Revealing thickness dependence of hardness, strain rate sensitivity, and creep resistance of nano-crystalline magnesium/titanium multilayers by nanoindentation

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Abstract
The size effect on mechanical properties of hcp/hcp multilayer has been short of understanding hitherto. In this study, we prepared Magnesium/Titanium (Mg/Ti) multilayers with various individual layer thicknesses ($h = 50, 100, 150, 200$ nm) by magnetron sputtering. Relying on nanoindentation, hardness and elastic modulus were little changed for the multilayers with $h$ of 50 $\sim$ 150 nm, while they were evidently lower when $h$ increased to 200 nm. The determined strain rate sensitivities were 0.029, 0.032, 0.035, and 0.062 for the samples with $h = 50, 100, 150, 200$ nm, respectively. According to evolution of grain size, it suggests that Hall-Petch law dominates the strengthen effect of this hcp/hcp multilayers, rather than blocking effect of interface and decrease of dislocation content by reducing individual layer thickness. On the other hand, creep resistance was gradually promoted as reducing $h$. It indicates that interface and dislocation content could be important for the time-dependent plastic deformation. The room-temperature creep mechanism was discussed based on the strain rate sensitivity of steady-state creep flow. It indicates that dislocation glide could be suppressed and grain boundary glide, dislocation climb, and even interface glade could be plausible when $h$ decreased to 50 nm.

1. Introduction

Nanoscale metallic multilayers (NMMs) have attracted lots of attentions for their excellent properties in mechanics [1–3], optics [4], and magnetics [5]. The NMMs are usually prepared by layer-by-layer deposition technology [6, 7]. Compared with traditional bulk alloys, metallic multilayers often exhibit special mechanical properties because of their periodic modulation structure. According to a large number of experiments, simulations, and theoretical studies, the strengthening properties observed in nanomaterials are mainly related to the increased interfaces in the multilayers [2, 8, 9]. At the same time, they are also the result of the combined effects of the individual characteristics of every single layer [10–12]. Therefore, by adjusting the single-layer thickness $h$, the intralayer structure and the interlayer interface structure of the metallic multilayers will change accordingly. And the microstructure and mechanical properties will show significant scale effects as well [6, 13–16]. At present, three deformation mechanisms under different $h$ have been determined, which are controlled by either the dislocation pile-ups mechanism ($h$: micron to sub-micron), the confined layer slip (CLS) ($h$: several tens of nanometers to several nanometers), and the interface barrier strength (IBS) ($h$:1–2 nm) [6, 17, 18].

Literature researches show that the studies on the strengthening mechanism of the multilayer system composed of cubic components are relatively extensive, such as face centered cubic/body centered cubic, or face centered cubic/faceted centered cubic type [9, 19–23]. And only a few works in literature have reported the multilayer system with the hexagonal close-packed (hcp) components [14, 16]. Recently, cubic/hexagonal close-
packed systems, such as Cu/Zr (fcc/hcp) [24], Mg/Nb (hcp/bcc) [25], Al/Ti (fcc/hcp) [14] multilayers, have begun to attract researchers’ attention. Compared with cubic metals, hcp metals own much fewer slip systems [26], which endows their NMM special microstructures and strengthening mechanisms [27]. And this also determines the poor plasticity of hcp metals. Common hcp lattice structure metals can exhibit suitable or even excellent properties in their own application. For example, magnesium metal with very high specific strength is used as structural materials [28], and zinc metal is widely used as a coating [29]. However, it cannot be ignored that the mechanical properties of hcp metal are generally poor. Therefore, the introduction of multi-layer strengthening to the hcp metal may greatly improve the mechanical properties of metallic thin films while retaining or even improving their original excellent properties.

In this study, two hcp metals which are magnesium and titanium were selected to prepare multilayers with various \( h \) (namely 50, 100, 150, and 200 nm, respectively). Magnesium and titanium layers are immiscible with each other and have a moderate lattice mismatch. The reports on Mg/Ti system in previous work are mostly focused on its hydrogen storage capacity and hydrogenation kinetics [30, 31], while the microstructure and mechanical properties of this system are not well-known. Lu et al [16] reported the microstructure of the magnetron sputtering Mg/Ti multilayers in detail. They provided two orientation relationships in this system and the strength difference between the micro-compression and the nanoindentation. However, the creep behavior of the Mg/Ti system has not been studied hitherto.

Nanoindentation is currently the most advanced technique in detecting mechanical responses at the micro/nano scales during shear–compressive deformation [32]. And it nowadays has been developed to study pretty a lot of mechanical and plastic properties, e.g., creep behavior [33], fracture toughness [34], spatial modulus distribution [35], strain rate sensitivity [36] and activation volume of dislocation nucleation [37] and/or shear transformation zone [38], more than the common hardness and elastic modulus. Relying on nanoindentation, Zhu et al [39] reported room temperature creep behavior of Ag/Fe multilayers, which prepared by the electron beam evaporation deposition method. And the length-scale-dependent indentation creep behavior of Ti/Al multilayers prepared by magnetron sputtering is revealed by Fu et al [14]. The current work aims to experimentally investigate the thickness-dependent strengthening behavior of Mg/Ti multilayers particularly their creep behavior using nanoindentation technique. And it also attempts to provide general implications on the thickness-dependent mechanical property and creep behavior of hcp/hcp multilayers.

2. Experimental details

Using a direct current (DC) magnetron sputtering system, Mg/Ti multilayers with a varying layer thickness (designed as ‘50’, ‘100’, ‘150’, ‘200’ nm, respectively) were alternately deposited on a clean silicon substrate at room temperature. Before deposition, the target was cleaned in acetone and ethanol for 15 min in an ultrasonic cleaning machine to remove surface stains and oxidation products. The base vacuum of the deposition chamber was maintained at less than \( 6 \times 10^{-4} \) Pa before each deposition. During the deposition, the working argon pressure in the chamber was about 0.5 Pa, and the sputtering power of both targets was 100 W. Under these conditions, the deposition rates of Mg and Ti were detected as 0.278 and 0.093 nm s\(^{-1}\), respectively. In all samples, a 5 nm thick Ti layer was firstly deposited on the silicon substrate since Si and Ti have relatively strong chemical bonds. Due to the active chemical properties of magnesium, the top layer of all samples is titanium for avoiding natural oxidation and corrosion. By using a scanning electron microscope (SEM, sigma hv-01–43) under the working voltage of 5.00 kV, the cross-sectional thickness and surface morphology of the multilayers were observed. The total thickness of each sample is \a~1200 nm and the true thickness ratio of magnesium to titanium increased from 1.15 to 1.32 as increasing \( h \). Energy dispersive x-ray spectroscopy (EDS) was used to analyze the chemical composition.

Nanoindentation measurement was carried out on the Agilent Nanoindenter G200 at an ambient temperature of 23 °C controlled by air conditioning. A standard Berkovich indenter was used to measure the hardness and elastic modulus through the continuous stiffness module (CSM) at a constant strain rate of 0.05 s\(^{-1}\). And the maximum pressed depth was 250 nm. Strain rate sensitivity (SRS) was obtained at pressed depth of 150 nm by performing the individual indentation method under three different strain rates of 0.25, 0.05, and 0.01 s\(^{-1}\), respectively. The room temperature creep behavior was studied by the constant-load holding method under Berkovich indenter and the initial holding depth was fixed at 150 nm. Creep tests were loaded by five different strain rates of 0.25, 0.1, 0.05, 0.025, and 0.01 mN/s, respectively. The load holding time was set as 250 s for all the tests. To avoid the substrate effect, the maximum indentation depth was less than 15% of the total thickness of the multilayers. In order to ensure the reliability of the nanoindentation results, at least twelve indents were performed on each sample.
3. Results and discussion

3.1. Microstructure

X-ray diffraction (XRD) patterns of pure Mg, Ti films, and Mg/Ti multilayers with different layer thickness $h$ are presented in figure 1. It is clear that the single-layer thickness $h$ has a great influence on the crystal structure of the Mg/Ti multilayer. Both Mg and Ti have strong Mg (0002) and Ti (0002) textures, with weak Mg ($\bar{1}100$), Ti ($\bar{1}100$), Mg ($\bar{1}012$) diffraction peaks. It can be seen that all the Bragg peaks of Mg (0002) and Ti (0002) shift to smaller angles slightly as $h$ increases. As $h$ decreases, Ti ($\bar{1}100$) peak gradually diminishes, and almost disappears when $h = 50$ nm. Additionally, Ti ($\bar{1}100$) peak is observed at $h = 200$ nm, but does not appear in smaller $h$. These indicate stronger preferentiality in orientation at smaller sizes of Mg/Ti multilayers [16].

Figures 2(a)–(d) show the cross-sectional microstructures of the Mg/Ti multilayers with different individual layer thickness $h$ under SEM. It can be seen that the interfaces of Mg/Ti multilayer are compositionally graded. The mixing enthalpy of Mg and Ti is positive, which means that there are no metal compounds according to the Mg/Ti binary phase diagram [40]. And it also shows strong columnar growth and obvious epitaxial growth for Mg/Ti multilayers in figures 2(a)–(d). Considering that Mg and Ti are both hcp metals with a moderate mismatch in lattice parameters between them, this growth pattern could be reasonable. And the strong Mg (0002)/Ti (0002) texture in figure 1 could also supports this point. According to the cross-section observation, the true thicknesses of Mg/Ti multilayers and individual layers could be obtained as listed in table 1. Notwithstanding the deposition conditions were fixed, the actual thickness ratio of Mg layer and Ti layer was approximately ranged from $1.15 \sim 1.32$ among the four multilayers. It is worthy noticing that the thickness of individual Mg layer is over enlarged with increasing $h$, of which real ratio increased from 1.048 (52.4/50) to 1.238 (247.6/200). While the true thickness of individual Ti layer is much closed to the designed value, of which ratio is around 0.91 ~ 0.93 for the four multilayers. Qualitatively, the increased oxidation of Mg layer at the higher $h$ could be the intrinsic reason for the higher ratio of Mg content. Hence, the larger the thickness of individual layer, the higher ratio of Mg to Ti content.

Figures 3(a)–(d) show the surface morphology of Mg/Ti multilayers with different layer thicknesses. It appears that the grain morphology of each sample presents hexagons in different sizes because of the double hcp crystal lattice system. Figures 4(a)–(d) is the statistical distribution of the grain size of Mg/Ti multilayer surface of figures 3(a)–(d). It shows that the grain sizes are approximately normally distributed. And grain sizes of the multilayers are nearly distributed in the range of 80–110 nm, 87–111 nm, 100–120 nm, 150–192 nm as the individual $h$ increased from ‘50 nm’ to ‘200 nm’, respectively. It can be seen that the surface grain size of the
multilayer increases with increasing individual layer thickness, whilst the enhancement is not pronounced when 
$h$ is below '150 nm'. It is suggested that the average grain sizes detected on surface are not consistent with the true 
thickness of individual layer $h$. This performance has also been observed in Al/Ti multilayers [14] and could be 
attributed to the similar deposition conditions by magnetron sputtering.

3.2. Nanoindentation hardness and elastic modulus

Figures 5(a) and (b) show the correlations between hardness, elastic modulus and pressed depth from 50 to 250 
nm of the multilayer with $h = 100$ nm by CSM as an illustration. Clearly, the results of ten independent indents 
exhibit well repeatability. For the herein multilayers belong to the typical soft film/hard substrate system, the 
substrate effect could easily occur as the pressed depth was beyond a certain value [41]. For the hardness, 
substrate effect could be recognized as the indenter pressed into about 180 nm that the CSM hardness appears to 
be slowly increased. While for the elastic modulus, substrate effect is more pronounced and appeared much 
earlier. It could be easily understandable that the elastic stress field beneath indenter is several times larger than 
the yield stress field, hence elastic response during nanoindentation testing could be much sensitive to the 
pressed depth for the film/substrate system [3]. Considering that the tip imperfection and surface roughness 
could also influence the accuracy of indentation result at the initial pressed depth [42], the average $H$ and $E$ in 
pressed depth range from 130–180 nm were selected.

![Figure 2. SEM cross-sectional views of Mg/Ti multilayers with different designed individual thickness of (a) $h = 50$ nm, (b) $h = 100$ nm, (c) $h = 150$ nm, (d) $h = 200$ nm. And the true individual layer thickness for both kinds of metals could be carefully obtained.](image)

| $h$ (nm) | Average thickness of Mg (nm) | Average thickness of Ti (nm) | Total Thickness (nm) | thickness ratio |
|----------|-----------------------------|-----------------------------|---------------------|----------------|
| 50       | 53.2                        | 46.0                        | 1191                | 1.157          |
| 100      | 107.5                       | 92.3                        | 1199                | 1.164          |
| 150      | 175.9                       | 136.1                       | 1248                | 1.292          |
| 200      | 247.6                       | 187.4                       | 1305                | 1.321          |

Table 1. The actual thickness of Mg/Ti multilayer with different layer thickness.
Figure 5 shows the obtained hardness of Mg/Ti multilayers with various layer thicknesses, respectively. The herein measured hardness values of pure Mg and pure Ti thin films are 0.9 and 6 GPa respectively, which are consistent with the previous report [43]. The hardness of Mg/Ti multilayers does not change monotonously with reducing layer thickness, as it was widely reported on the size effect of alloys and metals. By reducing individual layer thickness, the average hardness initially increased from 1.7 to 2.1 GPa (+23.5%) as the layer thickness reduced from 200 to 150 nm. However, hardness turns to be insensitive to individual layer thickness by further thinning the individual layer thickness, which gradually reduced to 1.98 GPa (-5.7%) and 1.87 GPa (-1.5%) as the layer thickness was reduced down to 100 and 50 nm, respectively. And clearly, the maximum hardness of Mg/Ti multilayers (2.11 GPa) appears in the sample with the individual layer thickness of 150 nm. For the multilayers with grain sizes ranging from 50 nm to 200 nm, the relationship between strength and grain size could be well described by the Hall-Petch law [14]. As shown in Figure 4, the increasing trend of the grain size on surface is insignificant as the layer thickness increases from 50 to 150 nm. Similar distributions in grain size of the multilayers indicate that the variation in hardness could be not obvious with changing layer thickness. However, the grain size of the multilayer at the individual layer thickness of 200 nm is greatly enhanced compared to other samples, which would result in a lower hardness value. This also indicates that the multi-layer modulation period has little effect on the grain size of Mg/Ti multilayers when the layer thickness ranges from 50 nm to 150 nm. Generally speaking, the deformation of Mg/Ti multilayers is mainly due to the dislocations slip in the soft phase layer Mg at this scale [21]. When the dislocations reach the interface, they are hindered by the interface and agglomerate [6]. As the individual layer thickness decreases and the number of interfaces increases, the hindering effect on dislocations becomes stronger. This always explains the strengthening mechanism of metal multilayers. However, the nanoindentation results suggest that the resistance of grain boundaries to dislocations is the key to dominating the hardness change. The hardness strengthening behavior in Mg/Ti multilayers is dominated by grain size, rather than the contribution of interface and individual layer size. As shown in Table 1, the ratio of Mg and Ti was little changed as increasing layer thickness from 150 to 200 nm. Hence, it should be clarified that the increase of soft component Mg could be insignificant to the hardness with increasing layer thickness.

As exhibited in Figure 5(d), the average values of elastic modulus were 77.14, 78.06, 78.62, and 72.52 GPa for the multilayers with layer thicknesses of 50, 100, 150, and 200 nm, respectively. Generally, $E$ is weakly correlated with individual layer thickness even though it is a little lower in the sample with 200 nm individual layer. Under the iso-stress condition, the ‘Mixed Rule’ can be used to estimate the average modulus of the two-component multilayer system [16], as follows:
where $E_{Mg}$ and $E_{Ti}$ are the elastic modulus of Mg and Ti respectively. $V_{fMg}$ and $V_{fTi}$ denote the volume fraction of Mg and Ti layer. Considering that the room temperature elastic modulus of pure Mg and pure Ti are 45 GPa and 120 GPa respectively, the elastic modulus of Mg/Ti multilayer calculated by equation (1) is also shown in figure 5(d), with a value of about 80 GPa. From the results, the Mg/Ti multilayer system well conforms to the mixing rule. It should be mentioned that the measured $E$ in the sample with $h$ of 200 nm was a littler lower than the predicted value (72.5 versus 77.3 GPa). Such tiny difference could be reasonable that the oxidation effect and lower density would influence the elastic response beneath indenter. The independences of individual layer thickness and grain size on the elastic modulus of multilayers could be reasonable for elastic modulus is an intrinsic mechanical parameter which concerns with the atomic bonding [39].

Strain rate sensitivity (SRS) plays an important role in studying the plastic deformation mechanism of materials [36, 44–46]. Figure 6(a) exhibits the typical nanoindentation load versus displacement ($P-h$) curves under various strain rates for the multilayer with $h = 50$, as an illustration. Clearly, higher load was required to reach the same displacement by increasing strain rate, which indicates a stronger resistance to the plastic deformation. Table 2 lists the hardness of Mg/Ti multilayers obtained under different strain rates and compared with the results by CSM test. Higher hardness could be detected under faster loading sequence, which means a positive strain rate sensitivity. Figure 6(b) shows the log-log correlation between the average hardness and strain rate for all the samples, it presents an almost linearly relationship. According to the following formula, the strain rate sensitivity index $m$ could be determined via linear fitting [47]:

$$m = \frac{\partial \ln H}{\partial \ln \varepsilon}$$

The specific values of $m$ are also given in the inset of figure 6(c), which are 0.029, 0.0322, 0.0348, and 0.0619 for the multilayers with individual layer thickness $h$ of 50, 100, 150, and 200 nm, respectively. Be similar with the variation trends of hardness on individual layer thickness, SRS is little changed for the samples with $h$ of 50, 100, 150 nm and precipitously enlarged when $h$ increased to 200 nm. It has a similar conclusion to the hardness change with individual layer thickness under CSM test. As for the metals with large grain sizes (larger than tens of
nanometers), the SRS of face-center-cubic (fcc) metals usually increases as the grain size decreases, while body-center-cubic (bcc) metals have the opposite behavior [48]. The current result manifests that the SRS behavior of the Mg/Ti (hcp/hcp) system is similar to the body-center-cubic (bcc) metals as the grain size changes. Besides, the herein estimated values of SRS for the hcp multilayers with nanoscale grain size are a bit higher than the reported data in fcc and bcc metals [49]. In comparison, SRSs of metal multilayers exhibited conflicted behavior as the individual layer size changes. For the fcc/bcc multilayers, SRS of Cu/V slightly increased from 0.018 to 0.024 as $h$ increased from 25 to 100 nm [50], SRS of Ag/W decreased from 0.08 to 0.047 as $h$ increased from 50 to 200 nm [51], and SRS of Cu/Ta is around 0.05 and independent on $h$ [52]. In Lu et al.’s work, SRS of Mg/Ti multilayer has been revealed as stable value in between 0.04 ~ 0.05 notwithstanding $h$ changed from 2.5 to 200 nm [16]. In another hcp/hcp (Mg/Zr) multilayer, SRS is also stable at 0.036 as $h$ increased from 10 to 100 nm [53].

3.3. Indentation creep behavior

Figure 7 exhibits the typical indentation creep $P$-$h$ curve by the constant-load holding method, as well as the load-time relationship in the inset. The irreversible displacement clearly occurs during the holding stage, which indicates the creep deformation happened at room temperature. The occurrence of indentation creep deformation in this Mg/Ti system is mainly high-stress activated beneath the Berkovich tip [54]. Be different to the conventional uniaxial creep test (always performed at high temperature), severe plastic deformation occurs during indentation process and abundant structure flaws such as dislocation, twins, and voids would be generated. Hence the time-dependent plastic deformation could be easily activated via the dislocation move and/or flaw evolution under the high stress (beyond the yield stress) during the holding stage, even though atom mobility is weak at room temperature. For all the samples, the creep flow curves, i.e., creep displacement versus holding time exhibit a well repeatability under the same loading condition as shown in the Supplementary Materials (available online at stacks.iop.org/MRX/9/046401/mmedia).

Figure 8 exhibits the typical creep curves under various loading strain rates for all the samples. In order to directly observe the creep flow, both the initial holding time ($X$-coordinate axis) and creep displacement ($Y$-coordinate axis) were set to zero. The experiment results clearly illustrate that creep deformation of Mg/Ti

![Figure 5. The correlations between (a) hardness, (b) elastic modulus and pressed depth for the multilayer with individual layer thickness of 100 nm by CSM testing. Accordingly, the average values of (c) hardness and (d) elastic modulus between 130 ~ 180 nm were recorded for the multilayers with four individual layer thicknesses.](image-url)
multilayers could be obviously enhanced by increasing loading strain rate. It is well-known that indentation creep deformation could be divided into two stages (lacks failure stage during traditional creep testing), namely instantaneous creep and steady-state creep. At the initial several to tens of seconds, creep displacement increased fast while the creep rate dramatically dropped with holding time, this stage could be called as instantaneous creep deformation. And then creep rate tended to be stable and creep displacement almost linearly increased with time, which named as steady-state creep stage. Based on the creep flow features in figure 8, it is obvious that loading strain rate effect mainly occurred at the initial holding stage. Both the creep rate and creep displacement of multilayers at the instantaneous creep stage were greatly enhanced by increasing strain rate. On the other hand, the creep rates of steady-state creep deformations under various loading strain rate were almost the same. This phenomenon could be explained from the perspective of apparatus data recording and intrinsic plastic deformation. At the ending of loading sequence, the indenter could not be completely static immediately and the ‘overshoot’ phenomenon would result in the increase of displacement at the very beginning of holding stage.

![Figure 6](image)

**Figure 6.** (a) Typical nanoindentation load versus displacement ($P$-$h$) curves under various strain rate for the multilayer with $h = 50$; (b) Hardness of Mg/Ti multilayers with different layer thickness under three different loading rates (0.25, 0.05, and 0.01 s$^{-1}$, respectively), SRS could be determined by linearly fitting the log-log correlation between hardness and strain rate.

**Table 2.** Hardness of Mg/Ti multilayers under various strain rates.

| Experimental conditions | $H_{h=50}$ (GPa) | $H_{h=100}$ (GPa) | $H_{h=150}$ (GPa) | $H_{h=200}$ (GPa) |
|-------------------------|------------------|------------------|------------------|------------------|
| 0.01 s$^{-1}$           | 1.86             | 1.79             | 1.98             | 1.46             |
| 0.05 s$^{-1}$           | 1.96             | 1.89             | 2.10             | 1.61             |
| 0.25 s$^{-1}$           | 2.05             | 1.99             | 2.21             | 1.78             |
| CSM (0.05 s$^{-1}$)     | 2.07             | 1.98             | 2.10             | 1.70             |
This could also be regarded as the continuation of quasi-plastic deformation during loading segment. Hence the higher loading strain rate, the larger the artificial error on the detected displacement of holding stage. On the other hand, the dislocation density beneath indenter is generally thought to be higher under higher strain rate, which is the reason for the stronger resistance to instant plastic deformation and higher strength or hardness.
While for the time-dependent plastic deformation, the higher existing dislocation density provides more ‘fertile place’ for dislocation move and therefore the pronounced creep flow.

Since the initial holding depth was constant at 150 nm for all the cases, the creep displacement at the end of holding stage could be briefly utilized to represent the creep resistance. Figure 9 records the total creep displacements which are plotted as a function of loading strain rates for all the samples. For all the multilayers, loading strain rate has a strong effect on the indentation creep deformation. Such phenomenon has been greatly reported in metals and alloys, which could be well explained from both extrinsic and intrinsic reasons [55, 56]. However, the loading history effect on the creep deformation of multilayers was rarely studied. Qualitatively, the enhancements of creep displacement for the multilayers with \( h = 50, 100, 150 \) nm were similar by increasing strain rate from 0.01 to 0.25 s\(^{-1}\). While for the multilayers with \( h = 200 \) nm, the loading strain rate effect on promoting creep flow is much more pronounced. Such result is coincidental with the behavior of hardness and SRS as changing individual layer thickness.

Figure 10 summaries the correlation between creep displacement and individual layer thickness under various loading strain rates. Clearly, the maximum creep displacement almost linearly increases with the increasing of layers’ thickness \( h \) for all the loading rates. It is worth noting that the layer thickness effect is pronounced on the creep deformation even for the \( h = 50, 100 \) and 150 nm. Be distinct to the aforementioned
hardness and strain rate sensitivity, it reveals the significant scale-dependent creep strengthening effect of Mg/Ti multilayer by reducing individual layer thickness and increasing interface. For the creep resistance is directly correlated with hardness, the enhanced creep resistance as \( h \) reduced from 200 to 150 nm could be expected due to the great promotion of hardness. On the other side, hardness is little changed or even slightly decreased by further reducing \( h \) to 100 and 50 nm as exhibited in figure 5(c). Hence it could be conceived that the promoted creep resistance is naturally attributed to the increased interfaces between Mg and Ti layers. The herein experimental result on the creep behavior of metal multilayers is well consistent with the previous reports [14, 39, 57, 58]. Although creep deformations, i.e., creep displacement and creep rate were various in those metal multilayers due to the different components (e.g., Cu/Ta [57], Cu/Co [58], Ti/Al [14], and Ag/Fe [39]), layer modulation and thickness, the exhibited creep resistances were generally enhanced by decreasing individual layer thickness. It has been widely reported that dislocation move such as dislocation glide and climb is the dominated indentation creep mechanism in alloys and metals at room temperature, rather than atomic diffusion and grain boundary glide [14, 57]. It could be indicated that the hindering effect on dislocation move by increasing interface between two nano-crystal layers plays an important role on the length scale effect on creep deformation in multilayers, which is independent on lattice structure. In the meanwhile, contents of dislocation and/or other defects in the sample could be conceived to be lower by reducing individual layer thickness, which in turn results in the suppression of creep flow via dislocation moves. However, it is difficult to recognize which one (introduce interlayer and reduce dislocation content) is the critical reason (or both are important) for the enhancement of creep resistance by decreasing \( h \).

In order to better understand the layer-thickness effect on creep deformation, the creep flow mechanism beneath indenter should be studied based on the strain rate sensitivity (SRS) [59]. Here we selected the multilayer film with \( h = 100 \) nm at strain rates of 0.05 s\(^{-1}\) to calculate the SRS during steady-state creep, as an illustration. Figure 11(a) shows a fitting curve of creep flow curve for the Mg/Ti multilayer by an empirical formula [45] (\( R^2 > 0.99 \)):

**Figure 11.** (a) Creep flow curve could be well fitted by an empirical equation. (b) Creep strain rate during holding stage. (c) Hardness during holding stage. (d) The log-log correlation between hardness and strain during holding stage, strain rate sensitivity could thus be determined by linear fitting the steady-state segment.
where \( h_0 \) and \( t_0 \) are the initial holding depth and the time at the beginning of the holding stage, and \( a, b, k \) are the fitting constants. Figure 11(b) shows the corresponding creep strain rate at each time point obtained by fitting the curve by:

\[
\dot{\varepsilon} = \frac{1}{h_c} \frac{dh_c}{dt}
\]  

Figure 11(c) shows the variation of hardness during holding stage by:

\[
H = \frac{P}{c h_c^2}
\]  

where \( P \) is indentation load, \( c \) is the coefficient of area function calibrated on standard fused silica and equal to 22.5 here, and \( h_c \) is the contact displacement which could be depicted as:

\[
h_c = h - \varepsilon \times \frac{P}{S}
\]

where \( h \) is pressed displacement, \( \varepsilon \) is a constant 0.72 for the three-pyramid tip, \( S \) is contact stiffness. Figure 11(d) shows a natural logarithm correlation between hardness and creep strain rate. The creep SRS could be estimated by linear fitting of its steady-state part according to equation (2). By this analysis, the SRS values of all the multilayers could be determined.

Figure 12 exhibits the SRS values as a function of individual layer thickness from the creep deformation with loading strain rate of 0.05 s\(^{-1}\). The SRS values determined by the rate-jump method were also presented as a contrast. The overall variation trends of SRS by two independent methods are similar, i.e., \( m \) enlarges with increasing \( h \), whilst the increasing rate of SRS is much high during creep deformation. The herein SRS variation trend confirms well with the previous reports in the indentation creep deformations of multilayers with different crystal components \([14, 39, 57, 58]\). It should be emphasized that the absolute value of SRS from nanoindentation creep is not a constant, which is strongly tied with creep deformation and hence changed with creep conditions. For instance, the estimated SRS would be enlarged with extending holding time, lowering the loading strain rate, and/or decreasing holding strain \([60]\). The non-uniform stress distribution and limited load-holding duration (be limited by thermal drift influence) are the critical reasons which result in the variable \( m \) during nanoindentation creep deformation. However, the current SRSs were determined from sufficiently steady-state creep flow beneath the self-similar Berkovich tip, which means the effects of loading history (mainly influences the instantaneous creep stage) and holding strain could be insignificant on the SRS values. Therefore, the herein SRS from creep test loaded by strain rate of 0.05 s\(^{-1}\) could be representable to qualitatively discuss the creep mechanism of Mg/Ti multilayers.

For the samples with \( h = 200, 150 \) and \( 100 \) nm whose SRS values are 0.144, 0.123 and 0.112, dislocation glide could be the dominating creep mechanism according to classic creep rule by conventional test when \( m \) falls in the range approximately from 0.1 \( \sim \) 0.3 \([61]\). While for the sample with \( h = 50 \) nm, the estimated SRS...
dramatically drops to 0.058, which is merely half of the sample with \( h = 100 \) nm. From the perspective of conventional creep mechanism, it implies that flow mechanism of creep deformation could be changed. Accordingly, we can conceive that the increased interface and decreased dislocation density as \( h \) decreased from 200 to 100 nm, are insufficient to change the dislocation-dominated creep mechanism beneath indenter. On the other hand, the size of 50 nm individual layer is much close to the required space for forming a developed dislocation \([62, 63]\). It could be expected that dislocation glide could be greatly suppressed during holding stage in the sample with \( h = 50 \) nm, due to blocking effect of abundant interfaces, very low density of residual dislocation, and limited space to activate new dislocations. Meanwhile, it has been revealed the average grain size of the sample with \( h = 50 \) nm could be lower than 100 nm as exhibited in figure 4. We are convinced that grain boundary (GB) glide, interface glide, and dislocation climb along the GBs and interfaces could appear rather than dislocation glide during steady-state creep deformation for the sample with \( h = 50 \) nm.

In summary, the current work reports a grain size-dominated strengthen mechanism during quasi-plastic deformation, i.e., hardness and strain rate sensitivity were mainly dependent on grain size of nano-crystalline multilayers, rather than individual layer thickness as it was widely reported. While for the time-dependent plastic deformation, i.e., creep resistance is strongly enhanced with reducing individual layer thickness. Based on the strain rate sensitivity of steady-state creep flow, the creep mechanism of Mg/Ti multilayers was carefully discussed.

4. Conclusions

In this work, we prepared nano-crystalline Mg/Ti multilayers with various individual layer thickness from 50 to 200 nm by magnetron sputtering. The thickness-dependent mechanical properties concern with both quasi-plastic and creep deformations were systematically revealed. According to experimental results, several conclusions could be obtained as following:

1. Hardness and elastic modulus were little changed for the samples with individual layer thicknesses \( (h) \) of 50, 100, and 150 nm, whilst they were obviously lower as \( h \) increased to 200 nm. Based on the statistical distribution of surface grain size, the Hall-Petch law well explains such ‘unusual’ weak dependence of strength on \( h \) in the nano-crystalline multilayers.

2. The correlation between strain rate sensitivity \( m \) of quasi-plastic deformation and individual layer thickness was revealed in the Mg/Ti multilayers. \( m \) quickly dropped from 0.062 to 0.035 as \( h \) decreased from 200 to 150 nm, and then it was stabilized around 0.03 for the samples with \( h = 100, 50 \) nm.

3. Indentation loading history plays an important role on the initial creep deformation that higher loading strain rate facilitates a more pronounced creep deformation for the Mg/Ti multilayers. The room-temperature creep resistance could be evidently strengthened by reducing individual layer thickness. The strain rate sensitivity of steady-state creep flow indicates creep mechanism could be changed when individual layer thickness was reduced to 50 nm.

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Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

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