loadings are needed, for example, one uni-axial loading and one bi-axial loading. The uni-axial loading condition (Figs. 5.9 (a) and (b)) is identical to Eq. (5.1); while the bi-axial loading (Figs. 5.9 (c) and (d)) is imposed as followings.

$$\begin{aligned}
u_x &= 0 & \text{at } x &= 0, \\
u_x &= \Delta & \text{at } x &= L, \\
u_y &= 0 & \text{at } y &= 0, \\
u_y &= \Delta & \text{at } y &= L, \\
u_z &= 0 & \text{at } x = y = z &= 0,
\end{aligned} \tag{5.3}$$

where Δ is the prescribed displacement. The symbols, u_x , u_y and u_z , denote the displacement along the direction x , y and z in the global coordinate system, respectively. The non-specified boundary conditions are “traction-free”.

In order to compare with the numerical results of the free-standing thin film, the substrate-attached copper thin film is modeled with ratio $t/d = 0.1$. It is a collection of 400 cubic grains with randomly assigned crystallographic orientation. The procedure to evaluate the effective Young’s modulus of the attached thin film is elaborately illustrated as follows.

Firstly, thirty different realizations for the real film/substrate system are simulated to determine the statistic data of the total strain energy stored in the real film under the uni-axial loading and bi-axial loading (shown in Figs. 5.9 (a) and (c)). Secondly, a pair of Young’s modulus and Poisson’s ratio (E , ν) is prescribed to the homogenized film. The homogenized film/substrate system is then simulated for evaluating the total strain energy of the homogenized film under the same uni-axial loading and bi-axial loading. Thirdly, comparison is made between these two systems. If these two films have identical energy under the uni-axial and bi-axial loading conditions, the assumed Young’s modulus and Poisson’s ratio are corresponding to the effective elastic constants of the real film. Otherwise, a new set of Young’s modulus and Poisson’s ratio is assigned to the homogenized film and the second step is repeated.

Figure 5.9 Homogenization of the attached copper film under the prescribed loading condition. In (a) and (c), the film formed by multiple grains with known crystal constants (C_{ij}) is subjected to uni-axial and bi-axial tensile loadings, respectively; in (b) and (d), the equivalently isotropic film with yet to be determined effective Young's modulus (E) and Poisson's ratio (ν) is subjected to uni-axial and bi-axial tensile loadings, respectively.

For illustration, a copper thin film attached on a diamond substrate with different thickness is considered. The thickness ratio, h/t , between the substrate and the film ranges from 1 to 100. Following the foregoing procedure, for a given value h/t , the energy discrepancy between the real film and the homogenized film is calculated as a function of the prescribed (E, ν).

Figures 5.10 (a)-(f) present the energy discrepancies under the uni-axial and bi-axial loadings versus the effective Young's modulus E for various values of Poisson's ratio ν . The dashed lines, denoted by the subscript u in the legend, are the results obtained from the uni-axial loading, while the dotted lines, denoted by the subscript b in the legend, are the results obtained from the bi-axial loading. The marked point, i.e. the intersection of the dashed line, the dotted line and the line with energy discrepancy equal to zero, denotes the predicted effective modulus E^* .

Figure 5.10 The energy discrepancy between the real film/substrate system (Figs. 5.9 (a) and (c)) and the equivalently homogenized film/substrate system (Figs. 5.9 (b) and (d)) in terms of various (E, ν) and h/t . E^* is the predicted effective Young's modulus. The subscripts, u and b , are referred to the simulations under uni-axial loading and bi-axial loading, respectively.

Figure 5.11 The mean value of the effective Young's modulus of polycrystalline copper thin film attached on various substrates. The thickness ratio h/t between the substrate and the film ranges from 0 to 50. The case, $h/t = 0$, refers to the free-standing thin film with $t/d = 0.1$.

Likewise, the foregoing procedure is carried out to evaluate the effective Young's modulus of the copper thin film attached to other substrates, i.e., polyimide, silicon and silicon carbide, with the ratio h/t varying from 1 to 50. All the numerical results for the effective Young's modulus of the copper thin film attached on different substrate are plotted in terms of the ratio h/t in Fig. 5.11.

It is observed that the effective modulus of the attached thin film varies dramatically when the ratio h/t is less than 10, and the value becomes stable after $h/t > 10$. It implies that the substrate material in the range $h/t = 10$ plays dominant role in controlling the deformation of the grains aggregating the film.

Also recognized from Fig. 5.11 is that the effective modulus of the attached thin film increases with increasing the substrate stiffness. When the substrate stiffness is much lower than the film stiffness, the deformation of the grains in the film is not constrained by the substrate. Hence, the effective modulus of the thin film attached on the polyimide is close to the equivalent value of the free-standing thin film. When the stiffness of the substrate increases, the constraint stemming from the underlying substrate goes up. The extent of the confinement depends on the stiffness contrast between the film and the substrate. For a copper film deposited on a silicon substrate, its effective modulus reaches to 126GPa which is close to the value of polycrystalline copper in bulk form. Because the substrate stiffness is comparable with the stiffness of the film, the deformation of the grains in the film cannot be completely restrained by the substrate. Therefore, the effective Young's modulus still deviates from the Voigt upper bound. In the case of a polycrystalline diamond substrate, the film's effective modulus is 130.0GPa. Only 1% discrepancy can be found in comparison with the Voigt bound of the effective Young's modulus of polycrystalline copper

aggregates in the state of plane stress (131.3GPa, obtained in Chapter 3). This observation can be explained as follows. For the substrate-attached thin film, the free surface leads the out-of-plane stresses, namely σ_{31} , σ_{32} and σ_{33} , to be zero everywhere. This stress state is corresponding to the state of plane stress. Because the Young's modulus of polycrystalline diamond is more than 7 times of the modulus of bulk polycrystalline copper (127GPa), the grains in the film deform almost uniformly together with the substrate under the prescribed loading. Such mechanical behavior agrees very well with the Voigt model in the plane stress condition. Thus, the predicted Young's modulus is so close to the analytical value.

5.3.2 Sensitivity of the effective Young's modulus to the film configuration

In the above subsection, the attention is given to the influence of the substrate. The attached film has the constant ratio $t/d=0.1$ and is modeled as columnar grain structure. In this subsection, we consider another two parameters in the film/substrate configuration. The first parameter is t/d . The value of t/d is prescribed to be 0.1, 0.5 and 1 respectively. The second new aspect is the films with multiple layers of grains along the direction of film thickness. The specimens containing 3, 4 and 5 layers of grains are simulated. Each layer consists of 100 cubic grains with identical size $d \times d \times d$. The ratio h/d is prescribed to be 10. Silicon carbide and polycrystalline diamond are selected as the substrate to understand the role of the film configuration on the effective modulus. Tables 5.7 and 5.8 present the numerical results with respect to different ratio t/d and film configuration for the copper film attached on silicon carbide and diamond, respectively.

Table 5.7 Statistic data for the effective Young's modulus of polycrystalline copper film attached on a silicon carbide substrate. The sample size is $n = 30$.

Layer of grains	Number of grains	t/d	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
		0.1	128.35	1.53	1.19
1	400	0.5	126.05	1.54	1.22
		1	126.00	1.52	1.21
3	300	3	127.22	1.74	1.34
4	400	4	127.60	1.49	1.17
5	500	5	127.67	1.46	1.14

Table 5.8 Statistic data for the effective Young's modulus of polycrystalline copper film attached on a polycrystalline diamond substrate. The sample size is $n = 30$.

Layer of grains	Number of grains	t/d	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
		0.1	130.00	1.69	1.20
1	400	0.5	128.19	1.55	1.21
		1	127.85	1.52	1.19
3	300	3	127.78	1.75	1.37
4	400	4	128.00	1.47	1.15
5	500	5	127.96	1.45	1.13

In the case of the attached film made up of one layer of the grains, as the ratio t/d increases, the constraint of the underlying substrate gradually decreases. The reduction in the degree of uniformity associated with the grain deformation results in the decrease of the modulus. When there are multiple layers of the grains in the film, only does the film material close to the film-substrate surface sense the substrate's control. The mutual interaction and constraint among the grains become significant. Hence, the behavior of the grains in the attached multiple-layered film becomes more similar to a bulk polycrystalline aggregate.

5.4 Simulation of the sandwiched thin films

Figure 5.12 schematically shows a sandwiched thin film with columnar grain structure. The surrounding materials are much thicker than the film. Due to the constraint on the deformation of the grains imposed by the surrounding materials, the effective modulus of the sandwiched thin film is expected to be higher.

Figure 5.12 A sandwiched thin film with columnar grain structure. The sandwiched thin film consists of $(L/d) \times (L/d)$ grains with equal size $d \times d \times t$. Grains are represented by different grey levels.

A thin film consisting of 400 columnar grains with randomly assigned crystallographic orientation is simulated. The ratio t/d between the film thickness t and the grain size d is 0.1. Silicon carbide and polycrystalline diamond are chosen as the surrounding material. The thickness ratio h/t of the surrounding bodies below and above the film is prescribed to be 10, with reference to Fig. 5.12. Following the identical procedure for estimating the effective modulus of the substrate-attached thin film, the effective modulus of the sandwiched copper thin film can be determined. The statistic data from thirty different simulations are listed in the Table 5.9.

Table 5.9 Statistic data for the effective Young's modulus of sandwiched copper thin film. The sample size is $n = 30$.

Surrounding material	Number of grains	t/d	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
Silicon carbide	400	0.1	131.40	1.60	1.22
Polycrystalline diamond	400	0.1	134.52	1.58	1.17

In comparison with the substrate-attached thin film, the sandwiched thin film exhibits higher modulus for both surrounding materials. In the case of the sandwiched thin film, the upper and lower surfaces of the film interact with the adjoining materials. As a result, the grains in the sandwiched film experience more confinement on the deformation than those in the attached film. Much more uniform deformation is thus observed in the sandwiched film.

The data in Table 5.9 indicate that the predicted modulus is still less than the Voigt upper bound (144.69GPa), even the surrounding material is polycrystalline diamond.

To understand this phenomenon, let us revisit the Voigt model. For example, in evaluating the upper bound of the shear modulus, the assumption of uniform strain implies that each grain has one non-zero shear strain component and all the other strain components vanish. However, our numerical simulations do not satisfy all the Voigt conditions. Therefore, the estimated modulus of the sandwiched thin film is lower than the Voigt bound.

5.5 Effect of micro-morphology

For a heterogeneous material, the spatial randomness or the micro-morphological randomness (e.g. the micro-morphology describes the grain shape, size and spatial distribution in a polycrystal) and the material randomness (e.g., the material randomness is associated with the random distribution of crystallographic orientation for a polycrystal) are two important factors in numerical simulation. However, their influences on the effective elastic modulus are coupled. A series of contributions by Torquato (1997, 1998, and 2002) provided us some qualitative and quantitative insight of the micro-morphological effect on the effective modulus, although the author studied two-phase isotropic composites. Torquato (1997) expanded the effective modulus of the composites in terms of the modulus difference of the two phases and n -point correlation function characterizing the microstructure. He pointed out that the micro-morphology has a limited influence on the macro-scale effective modulus when the modulus contrast between the two phases is low. For a polycrystalline aggregate, which is a collection of grains with random orientation and geometrical feature described by the grain shape and size, Torquato's formula (1997) cannot be applied directly. Nevertheless, for the polycrystalline aggregates assembling the grains with

low or medium anisotropy, limited influence of the micro-morphology on the effective modulus is expected.

In the previous sections, our emphasis is put on studying the influence of the crystallographic orientation, while the grain shape and size are identical for each grain in an aggregate. Now we use a planar Voronoi tessellation to characterize the variation of the grain shape and size. We call it as Voronoi model to distinguish the foregoing uniform model. In the present section, the framework of Voronoi tessellation is demonstrated in brief. It is then used to estimate the corresponding effective modulus of the polycrystalline aggregates in the state of plane stress. A comparison is made between the effective modulus obtained from the Voronoi model and that obtained from the uniform model.

5.5.1 Voronoi tessellation

The Voronoi tessellation is a process of decomposing a n -dimensional space into space-filling Voronoi cells. Due to the topological similarity between the Voronoi tessellation and the real microstructure of a polycrystalline aggregate, it has been widely adopted in researching the polycrystalline aggregates (Kumar, 1992; Kumar *et al.*, 1996; Mullen *et al.*, 1997; Barbe *et al.*, 2001a, b; Weyer *et al.*, 2002; Zhao and Tryon, 2004; Kamaya, 2006).

The basic procedure of tessellating a polycrystalline aggregate by Voronoi algorithm is demonstrated as follows (Aurenhammer, 1991). Consider a polycrystalline aggregate composed of N grains in a given space (e.g., a unit cube in three-dimensional space or a unit square in two-dimensional space). Firstly, N points, which are regarded as the nuclei of the grains, are randomly generated in the space with uniform distribution.

Secondly, the grain boundary is formed according to the nearest-neighbor rule, that is, the points in the space which are nearer to nucleus i than to any other nuclei are assigned to the nucleus i . For example, there are two distinct points or nuclei, p and q , in the space. A perpendicular line (two-dimensional case) or plane (three-dimensional case), which forms a boundary and divides the original space into two sub-spaces, is generated to bisect the line joining points p and q . Apparently, each sub-space contains the points that are closer to one of the two nuclei. By determining the boundaries between each nucleus and its neighboring nuclei, the given space is thus discretized into N polygons (two-dimensional case) or polytopes (three-dimensional case). Such polygons or polytopes are usually called Voronoi cells and the set of complete polygons or polytopes is called Voronoi diagram. Figure 5.13 illustrates a Voronoi diagram with six points in the plane. The dashed lines denote the lines joining the nearest two nuclei. The solid lines, which bisect the dashed lines, correspond to the boundaries of Voronoi cells.

Figure 5.13 Planar Voronoi diagram with six points.

5.5.2 Effect of micro-morphology on the effective modulus

To shed some light on the influence of micro-morphology on the effective modulus, a two-dimensional specimen in the state of plane stress is considered. The specimen is

tessellated into 400 Voronoi cells via the Voronoi tessellation algorithm. Each Voronoi cell is regarded as one grain. The crystallographic orientations are randomly assigned to the grains. Hence, all the statistic features, such as the grain shape, size and orientation, are considered in the Voronoi model. For visualization, a realization of the Voronoi model containing 400 Voronoi cells is schematically plotted in Fig. 5.14 (a). Different grey level stands for different grain. The histograms for the number of edges made up of Voronoi cell and the area of Voronoi cell are presented in Figs. 5.14 (b) and (c). The solid lines are fitted by using normal distribution.

Figure 5.14 (a) A realization of planar Voronoi tessellation of the specimen consisting of 400 Voronoi cells; (b) histogram of the number of edges made up of Voronoi cell; (c) histogram of the area of Voronoi cell. The solid lines are fitted by normal distribution.

After a Voronoi model is built up, a standard two-dimensional free meshing technique is employed to ensure a proper meshing of grain boundaries. Considering the irregular shape of Voronoi cells treated as grains, six-node triangular plane stress element (i.e.,

Plane183) is selected to discretize the Voronoi cells. No element straddles the adjoining grains. Figure 5.15 shows a meshed Voronoi model containing 105,227 nodes and 52,344 elements after mesh refinement.

It is known that the stress singularity exists at the triple junctions of the grains, and the strength of the singularity is dependent on the degree of mismatch in the material properties of the adjoining grains at the triple junction. To fully characterize all the details in the stress field variation such as the stress singularity at the grain triple points, the requirement of computational capacity and time is overwhelming, especially when there are substantial grains in a specimen.

Figure 5.15 Schematic illustration of a meshed Voronoi model. The mesh is performed by using six-node triangular element.

To guarantee the accuracy of numerical results, not only does the high-ordered element type is employed, but also the process of mesh refinement is carried out. Although such mesh may not completely capture the precise stress distribution, the small zone of high stress concentration may have little influence on the total strain

energy via finite element analysis. Because our interest is to evaluate the homogenized effective modulus of polycrystalline aggregates in different geometric configuration based on the principle of energy balance, the impact of stress singularity at triple point is neglected.

Table 5.10 Statistic data for the effective Young's modulus of polycrystalline aggregates via plane stress approximation with Voronoi model and uniform model. The sample size is $n = 30$.

Material type	Micro-morphological model	Number of grains	Effective Young's modulus		
			Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
Aluminum	Voronoi model	400	70.14	0.13	0.18
	Uniform model	400	70.18	0.16	0.23
Copper	Voronoi model	400	116.32	1.91	1.64
	Uniform model	400	116.46	1.71	1.43
β -brass	Voronoi model	400	85.47	1.39	1.62
	Uniform model	400	85.12	1.96	2.30
Lithium	Voronoi model	400	7.46	0.19	2.51
	Uniform model	400	7.48	0.21	2.75

Four types of polycrystalline aggregates are simulated by the Voronoi model and uniform model to evaluate the corresponding effective Young's modulus. They are aluminum, copper, β -brass and lithium. The mean value and standard deviation of the effective modulus are listed in Table 5.10. It is noticed that the mean values of the predicted effective modulus via the Voronoi model and uniform model are very close, even for the highly anisotropic lithium. Conclusion may be made that the micro-

morphology, such as the grain shape and grain size, only slightly affects the effective modulus of the polycrystalline aggregates. A similar result has been obtained in Chapter 4 for the bulk polycrystalline aggregates. Note that the negligible influence of the micro-morphology is obtained for the selected polycrystalline materials and specific structure. For the polycrystalline aggregates with imperfect grain boundaries or the heterogeneous material with complex microstructure or with high material contrast among the constituent phases, further investigation should be conducted to understand the role of the micro-morphology.

In addition, it should be pointed out that the grain size in our simulation is used to normalize all the geometric dimensions. Therefore, the aforementioned study of the grain shape and size via the Voronoi model should be considered as a “relative grain shape and size” effect.

5.6 Summary

In this chapter, the proposed finite element scheme with statistic consideration is carried out to estimate the effective Young’s modulus of polycrystalline copper in the free-standing, substrate-attached and sandwiched film configurations.

For the free-standing thin film with columnar grain structure, the effective modulus decreases with reducing the ratio t/d (i.e., the ratio between the film thickness t and the grain size d). When the ratio t/d approaches to zero, the plane stress approximation can be considered as the limit of the free-standing thin film. When the number of the grains in one of the dimensions (i.e., y -direction) in the film plane shrinks, the corresponding effective Young’s modulus approaches the Reuss lower bound from the

above. For the free-standing film with multiple layers of the grains, the estimated effective modulus comes close to the value of its bulk counterpart from the below.

For the substrate-attached thin film, the influence of the substrate is examined. Four materials, i.e., polyimide, silicon, silicon carbide and polycrystalline diamond, are selected as the substrate. The thickness ratio h/t between the substrate and film varies from 1 to 50. The numerical results indicate that the effective modulus is affected not only by the ratio h/t , but also by the relative stiffness of the substrate to the film. The effective modulus of the attached copper thin film increases and becomes stable when the ratio h/t exceeds 10. When the substrate is “much softer” (i.e., polyimide) than the film, the film modulus is approximately identical to the equivalent value of the free-standing thin film; while the film modulus exceeds that of its bulk counterpart if the substrate is “much harder” (i.e., polycrystalline diamond) than the film. For the sandwiched thin film, silicon carbide and polycrystalline diamond are chosen as the surrounding material. The enhancement of the film modulus is observed in comparison with that of the attached thin film.

In summary, the effective modulus may vary from the Reuss lower value to the Voigt upper value when the geometric configuration of the film changes. The spread of the effective modulus could be as large as 20% for the polycrystalline copper aggregates.

The micro-morphology influence is investigated by using planar Voronoi tessellation to simulate the grain structure of the polycrystalline aggregates. Negligible difference in the modulus is observed by comparing the result obtained from the Voronoi model with that from the uniform model, even for the crystalline lithium with anisotropic ratio of 8.52. This indicates that the micro-morphology does not contribute meaningfully to the effective modulus of the polycrystalline aggregates.

Chapter 6 Copper Thin Wires

Polycrystalline thin wire is another important geometric configuration in MEMS and microelectronic devices. Its cross-sectional dimension is scaled down to the level of grain size due to the need of miniaturization. The effective mechanical properties and behavior of the thin wires would deviate from those of bulk counterparts. In this chapter, the finite element scheme with statistic consideration is applied to estimate the effective Young's modulus of polycrystalline copper in the thin wire configuration.

6.1 Polycrystalline thin wire

Polycrystalline thin wires of copper, aluminum, and gold have been widely used as interconnects in microelectronic packages such as microelectronic devices, sensors and MEMS (Khatibi *et al.*, 2005). During the process of fabrication and application, the thin wires often subject to large temperature cycles or mechanical loading. The mismatch of thermal behavior between the wire and the surrounding materials results in cyclic stresses. Micro-cracks or micro-voids may develop in the thin wires and thus increase the resistance in the circuit. Hence, to ensure the reliability and stability of these devices, we need to have a qualitative understanding of the mechanical properties of the polycrystalline thin wires.

It has been noticed that the microstructure of polycrystalline copper wire is different from those of thin film and bulk form, and may change with the line width and

fabrication technique (Besser *et al.*, 2001; Paik, *et al.*, 2006, Zhang *et al.*, 2009; Brandstetter *et al.*, 2010). An example of these observations was given by Zhang and co-workers (2009), where the microstructures of copper damascene lines with three different line widths of 850, 185 and 60nm were studied. The plane-view transmission electron microscopy (TEM) images indicated that the 850nm line has a mixture of polycrystalline and bamboo-like grain structures, while the 185nm and 60nm lines have almost bamboo-like structure. The bamboo-like grain structure means that there is only one grain spanning across the entire width of the wire.

Many researchers tried to characterize the mechanical properties of the thin wires through experimental measurements (Fallen *et al.*, 2001; Weiss *et al.*, 2002; Khatibi *et al.*, 2005; Dixit *et al.*, 2007). Khatibi and co-workers (2005) reported a lower Young's modulus for the copper wires with a typical bamboo-like structure and diameter ranging from 10 to 125 μm through micro-tensile testing, in comparison with the value of bulk form. Dixit *et al.* (2007) observed different values of hardness and Young's modulus at the top and bottom of the electroplated copper interconnects through nanoindentation test. It was explained as the result of variation in microstructural morphology, like the grain structure and crystallographic orientation. These experimental data shed the light that the polycrystalline wires could exhibit different modulus in comparison with the polycrystalline bulk solids.

6.2 Description of polycrystalline thin wire

A thin wire is defined, in the present work, as a structure with one dimension much larger than the other two dimensions. It means that the length of the wire is “very” long. The grain size d is the characteristic dimension of a polycrystalline wire. All the

macroscopic dimensions (i.e., the length of wire, the size of wire's cross-section) are normalized by the grain size.

To have some “common sense”, a set of simplified wires, shown in Fig. 6.1, is adopted. Figure 6.1(a) is called bamboo-structured thin wire with only one grain across its cross-section, and the wires with 2×2 , 3×3 and 4×4 grains in the cross-section are given in Figs. 6.1 (b), (c) and (d) respectively. As illustrated, each grain, represented by different grey level, has uniform shape (cube) and randomly assigned crystallographic orientation.

Figure 6.1 Schematic illustration of polycrystalline wires containing various numbers of cubic grains in the cross-section. (a) 1×1 ; (b) 2×2 ; (c) 3×3 ; (d) 4×4 . Grains are represented by different grey levels.

With the micro-morphology related discussion in Chapters 4 and 5, we know that the grain shape has negligible influence on the effective modulus of the polycrystalline

aggregates. In the following simulations, the cubic grain is used to model the polycrystalline wires in the grain structure. This simplification gives us an easier way to characterize the microstructure of the thin wire.

6.3 Simulation of the free-standing thin wires

A free-standing thin wire is modeled by a set of cubic grains with identical size $d \times d \times d$ and randomly assigned crystallographic orientation. The interface between the grains is assumed to be perfect. The effective Young's modulus in the direction of the wire length is evaluated by uni-axially stretching a thin wire.

With reference to Fig. 6.1, the boundary conditions are imposed as follows.

$$\begin{aligned}
 u_x &= 0 & \text{at } x = 0, \\
 u_x &= \Delta & \text{at } x = L, \\
 u_y &= 0 & \text{along } x = 0, y = 0, \\
 u_z &= 0 & \text{along } x = 0, z = 0,
 \end{aligned} \tag{6.1}$$

where Δ is the prescribed displacement. The symbols, u_x , u_y and u_z , denote the displacement along the direction x , y and z in the global coordinate system, respectively. The non-specified boundary conditions are “traction-free”. From the finite element numerical data, we obtain

$$E = \frac{2LU}{\Delta^2 A}, \tag{6.2}$$

where A is the cross-sectional area of the specimen at $x = L$ and U is the total strain energy of the grains aggregating the polycrystalline wire.

6.3.1 Fundamental considerations in numerical simulation

In order to numerically estimate the effective Young's modulus, the modeled specimen should consist of sufficient grains so that it is a RVE. Furthermore, certain level of mesh refinement is needed to ensure “apparent” convergence of the numerical results. For the sake of illustration, a specimen, which geometric configuration is identical with that shown in Fig. 6.1(b), is used to show the “apparent” convergence. There are 4 grains spanning across the cross-section and 80 grains along the length of the wire in the specimen. Twenty-node hexahedral element, Solid186, is employed to discretize each grain.

Table 6.1 Statistic data for the effective Young's modulus of polycrystalline copper thin wire consisting of 4×80 grains shown in Fig. 6.1 (b). The sample size is $n = 30$.

Mesh level	Number of elements per grain	Effective Young's modulus		
		Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
R0	1	119.50	2.10	1.76
R1	8	118.98	2.07	1.74
R2	27	118.73	2.07	1.74
R3	64	118.65	2.07	1.75

The statistic data of the calculated effective modulus, based on a sample of $n = 30$ realizations, is listed in Table 6.1, with respect to the level of mesh refinement. The data indicate that the effective modulus converges from the “stiff” side with increasing the level of mesh refinement or the number of elements per grain. There is only 0.3% difference between the result obtained at the first mesh level (R1, each grain is subdivided into 8 elements) and the value obtained at the third mesh level (R3, each

grain is subdivided into 64 elements). Therefore, in the following simulations, all the numerical results are calculated at the R1 mesh level.

In order to determine how many grains are needed for a specimen to be considered as a RVE, three specimens, which geometric configuration corresponds to Fig. 6.1(b), are constructed. They are composed of 40, 80 and 160 grains along the wire length. Thus the total number of the grains for these three specimens is $40 \times 4 (=160)$, $80 \times 4 (=320)$ and $160 \times 4 (=640)$, respectively. For each specimen, thirty different simulations are sampled to obtain the effective Young's modulus. Table 6.2 lists the numerical results at R1 mesh level (i.e. each grain is meshed into 8 elements). It is in good agreement with the statistic sense that the standard deviation of the predicted modulus reduces as the number of the grains increases. The mean value is insensitive to the number of the grains when there are enough grains in the specimen. According to Eq. (4.4) and engineering acceptable tolerance ($|\mu - \bar{x}| / \bar{x} < 1\%$), it may be inferred that the specimen containing more than 160 grains can be considered as a RVE from the data in Table 6.2.

Table 6.2 Statistic data for the effective Young's modulus of polycrystalline copper thin wire shown in Fig. 6.1(b) aggregating various numbers of the grains. Each grain is meshed into 8 twenty-node hexahedral elements. The sample size is $n = 30$.

Number of grains	Effective Young's modulus		
	Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
4×40	118.71	2.56	2.16
4×80	118.98	2.07	1.74
4×160	118.87	1.24	1.04

6.3.2 Sensitivity to the grain structure

As shown in Figs. 6.1, there may be one or multiple grains forming the cross-section. Based on the aforementioned study, we construct the specimen aggregating about 640 grains in different geometric configuration as shown in Figs. 6.1. They have 1, 4, 9 and 16 grains in the cross-section of the wire. The numerical data of the effective modulus are presented in Table 6.3. Figure 6.2 presents the effective Young's modulus in terms of the number of the grains containing in the specimen's cross-section. Also included is the numerical result of bulk polycrystalline copper aggregates (127.13 GPa, the estimated effective Young's modulus of the cubic specimen containing $8 \times 8 \times 8$ grains).

Table 6.3 Statistic data for the effective Young's modulus of polycrystalline copper wire corresponding to Figs. 6.1 (a), (b), (c) and (d), respectively. Each grain is meshed into 8 twenty-node hexahedral elements. The sample size is $n = 30$.

Number of grains	Effective Young's modulus		
	Mean value (GPa)	Standard deviation (GPa)	Coefficient of variation (%)
1×640 (Fig. 6.1(a))	111.11	1.15	1.03
4×160 (Fig. 6.1(b))	118.98	1.24	1.04
9×70 (Fig. 6.1(c))	122.85	1.40	1.14
16×40 (Fig. 6.1(d))	124.86	1.24	0.99

6.4 Discussion and summary

The thin wire configurations studied in our numerical simulation are only indicative as they do not by any means cover all the possible configurations of the polycrystalline thin wires. Nevertheless, they demonstrate that the effective Young's modulus of a

free-standing thin wire can dramatically deviate from the equivalent value of its bulk counterpart, as shown in Fig. 6.2.

Figure 6.2 The mean value of the effective Young's modulus of polycrystalline copper wire with respect to the number of grains in the cross-section, along with the value of bulk polycrystalline copper (127.13GPa). Each value marked by '■' is based on a sample of thirty simulations.

In the case of the thin wire with only one grain in the cross-section, shown in Fig. 6.1 (a), the grains are serially connected together so that the stress field under the uni-axial tension is approximately uniform. Such condition is similar to the assumption of the Reuss model. As expected, there is only 1.5% difference between numerically estimated effective Young's modulus (111.11GPa) and the Reuss lower bound (109.44GPa).

When there are multiple grains spanning across the wire's cross-section, the deformation of grain is constrained by the neighboring grains in the same cross-section or along the length. Consequently, the induced stress field under the uni-axial tension

would depart from the uniform stress field. When more and more grains are added in the cross-section, the deformation field in the grains approaches to the situation of three-dimensional bulk configuration. Figure 6.2 presents the trend that the numerically estimated effective modulus of the polycrystalline copper wire approaches to the value of polycrystalline bulk solids as the number of the grains in the cross-section increases from 1 to 16.

In summary, the spread of the effective moduli could be as large as 15% for polycrystalline copper wire with various grain structures in the cross-section. Therefore, it is necessary to take the geometric configuration into account in determining the effective Young's modulus of the polycrystalline copper wires.

Chapter 7 Summary and Conclusions

7.1 Summarizing the results

To have a complete picture correlating the effective Young's modulus with the geometric configuration in micro- and macro- scale (i.e., Fig.1.2), all the numerical results are summarized in Table 7.1 and illustrated in Fig. 7.1 for reference. Also included in Fig. 7.1 are the Reuss lower bound and Voigt upper bound in the three-dimensional (3D) bulk configuration and the state of plane stress (p, σ).

As shown in Fig. 7.1, depending on the geometric configuration, the effective Young's modulus of the polycrystalline copper can be anywhere in between the Reuss lower bound and Voigt upper bound. Because the geometric configuration associated with the Voigt upper bound is not seen in reality, it is absent in Fig. 7.1. The relation between the geometric configuration and its corresponding effective modulus can be expressed as

$$\begin{aligned} E_{lower\ bound} &\leq E_{free-standing\ film} \leq E_{bulk} \leq E_{substrate-attached\ film} \leq E_{upper\ bound} \\ E_{lower\ bound} &\leq E_{free-standing\ wire} \leq E_{bulk} \leq E_{upper\ bound} \end{aligned} \quad (7.1)$$

It should be emphasized that no “damage mechanism” (e.g., micro-cracks, micro-voids, grain boundary sliding and compliant grain boundary) is introduced in our numerical simulations. The modulus variation stems only from the change of geometric

configuration. The mutual interaction and constraint among the grains are the essential factors.

Table 7.1 Summary of the effective Young's modulus of polycrystalline copper in various geometric configurations.

Geometric configuration	Number of grain rows	Number of grain layers	Effective modulus (GPa)	Remarks
Fig. 1.2 (a)	1	1	109.4	Reuss lower bound
Fig. 1.2 (b)	2	1	111.8	Plane stress simulation of the free-standing thin film ($t/d \ll 1$) with various rows of grains
	5	1	114.8	
	10	1	115.4	
Fig. 1.2 (c)	20	1	116.4	Plane stress simulation of the free-standing thin film ($t/d \ll 1$)
	20	1	120.4	
Fig. 1.2 (d)	20	2	124.2	3D simulation of the free-standing film with various layers of grains
	20	3	125.8	
	20	5	127.0	
Fig. 1.2 (e)			127.1	3D simulation of bulk configuration
Fig. 1.2 (f)	1	1	111.1	3D simulation of the free-standing thin wire with bamboo-like structure
	2	2	119.0	3D simulation of the free-standing thin wire with multiple grains in the cross-section
	3	3	122.9	
	4	4	124.9	
	10	5	128.0	3D simulation of the film with various layers of grains attached to a diamond substrate ($h/d = 10$)
	10	3	127.8	
Fig. 1.2 (h)	20	1	127.9	3D simulation of the thin film attached to a diamond substrate ($t/d = 0.1$, $h/d = 10$)
	20	1	130.0	
Fig. 1.2 (j)	20	1	134.5	3D simulation of the thin film sandwiched between two diamond solids ($t/d = 0.1$, $h/d = 1$)
Voigt model			144.7	Voigt upper bound

Figure 7.1 Summary for the effective Young's modulus of polycrystalline copper in various geometric configurations (thin wire, thin film and bulk form) through finite element simulation. The Voigt and Reuss bounds on the effective Young's modulus of three-dimensional (3D) bulk configuration and two-dimensional plane stress state (p, σ) are included.

The anisotropy of a crystallite is defined by anisotropic ratio (AR). The spread between the lower and upper bounds of the polycrystalline aggregates positively correlates with its single crystal AR value. For the cubic crystal with low anisotropy, such as aluminum which has AR of 1.22, its effective modulus associated with the polycrystalline aluminum varies in the range from 70.1GPa (Reuss lower bound) to 70.7GPa (Voigt upper bound). Thus, we do not expect that the modulus of the polycrystalline aluminum in the film or wire configuration sufficiently deviates from the equivalent value of its bulk counterpart. Single crystal copper is moderately anisotropic with an AR of 3.21. Therefore, the geometric configuration should be taken into account for the polycrystalline copper in evaluating the effective modulus.

For the cubic crystal with high anisotropy, the modulus variation should be drastically large in different geometric configurations.

7.2 Conclusions

The objective of this thesis was to explain the modulus difference of polycrystalline aggregates in various geometric configurations (i.e., wire, film and bulk configurations) from a new perspective, i.e., the constraint and interaction among the grains. A finite element scheme in combination with statistic consideration was developed to systematically investigate the effective Young's modulus of the polycrystalline copper in different geometric configuration.

Grains in a polycrystalline aggregate were given a random distribution of crystallographic orientation. Each of the grains was simplified as cubic shape in three dimensions or square shape in two dimensions. Statistic concept was incorporated to infer the statistic data (population mean value and standard deviation) of the effective elastic properties from a sample of limited simulations (sample mean value and standard deviation).

Firstly, we applied the proposed finite element scheme with statistic consideration to predict the effective elastic constants of the polycrystalline aggregates in a three-dimensional bulk configuration. Four materials with cubic symmetry, i.e., aluminum, copper, β -brass and lithium which anisotropic ratios are 1.22 (low), 3.21 (moderate), 5.30(very high) and 8.52(extremely high) respectively, were considered. To determine the necessary number of the grains in a representative volume element, the specimens consisting of different number of the grains were simulated to see the convergence of the elastic constants. To understand the effect of the microstructural morphology (i.e.,

the grain shape, size and distribution), two models were further established. The first one was to use tetrahedron-shaped grain. The second one was to model the process of regular crystallization. The numerical simulations are summarized as follows.

The mean values of the effective elastic constants (Young's modulus and Poisson's ratio) become stable and approach the analytical solutions (Hill's average) with increasing the number of the grains or the specimen size, while the corresponding standard deviations inversely reduce. The required number of the grains in a representative volume element is positively correlated with the anisotropic ratio (AR) of the crystal. For the copper ($AR = 3.21$) of our interest, more than 196 grains are needed. When the specimen contains sufficient grains, convergence of the numerical results is observed. The micro-morphology slightly contributes to the effective elastic properties of the polycrystalline aggregates, even for the extremely anisotropic crystalline lithium ($AR > 8$). Hence, it is reasonable to use a simple uniform model to simulate the real microstructure of the polycrystalline aggregates.

Secondly, the proposed finite element scheme with statistic consideration was carried out to investigate the effective Young's modulus of the polycrystalline copper in the free-standing, substrate-attached and sandwiched film configurations, respectively.

For the free-standing film with columnar grain structure, referred to as "thin film" in the present work, the modulus sensitivity to the film thickness was studied by simulating specimen with t/d (i.e., the ratio between the film thickness t and the grain size d) varying from 0.1 to 1. It is noted that the effective Young's modulus reduces with decreasing the ratio t/d . The plane stress condition can be considered as the limit of the free-standing thin film for t/d tends to zero. In addition, the influence of the grain structure in the film plane and along the film thickness was investigated. When

the number of the grain rows in the y direction (shown in Fig. 1.2) shrinks from 20 to 2, the effective Young's modulus reduces from a value associated with the free-standing thin film (116.4GPa) to a value (111.8GPa) closed to the Reuss bound (109.4GPa). This phenomenon is attributed to the reduced constraint on the deformation of a grain from the neighboring grains. For the specimen with two rows of the grains, each grain subjects to almost uniform stress under the uni-axially tensile loading. Such condition is similar to the assumption of the Reuss model. As expected, our numerical simulation predicts the Young's modulus of 111.8GPa. For the free-standing multiple-layered film, the effective Young's modulus increases towards the equivalent value of bulk counterpart as more and more grain layer are added. This is due to the increased grain interaction relative to that in the thin film. It is seen that the predicted effective moduli are 125.8GPa and 127.0GPa for the specimen containing three and five layers of the grains, respectively.

For the substrate-attached thin films, we examined the influence of substrate stiffness and substrate thickness on the effective modulus of the attached copper film. Four materials, i.e., polyimide, silicon, silicon carbide and polycrystalline diamond, were selected as the substrate. The thickness ratio h/t between the substrate and the film varied from 1 to 50 when the attached thin film with the ratio t/d of 0.1 was simulated. Two features can be observed from the numerical results. The first feature is that the attached film behaves like a free-standing thin film when the substrate stiffness (polyimide) is much softer than the film; while the attached film deforms together with the underlying substrate when the substrate stiffness (polycrystalline diamond) is much higher than the film. For the latter case, there is only 1% discrepancy between the evaluated Young's modulus and the Voigt upper bound in the state of plane stress. The second feature is that the effective Young's modulus of the attached thin film

dramatically goes up when the ratio h/t is less than 10, and the value becomes stable after $h/t > 10$. Furthermore, the dependence of the effective Young's modulus on the attached film configuration was studied. When the ratio t/d for the attached film with columnar grain structure increases from 0.1 to 1, the evaluated Young's modulus approaches to the equivalent value of bulk counterpart from the above. The simulation for the attached film with multiple layers of the grains indicates that the evaluated Young's modulus approaches to the value of bulk form. It is the result of increasing grain interaction and reducing constraint from the underlying substrate. For the sandwiched thin films, silicon carbide and polycrystalline diamond were chosen as the surrounding material. The enhancement of the film modulus is observed in comparison with that of the attached thin film.

In summary, the effective modulus may vary from the Reuss lower value to the Voigt upper value when the geometric configuration associated with the film changes. The spread of the effective moduli could be as large as 20% for the polycrystalline copper film.

Thirdly, we simulated the free-standing polycrystalline copper thin wire with various grain structures in the cross-section. When the thin wire has a bamboo-like grain structure, the estimated Young's modulus is very close to the Reuss lower bound. As the number of the grains spanning across the cross-section increases, the effective modulus increases and approaches to the equivalent value of bulk counterparts. The spread of the effective moduli could be as large as 15% for the polycrystalline copper wire with various grain structures in the cross-section.

Lastly, the influence of the micro-morphology on the effective Young's modulus was studied by using planar Voronoi tessellation to simulate the grain structure of the

polycrystalline aggregates. Comparison was made between the numerical results obtained from the Voronoi model and those from the uniform model. It is noted that the micro-morphology does not contribute meaningfully to the effective modulus. Even for crystalline lithium with a very high anisotropic ratio ($AR > 8$), the micro-morphologic influence on the effective modulus is negligible.

With all the numerical simulations in mind, we can conclude

1. Anisotropic ratio is the most important parameter in the simulation of the polycrystalline aggregates.
2. The geometric configuration of device is a key factor in determining the effective Young's modulus of the polycrystalline aggregates.
3. The real micro-morphology has negligible influence on the effective Young's modulus of the polycrystalline aggregates.

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