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A nano-indentation study on the plasticity length scale effects in LIGA Ni MEMS structures

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This paper presents the results of a nano-indentation study of the effects of strain gradient plasticity on the elastic-plastic deformation of LIGA Ni MEMS structures plated from sulfamate baths. Both Berkovich and North Star indenter tips were used in the study to investigate possible effects of residual indentation depth between the micro and nano scales on the hardness of LIGA Ni MEMS structures. A microstructural length scale parameter, $\hat{l} = 0.89\ \mu$m, was determined for LIGA nickel films. This is shown to be consistent with a stretch gradient length scale parameter, l_s, of $\sim$0.36 μm.

1. Introduction

In recent years, LIGA (an acronym of the German words "Lithographie, Galvanoformung, Abformung") nickel MEMS structures have been developed for applications in larger and thicker devices [1–4]. These include applications in micro-switches [1, 2, 4] and accelerometers [3] that are used for the deployment of air bags. However, a basic understanding of the mechanical behavior of LIGA nickel MEMS structures is yet to be developed. Furthermore, the possible effects of size-scale on plasticity in LIGA Ni MEMS structures are yet to be fully explored.

The nano-indentation method was chosen in present study of size effects because it is the simplest and most controllable experimental techniques for the characterization of size effects on the plasticity of metals between the micro and nano scales. Traditionally, indentation tests can be used to estimate the yield stress by measuring the hardness at the macro-scale [5]. More recently, however, there have been several studies [6–10] which show that the measured hardness is dependent on the size or depth of indent when the residual indent size is on the order of tens of microns. This size dependence cannot be explained by the conventional continuum plasticity theories in which there are no length scale parameters [6, 9, 11].

During last three decades, there have been significant efforts to develop new plasticity theories that include: the effects of plasticity length scale parameters [6, 9, 11]. Two classes of plasticity theories have been developed. These include the phenomenological strain gradient plasticity theories proposed by Fleck and Hutchingson [11] and the mechanism-based strain gradient plasticity (MSG) theory developed by Nix and Gao [9], Gao *et al.* [12] and Huang *et al.* [13]. Most recently, micro-bend experiments have been conducted on LIGA Ni MEMS structures [14]. These show that plastic flow stresses in LIGA Ni MEMS structures are strongly affected by structural thicknesses in the range of between $\sim$25 and 200 μm. However, there have been no prior studies of size effects in LIGA Ni MEMS structures in the regime between the micro and nano-scales.

This paper presents the results of a combined experimental and theoretical study of the effects of strain gradient plasticity on the elastic-plastic deformation of LIGA Ni MEMS structures plated from sulfamate baths. The structure of as-received LIGA Ni films is characterized using scanning electron microscopy, and orientation imaging microscopy. Both Berkovich and North Star (cube corner) indenter tips were then used in the study to investigate possible effects of indenter tip and residual indent size on the hardness of LIGA Ni structures within micro and nano-scales. The results of the nano-indentation tests were used to extract the microstructural length scale parameter, using a mechanism-based model proposed by Nix and Gao [9]. This gives a microstructural length scale parameter, $\hat{l}$, of $\sim$0.89 μm, and a stretch gradient length scale parameter, l_s, of $\sim$0.36 μm.

2. Materials

LIGA is an additive microfabrication process in which structural material is deposited into a polymethylmethacrylate (PMMA) molds realized by deep X-ray photolithograpy (DXRL) [3]. The LIGA Ni film specimens used in this study were electrodeposited using sulfamate bath chemistry at Sandia National Laboratories, Livermore, CA. The bath composition and operating conditions are summarized in Table I. The details of the fabrication process were outlined by Christensen *et al.* [1] and Buchheit *et al.* [15].

The as-received samples were examined using optical microscopy (OM), scanning electron microscopy (SEM) and orientation imaging microscopy (OIM). These were used, respectively, to reveal the microstructures (Fig. 1a and b) and micro-textures (Fig. 2). The optical micrograph of the sample cross-section indicates the presence of predominantly columnar structures oriented parallel to the deposition direction (Fig. 1a). The columnar structures are approximately 5 μm wide and 5–25 μm long. At higher magnifications, the SEM micrograph of the surface of the as-received sample revealed very fine grains with sizes on the order of tens of nanometers (Fig. 1b). The predominantly red color in Fig. 2 is consistent with a sharp $\langle 100\rangle$ crystallographic fiber texture of the columnar laths shown in Fig. 1. A small fraction of smaller grains between the laths had distinctly different orientations from the predominant $\langle 100\rangle$ microstructure.

3. Hardness measurement

As-received LIGA Ni samples with rectangular dimensions (7 mm × 1 mm) and thickness of 140 μm were fabricated at Sandia National Labs, Albuquerque, NM. Hardness measurements were then performed on these specimens using nano-indenter tips of different types. Different peak loads were applied during indentation in an effort to explore the plasticity length-scale effect on the elastic-plastic deformation of LIGA Ni films between the micro and nano-scales.

The hardness measurements were performed using a TriboScope® (TriboScope is a registered trademark of Hysitron, Inc., Minneapolis, MN) Nanomechanical Testing System at Hysitron, MN. The force and displacement resolution of the Triboscope system are 1nN and 0.0002 nm, respectively. The noise floor for the force is 100 nN and the noise floor for the displacement is 0.2 nm. A sharp North Star indenter (a three-sided cube-corner type tip) and a Berkovich indenter (a three-sided pyramid type tip) were used in the hardness measurements. The loading rate was kept at 1000 μN/s, and a holding period of 15 s was applied. Peak load ranges between 200 μN to 11000 μN were explored in an effort to study the effects of impression size from a few microns to tens of nanometers.

Contact mode atomic force microscopy scans were also obtained before and after the indentation tests. These were used to ensure that the indentations were only performed on spots with relatively low roughness. This minimized the possible effects of RMS on hardness measurement [16]. A load-displacement curve was recorded during each test. Finally, from known area functions of the calibrated tips, hardness and Young's modulus were calculated [17].

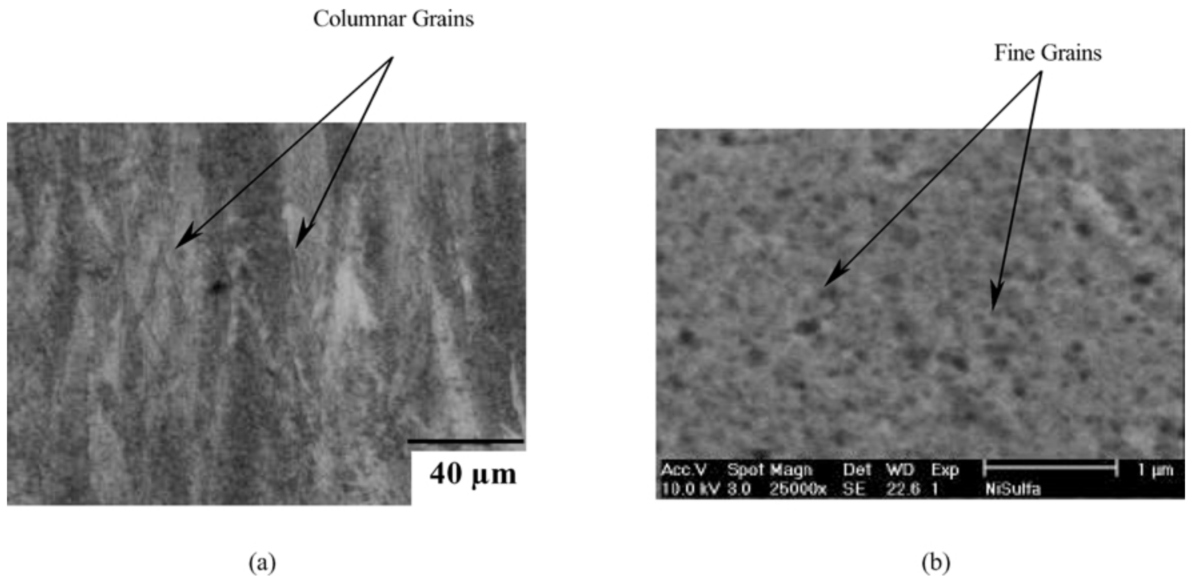
Following Oliver and Pharr's approach [17], the measured load-displacement curves were analyzed according to the Equation:

$$S = \frac{\mathrm{d}P}{\mathrm{d}h} = \frac{2}{\sqrt{\pi}} E_\mathrm{r}\sqrt{A} \qquad (1)$$

where S is the experimentally measured stiffness of the upper portion of the unloading data, A is the projected area of the elastic contact, E_r is the reduced modulus, which is defined for effectively accounting

TABLE I Composition and operating conditions of nickel sulfamate plating baths

$Ni((NH_2SO_3)_2 \cdot 4H_2O$	440.1 g/l
Boric acid	48 g/l
Wetting agent	0.2%/vol
Temperature	50°C
PH	3.8–4.0



(a) (b)

Figure 1 Cross-sectional optical micrograph and surface SEM images of as-received IGA Ni sample: (a) Optical image and (b) SEM image.

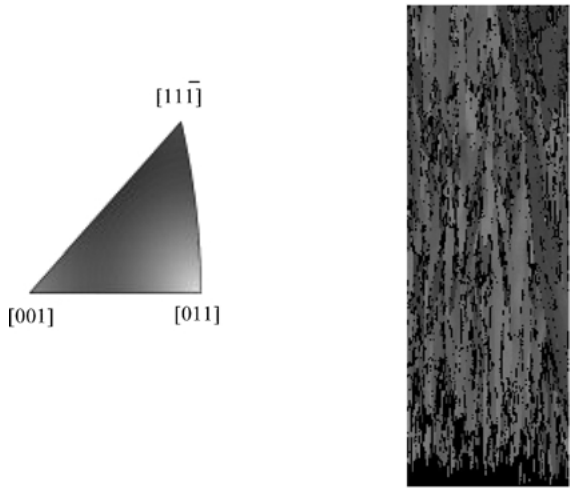


Figure 2 Micro-texture of the as-plated LIGA Ni obtained from sulfamate bath.

non-rigid indenters on the load-displacement behavior as following:

$$\frac{1}{E_r} = \frac{(1-\nu^2)}{E} + \frac{\left(1-\nu_i^2\right)}{E_i} \tag{2}$$

E and ν are Young's modulus and Poisson's ratio for the specimen and E_i and ν_i are the same parameters for the indenter. Furthermore, rearranging Equation 1 gives:

$$E_r = \frac{\sqrt{\pi}}{2}\frac{S}{\sqrt{A}} \tag{3}$$

So if the contact area at peak load is known, the reduced modulus can be calculated from the measured stiffness.

Then the hardness of the material is defined in the usual way [5] to be the mean pressure exerted by the indenter at maximum load. This gives:

$$H = \frac{P_{max}}{A} \tag{4}$$

where P_{max} is the maximum load applied during the identation and A is, again the projected contact area.

4. Results and discussions

4.1. Effects of the indent size and indenter tip on hardness

In the case of the as received LIGA Ni sample that was characterized using the Berkovich indenter, no apparent indent size-dependence was observed (Fig. 3). The hardness values at all the peak loads were around 4.2 GPa, which is close to the Vickers hardness results (∼4.8 GPa) reported in the literature for polycrystalline nickel [18]. The possible reason could be that the radius of tip is around 150 nm, which is too dull to impose any plasticity strain gradient in this very fine poly-crystalline material.

However, the hardness results obtained from the North Star indenter (tip radius is around 30 nm) exhibited the size dependence effect very clearly. A dramatic increase of hardness values was observed with decreasing residual indent depth. This is shown clearly in Fig. 4. Since the indenter tip is very sharp, using a smaller peak load to obtain a detectable residual depth is possible. This makes it possible to explore the effects of very small indent residual depth (as low as tens of nanometers). The measured hardness values are between 3.5 GPa (at larger indent residual depth of ∼400 nm) and 12.5 GPa (for the smallest indent residual depth of ∼18 nm), almost triple the initial value. Compared with Fig. 3, the effect of the residual indent depth on hardness is very clear in the case of the sharper North Star (cube corner) tip.

4.2. Strain gradient plasticity length scale parameters

In order to fully characterize the size dependence of hardness observed in nano-indentation tests, a modeling approach, based on the geometrically necessary dislocations, which are associated with the strain gradient phenomena, was pursued first by Stelmashenko *et al.* [19] and De Guzman *et al.* [20]. The model was subsequently refined by Nix and Gao [9]. In these models, it is assumed that the indentations made by rigid cones are accommodated by circular loops of geometrically necessary dislocations, as shown schematically in Fig. 5. Also, the statistically stored dislocations that

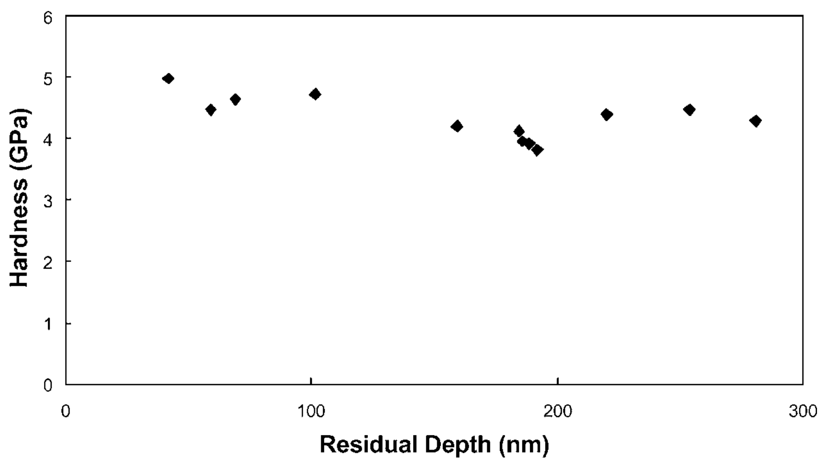


Figure 3 The hardness as a function of residual depth for as-received LIGA Ni sample using berkovich indenter.

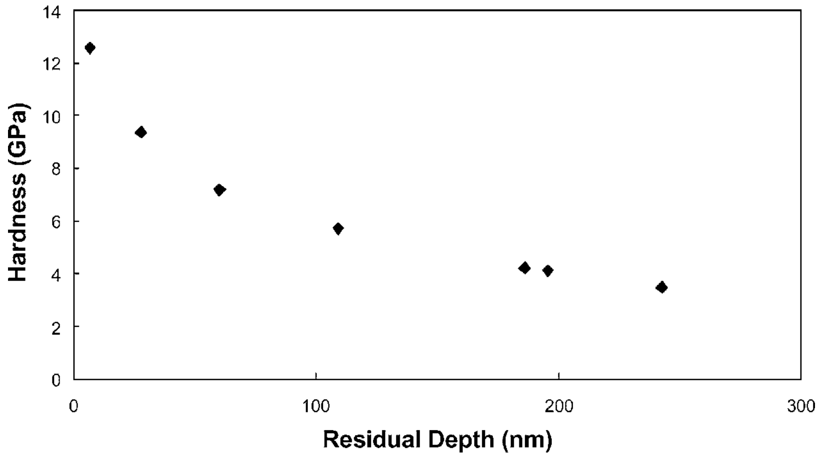


Figure 4 The hardness as a function of residual depth for as-received LIGA Ni sample using north star cube corner indenter.

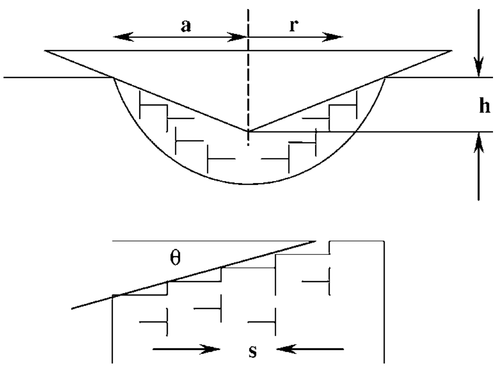


Figure 5 Schematic illustration of the geometrically necessary dislocations created by a rigid conical indentation (taken from Nix and Gao [9]).

are mainly associated with deviatoric strains make contributions to the overall deformation. Assuming that the spacing between the dislocation loops is equal, then we may write [9]:

$$\tan\theta = \frac{h}{a} = \frac{b}{s} \tag{5}$$

where θ is the angle between the surface of the indenter and the plane of sample surface, h is the depth of indentation, a is the contact radius, s is the spacing between individual slip steps, and b is the Burgers vector. Integrating through the radius of indenter, the total length of the injected dislocation loops is given by:

$$\lambda = \int_0^a 2\pi r \frac{\mathrm{d}r}{s} = \int_0^a \frac{h}{ba} 2\pi r \mathrm{d}r = \frac{\pi h a}{b} \tag{6}$$

Then by assuming that all the dislocation loops are confined within the hemispherical volume determined by the contact radius, the density of the geometrically necessary dislocations, ρ_{G}, can be expressed as:

$$\rho_{\mathrm{G}} = \frac{\lambda}{V} = \left(\frac{\pi h a}{b}\right)\left(\frac{3}{2\pi a^3}\right) = \frac{3}{2bh}\left(\frac{h^2}{a^2}\right) = \frac{3}{2bh}\tan^2\theta \tag{7}$$

The shear flow stress, τ, is given by the Taylor's relation to be:

$$\tau = \alpha\mu b\sqrt{\rho_{\mathrm{G}} + \rho_{\mathrm{s}}} \tag{8}$$

where α is a constant to be taken as ∼1/3 for f.c.c. metals, μ is the shear modulus and ρ_{s} is the density of the statistically stored dislocations.

If the von Mises yielding condition of $\sigma = \sqrt{3}\tau$ is applied, we can substitute Equation 7 into Equation 8 to obtain the following expressions for the flow stress:

$$\sigma = \sqrt{3}\alpha\mu b\sqrt{\rho_{\mathrm{s}}}\sqrt{1 + \frac{\rho_{\mathrm{G}}}{\rho_{\mathrm{s}}}} = \sqrt{3}\alpha\mu b\sqrt{\rho_{\mathrm{s}}}\sqrt{1 + \frac{3\tan^2\theta}{2bh\rho_{\mathrm{s}}}} \tag{9a}$$

From the Tabor relation, $H = 3\sigma$. Hence, we may rewrite Equation 9a as:

$$H = 3\sigma = 3\sqrt{3}\alpha\mu b\sqrt{\rho_{\mathrm{s}}}\sqrt{1 + \frac{3\tan^2\theta}{2bh\rho_{\mathrm{s}}}} \tag{9}$$

Reorganizing Equation 9, a simple relationship between hardness and the indent depth can be defined as:

$$\frac{H}{H_0} = \sqrt{1 + \frac{h^*}{h}} \tag{10}$$

where

$$H_0 = 3\sqrt{3}\alpha\mu b\sqrt{\rho_{\mathrm{s}}} \tag{11}$$

and

$$h^* = \frac{81}{2} b\alpha^2 \tan^2\theta \left(\frac{\mu}{H_0}\right)^2 \tag{12}$$

The average strain gradient may be defined as:

$$\chi = \frac{\tan\theta}{a} \tag{13}$$

Equation 10 can now be rewritten as:

$$\left(\frac{\sigma}{\sigma_0}\right)^2 = 1 + \frac{1}{2}b\left(\frac{\mu}{\sigma_0}\right)^2 \chi \tag{14}$$

where σ is the effective flow stress in the presence of strain gradient, and σ_0 is the flow stress in the absence of strain gradient.

As suggested by Nix and Gao [9], the term before the strain gradient can be identified as the microstructural length scale parameter, $\hat{l}$, as shown below:

$$\hat{l} = \frac{1}{2}b\left(\frac{\mu}{\sigma_0}\right)^2 \tag{15}$$

Furthermore, it can be shown that, if the density of the statistically stored dislocations is defined as:

$$\rho_s \approx \frac{1}{L_s^2} \tag{16}$$

where L_s is the mean spacing between statistically stored dislocations, then this microstructural length scale parameter, $\hat{l}$, depends on the mean spacing between dislocations and the Burgers vector, as shown below:

$$\hat{l} = \frac{3}{2}\frac{L_s^2}{b} \tag{17}$$

Following the approach described above, the hardness data obtained by indenting with the North Star cube corner tip was analyzed. This data exhibited a strong size dependence for the as-received LIGA Ni film (Fig. 4). As suggested by Equation 10, the square of the hardness can be plotted against the reciprocal of the depth of indentation. A very good linear relationship was found for the data. This linear fit gives estimates of H_0 and h^* as 2.6 GPa and 343.3 nm, respectively. Also, the data can be displayed as a plot of $(H/H_0)^2$ vs. $1/h$, as shown in Fig. 6. From Equation 12, a self-consistency check can be done by computing the expected value of h^* from the measured H_0. For the North Star cube corner tip used in the study, the area function can be described as:

TABLE II Comparison of predicted and measured values of the characteristic length, h^*

Indenter tip	H_0 (GPa)	h^* (nm)	μ (GPa)	b (nm)	α	h^* (predicted) (nm)
Cube corner tip	2.60	343.3	73	0.25	0.33	350.4

$$A = 2.598h^2 = \pi a^2 \tag{18}$$

Then from Equation 1, $\tan\theta$ can be calculated as following:

$$\tan\theta = \frac{h}{a} = \sqrt{\frac{\pi}{2.598}} = 1.10 \tag{19}$$

The data used in the calculation and the results are presented in Table II. The predicted value of h^* of 350.4 nm is in very good agreement with the measured value of h of 343.3 nm. This suggests that the mechanism-based strain gradient plasticity model can be extended to nano-scale regime, for very fine grained poly-crystalline material, in the presence of a sharp enough indenter tip, i.e., significant strain gradients.

The microstructural length scale parameter, $\hat{l}$, can be computed with Equation 15. Using measured value of H_0, and taking the Tabor factor as 3, b as 0.25 nm for the f.c.c. nickel structure, and the shear modulus as 73 GPa from a previous study of LIGA Ni films [21]. We obtain $\hat{l} = 0.89\ \mu$m.

4.3. Comments and implications

Before concluding, it is important to note that some special cautions were taken in the experiments to ensure that the results provided true indications of size effects. First, to account for the load frame compliance, the contact compliance was obtained by subtracting the machine compliance from the total measured

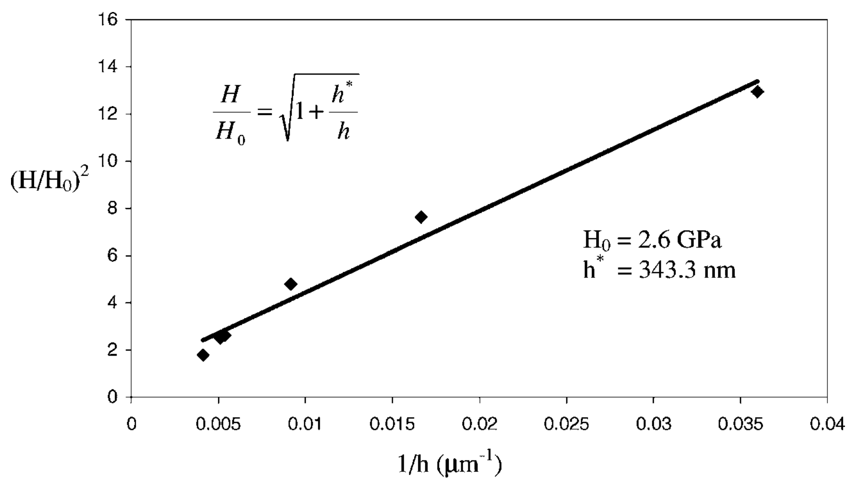


Figure 6 Depth dependence of the hardness of the as-plated LIGA Ni from sulfamate bath.

compliance of the testing system. Also, the tests were done in a quiet room with temperature maintained at a constant level of 20°C. Furthermore, an enclosure was used to minimize the airflow current effect. Relatively short holding periods were chosen in order to reduce the effects of thermal drift in our small load tests. Furthermore, a sharp cube corner indenter tip, with a tip radius of ∼30 nm, was used in the present study. This ensured that the shape of indenter remained self-similar, even at small indentation depths. It also minimized the uncertainty in the projected contact area at small depths of indent. Excellent self-consistent agreement was observed in the prediction by using above approach (Table II).

As suggested by Nix and Gao [9], the extracted microstructural length scale parameter $\hat{l}$ is actually related to the material length scale parameter, l, defined by Fleck and Hutchinson [11]. The relationship is given by the following Equation 9:

$$l \approx \hat{l}\left(\frac{\sigma_0}{\sigma_{\text{ref}}}\right)^2 \tag{20}$$

where σ_{ref} is taken to be a measure of the yield stress and σ_0 is the flow stress in the absence of strain gradient.

From the derivation presented earlier, we know that σ_0 is the yield stress in absence of strain gradient, i.e., in the case of the primarily dislocation interaction strengthened f.c.c. pure metal, the contributions from statistically stored dislocations. So, it is not unreasonable to take yield stress from a larger sample [15] of 277 MPa for σ_0. Using the yield stress of 437 MPa obtained from a prior study of LIGA nickel MEMS structures [21], the material length scale parameter can be calculated from Equation 20 to be $l \sim 0.36\ \mu$m. This length scale parameter corresponds essentially to the stretch gradient parameter, l_s, which plays a dominant role in the deformed regions that surround the nano-indents (Fig. 5). The computed parameter is on the same order of what Begley and Hutchinson [6] obtained from earlier studies on W, Cu and Ag single crystal, as well as Cu polycrystals. These gave estimates of l to be on the order of ∼0.25–1 μm. Finally, we may now estimate the rotational gradient parameter [14, 22] obtained from prior micro-bend experiments on LIGA Ni foil. This may be extracted from a composite length scale parameter, l_c, obtained from micro-bend experiments proposed originally by Stölken and Evans [22]. This gives:

$$l_c = l_R\sqrt{1 + \frac{8}{5}\left(\frac{l_s}{l_R}\right)^2} \tag{21}$$

where l_c is the composite length scale parameter, l_R is the rotational gradient parameter and l_s is the stretch gradient parameter.

Shrotriya *et al.* [14] have estimated the composite length scale parameter, l_c, for LIGA Ni films obtained from same fabrication process to be ∼4.8 μm. Hence, the rotational gradient parameter of the as-received LIGA nickel MEMS structure examined in this study can be estimated from Equation 21 to be ∼4.78 μm. The measured values of l_R and l_s can now be incorporated into the extended J_2 theory and used for the constitutive modeling of plasticity in LIGA Ni MEMS structures plated under similar conditions to those explored in this work.

5. Conclusions

1. Nano-indentation tests have been performed on as-received LIGA Ni films (plated from sulfamate baths) with 2 different indenter tips, Berkovich and North Star cube corner tips. No apparent size dependence has been found for LIGA Ni films indented with the Berkovich tip. However, a dramatic increase of hardness was observed with decreasing residual indent depth, for films indented with the North Star tip. These are associated with the increased strain gradients that are induced during indentation with the North Star tip.

2. A microstructural length scale parameter, $\hat{l} = 0.89\ \mu$m, has been determined in LIGA nickel films (indented with a sharp North Star tip). This was obtained using a mechanism-based strain gradient plasticity approach based on the concept of geometrically necessary dislocations [9]. The stretch gradient plasticity length scale parameter, l_s, associated with the microstructural length scale parameter, $\hat{l}$, has also been shown to be ∼0.36 μm. This is in the same range ($l_s \sim 0.25$–1 μm) as prior reports by Begley and Hutchinson [16] for single crystal and polycrystalline metals.

3. Using the measured composite gradient, l_c, of ∼4.8 μm obtained from prior work on LIGA Ni films [14], the rotational gradient, l_R, for the as-received LIGA Ni films in current study was determined to be ∼4.78 μm. The measured length scale parameters can now be incorporated into the extended J_2 theory, and used in the constitutive modeling of plasticity in LIGA Ni MEMS structures plated under the similar conditions to those employed in this work.

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