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Interfacial effect on strengthening nanoscale metallic multilayers - a combined Hall-Petch relation and atomistic simulation study

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ABSTRACT

Besides the modulus difference, coherent stress and structural barrier mechanisms, the critical shear stress (CSS), which represents the resistance to the transmission of dislocation cross the interface, is another key factor influencing the hardness of nanoscale metallic multilayers (NMMs). However, it is very difficult to measure the CSS experimentally. In this study, we estimate the CSS based on the Hall-Petch relation via the available experimental hardness versus layer-thickness data. The obtained values are then verified by molecular dynamics simulations. With the proposed approach, 20 different NMMs are investigated systematically. The relative hardening contribution from the layers of the two constituent elements are quantified and these 20 NMMS grouped into two types except for one NMM. One group includes 12 NMMs whose interface hardening is contributed from both constituent element layers with different percentage. The other group includes 7 NMMs whose hardening contribution is mainly from the soft layer. We find that the peak hardness is not very sensitive to the CSS between the two constituent elements, rather is mainly determined by the average hardness of constituent layers. Moreover, it is more beneficial if the hard layer is about twice as hard as the soft layer for achieving high peak hardness.

1. Introduction

Nanoscale metallic multilayers (NMMs) represent an important class of engineering materials that are made up of alternating nanometer-scale layers of two different metallic materials. A range of technologically interesting properties including increased hardness and yield strength achieved in these materials at the nanometer scale make them attractive for a wide range of applications [1–6]. Experimental data accumulated over the past few decades have clearly demonstrated that the hardness and yield stress of NMMs may increase five to ten times when the grain size is reduced from the macroscopic to the nanometer range [1,6,7]. Several models for the strengthening have been proposed, including Hall–Petch (HP) dislocation pile-up [7], Orowan strengthening [8] and the combination of these two models. As suggested by Clemens et al. [9], the strengthening of NMMs is ultimately controlled by two parameters, one is the size effect, i.e., the layer thickness and grain size, and the other is the interfacial strength or the grain boundary strength to resist dislocation transmission.

There are two types of barriers for dislocation transmission, phase interfaces and grain boundaries, both of which are present in polycrystalline NMMs while only phase interfaces are present in single crystal NMMs. It is noted that the interfacial strength, the critical shear stress (CSS, τ^*) for the dislocation to transmit across the interface, is an explicit parameter in the HP model, while it is the lower limit for the Orowan model [11]. Hence, an estimate of the CSS required for dislocations to transmit across the interfaces is important to evaluate the mechanical strength of various NMMs. However, it is very difficult to experimentally obtain the CSS value for NMMs.

On the other hand, the hardness (H) of NMMs increases with the decrease of the individual layer thickness until H reaches its peak value at around layer thickness of ~10 nm [6,10,11]. Here we only consider NMMs with layer thickness that does not vary. One of the commonly used models for predicting the size-dependent hardness of NMMs is the HP relation, which takes the form of $H = H_0 + kh^{-1/2}$, where H is the hardness of the NMMs, h is the thickness of each layer, k is the HP slope and H_0 is the hardness of the multilayers with infinitely large layer thickness, representing

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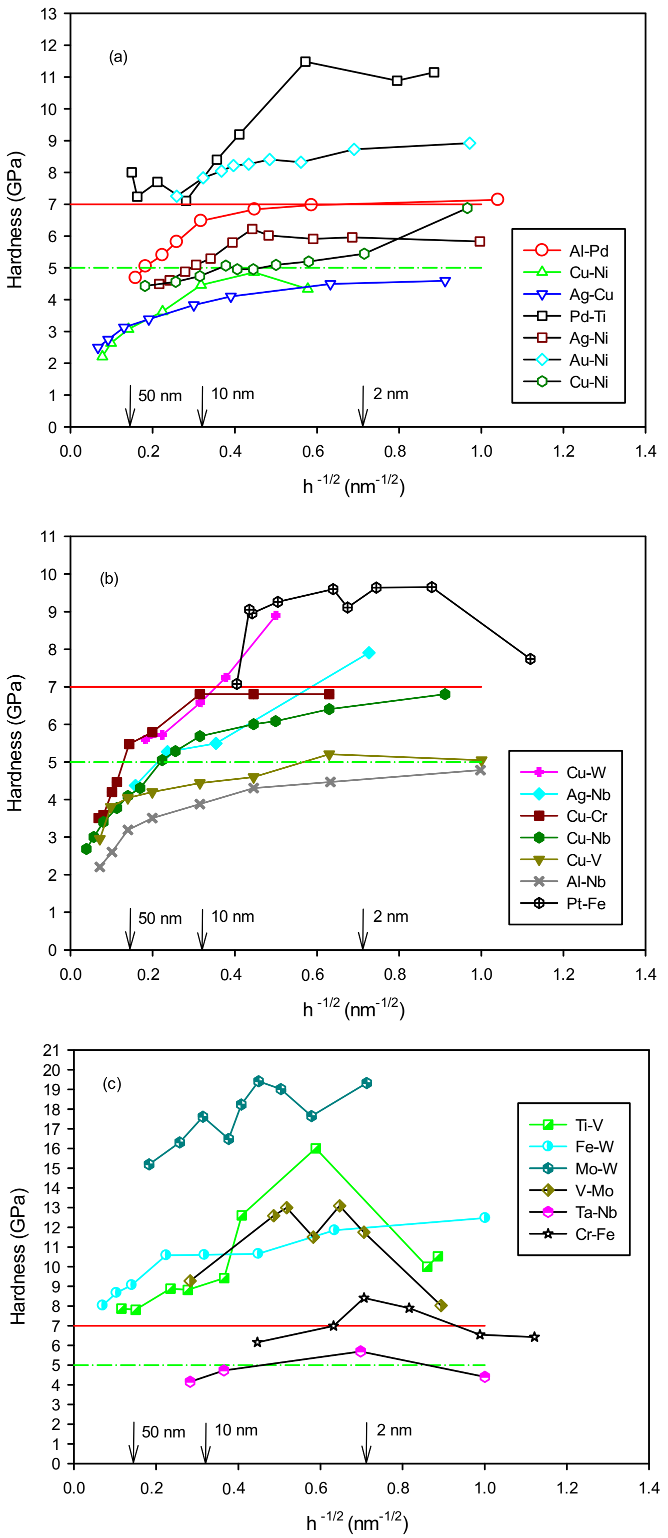

Fig. 1. Experimental data showing the H-h$^{-1/2}$ relationships from literature (The references are listed in Table 1) for the 20 NMMS, which are grouped as (a) fcc/fcc, (b) fcc/bcc, and (c) bcc/bcc types, with the green dashed line at 5 GPa and the read solid line at 7 GPa (red solid) for easy comparison. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the intrinsic resistance of the crystal lattice to dislocation movement. However, this model is only valid at the length scale where dislocation pile-ups can be treated as a continuum. At smaller scales, too few dislocations reside in the pile-ups to be treated as a continuum. As a result, a modified model considering other strengthening mechanisms is required.

Table 1
Crystal structure, enthalpy of formation predicted from Miedema's theory [24], peak hardness (H_{peak}), nanohardness (H_{nano}) for each constituent element, and average hardness (H_{ave}) from rule-of-mixture for the 20 metallic multilayers.

	A	B	ΔH (J/mol)[a]	H_{Peak} (GPa)	H_{nano} A (GPa)	H_{nano} B (GPa)	H_{ave} (GPa)	Ref.
Al/Pd	fcc	fcc	−46	7	1.57	4.27	2.92	[23]
Cu/Ni	fcc	fcc	3.5	4.8	1.8	6.4	4.1	[6]
Cu/Ni[b]	fcc	fcc	3.5	6.6	1.8	6.4	4.1	[17]
Ag/Cu	fcc	fcc	2	4.2	2	2.1	2.05	[6]
Ag/Ni	fcc	fcc	15.2	6.2	2	6.4	4.2	[17]
Au/Ni	fcc	fcc	7	8.9	4.6	6.4	5.5	[17]
Pd/Ti	fcc	fcc, hcp	−64	11.5	3.9	8.6	6.25	[15]
Cu/W	fcc	bcc	22	8.9	2.1	12.12	7.11	[13]
Ag/Nb	fcc	bcc	16	8	2	4.97	3.49	[14]
Cu/Cr	fcc	bcc	12	6.8	2.1	2.82	2.46	[6]
Cu/Nb	fcc	bcc	3	6.8	2.1	4.97	3.54	[6]
Cu/V	fcc	bcc	5	5.2	2.1	2.36	2.23	[12]
Al/Nb	fcc	bcc	−18	4.8	1.57	4.97	3.27	[12]
Pt/Fe	fcc	bcc	−13	9.64	1.97	4.81	3.39	[20]
Ti/V	bcc, hcp	bcc	−1.6	16	8	12	10	[16]
Fe/W	bcc	bcc	0.02	12.5	4.6	8.8	6.7	[22]
Mo/W	bcc	bcc	−0.2	19.4	9.4	16.3	12.85	[18]
V/Mo	bcc	bcc	0.02	13.09	2.36	4.98	3.67	[19]
Ta-Nb	bcc	bcc	0.02	5.69	1.35	4.97	3.16	[21]
Cr/Fe	bcc	bcc	−1.5	8.41	2.82	4.81	3.82	[20]

[a] The data are collected from ref. [24].
[b] The second set of experimental data for Cu/Ni NMM from ref. [17].

Fig. 1 reports the experimentally measured H-$h^{-1/2}$ relationships for 20 NMMs from the open literature [6, 12–23]. For the purpose of simplicity, we have used "A/B" to label each nanoscale metallic multilayer which is in fact in the format of "A/B/A/B…A/B/A/B". And These 20 NMMs are grouped as fcc/fcc, fcc/bcc, and bcc/bcc types. Their crystal structure, enthalpy of formation predicted from Miedema's theory [24], peak hardness (H_{peak}), nanohardness (H_{nano}) for each constituent element, and average hardness (H_{ave}) from the rule-of-mixture for 20 metallic multilayers are listed in Table 1. From Fig. 1, it can be found that a linear relation exists between H and $h^{-1/2}$ when h is larger than ~50 nm for most of the reported NMMs. In other words, the HP relation holds down to ~50 nm of h for these NMMs. The peak hardness (H_{peak}) of many NMMs occurs below ~10 nm of h. To predict H_{peak} of the NMMs with small h (< 50 nm) where the HP extrapolation does not apply any more, several models have been developed. By considering the modulus difference of constituents, dislocation core effect, and finite width of layer interface, Chu and Barnett [25] proposed a theoretical model for calculating the upper and lower bounds of H for NMMs. Moreover, first proposed by Anderson et al. [26] and then developed by Misra et al. [27], the confined layer slip (CLS) method of single dislocation loops combined with the HP model (CLS-HP) can explain an increase in strength with decreasing h at length scales where dislocation pile-up-based HP model does not apply. Many applications are motivated since this model proposed [28–33]. However, one of the challenging tasks here is how to determine the values of H_0 and k in the HP relation for a given NMM as these two parameters represent the intrinsic properties of multilayers with infinitely large layer thickness.

Theoretically, the value of the HP slope k can be obtained by delineating the experimentally measured H-$h^{-1/2}$ data points at large layer thicknesses. As can be seen from Fig. 1, however, it is very difficult to precisely determine the value of k by delineating the experimental data as the range of layer thickness h suitable for the HP relation is not well defined. For example, Fig. 2 shows the collected H-h$^{-1/2}$ experimental data for Fe/W NMM. The blue line