

Appendix A. Calculation of plastic strain amplitude and accumulated plastic strain

When cyclic deformation of GNG/CG Cu specimens is performed under the constant total strain amplitude ($\Delta\epsilon_t/2$) control, the sum of elastic strain amplitude ($\Delta\epsilon_e/2$) and plastic strain amplitude ($\Delta\epsilon_{pl}/2$) is maintained constant along the depth from GNG surface to CG core during cyclic loading:

$$\Delta\epsilon_t/2 = \Delta\epsilon_e/2 + \Delta\epsilon_{pl}/2 \quad (1)$$

In general, $\Delta\epsilon_e/2$ is defined as

$$\Delta\epsilon_e/2 = \sigma_a/E \quad (2)$$

Where σ_a is the stress amplitude, i.e. $\Delta\sigma/2$; while E is the Young's modulus (120 GPa for Cu).

In this study, although GNG/CG Cu specimens were cyclically deformed under the iso-total strain amplitude, the gradient plastic strain distribution in cyclically deformed GNG/CG Cu arises due to progressive yielding from the coarse-grained core to nanometer surface. These two regions exhibit large differences in yield strength and hardness, as shown in Fig. 6a.

For the nano or ultrafine grains in the GNG layer (with limited strain hardening capacity) during cyclic deformation, their yield strength and tensile strength (σ_{TS}) (both are equal) are estimated to be approximately one third of the hardness value. Thus, the σ_a value in the GNG layer at different cycles (namely $N= 1, 4\%, 20\%N_f$) can be estimated, using the measured hardness at the corresponding depth:

$$\sigma_a \approx H_v/3 \quad (3)$$

For the CG core and deformed CG layer with high work hardening capacity and the GNG layer at 40% and 100% N_f cycles with large coarsened grain size, σ_{TS} is much higher than σ_y . Thus, σ_a in these layers is not accurately estimated using the Hv data which should be proportional to σ_{TS} . Instead, σ_a in CG core at different cycles can be taken to be same with that of homogeneous CG at the same $\Delta\varepsilon/2$, as shown in Fig. 2b. Similarly, at any cycle, σ_a in the deformed CG layer and/or the GNG layer at 40% and 100% N_f cycles is approximately proportional to σ_a in CG core. Take σ_a in the deformed CG layer as an example:

$$(\sigma_a)_{\text{Deformed CG}}/(\sigma_a)_{\text{CG core}} \approx (Hv)_{\text{Deformed CG}}/(Hv)_{\text{CG core}} \quad (4)$$

Based on the above Equations (1) to (4), in combination of Hv data in GNG layers and stress amplitude data of homogeneous CG, the variation of the $\Delta\varepsilon_{pl}/2$ as a function of depth for cyclically deformed GNG/CG Cu at different cycles are estimated and plotted in Fig. 6b.

In general, the degree of damage accumulation under cyclic loading is evaluated using the cumulative plastic strain, which is generally estimated as [1].

$$4 \sum_{i=1}^N \frac{\Delta\varepsilon_{pl,i}}{2}$$

Here $\Delta\varepsilon_{pl,i}/2$ is the plastic strain amplitude in the i th cycle and N is the total number of cycles. For GNG/CG Cu sample in this study, $\Delta\varepsilon_{pl}/2$ in different depth from GNG surface to CG core varied with cycles, as shown in Fig. 6b. Approximately, for different layers of GNG/CG, $\Delta\varepsilon_{pl}/2$ in a certain period, such as from 4% N_f to 20% N_f , is supposed to equal with a half of $\Delta\varepsilon_{pl}/2$ at 4% N_f and that at 20% N_f .

For example, the accumulated plastic strain at 20% N_f is calculated as:

$$4 \sum_i^{20\%N_f} \frac{\Delta \varepsilon_{pl}}{2} \approx 4 \left[4\%N_f \frac{\Delta \varepsilon_{pl,0} + \Delta \varepsilon_{pl,4\%N_f}}{4} + (20\%N_f - 4\%N_f) \frac{\Delta \varepsilon_{pl,4\%N_f} + \Delta \varepsilon_{pl,20\%N_f}}{4} \right] \quad (5)$$

Similarly, the cumulative plastic strain in different layers of GNG/CG at different cycles can be estimated by adding the cumulative plastic strains in different time periods (0-4% N_f ; 4% N_f - 20 N_f ; 20% N_f - 40 N_f ; 40% N_f - N_f)..

It is obvious that both $\Delta \varepsilon_{pl}/2$ in GNG layer and in CG core at N_f , estimated either from hardness or from stress amplitude data of homogeneous CG, are comparable, as shown in Fig. 6b. Furthermore, the estimated value of $\Sigma 4\Delta \varepsilon_{pl}/2$ in different layers (especially in CG core) of GNG/CG Cu (Fig. 7) is also comparable to that calculated from its hysteresis loop as well. These results suggest that the estimates of $\Delta \varepsilon_{pl}/2$ and $\Sigma 4\Delta \varepsilon_{pl}/2$ in this study are reasonable and reliable. Recent studies of GNG/CG metals by recourse to finite element model employing crystal plasticity add further insights into strain and stress distribution in the GNG/CG material during tensile deformation [57, 58].

Appendix B. Transition life

It is well accepted that in the high-cycle fatigue (HCF) regime under stress control (elastic deformation), the fatigue lives of a metal at different stress amplitudes are dominated by its strength, based on the Basquin equation[1]. Fatigue limit (generally the maximum stress amplitude at which the specimen exhibits a fatigue life of 10^7 cycles) is traditionally used to assess its high cycle fatigue response. By contrast, in the low-cycle fatigue (LCF) regime under strain control (with significant plastic deformation), the fatigue lives ($2N_f$) at different plastic strain amplitudes ($\Delta\varepsilon_{pl}/2$) are governed by ductility, according to the Coffin-Manson equation, i.e.

$$\Delta\varepsilon_{pl}/2 = \varepsilon'_f (2N_f)^c \quad (6)$$

where ε'_f is the fatigue ductility coefficient, which approximately correlates with the extent of elongation to failure in a tensile test; c is the fatigue ductility exponent which ranges from -0.5 to -0.7 for most metals. Typically, the larger ductility, the longer is the low-cycle fatigue life.

Variations of the elastic, plastic and total strain amplitudes as functions of $2N_f$ are plotted in Supplementary Fig. 1, based on the above equations[1]. In order to isolate the LCF and HCF regimes in Supplementary Fig. 1, the transition life is defined as the number of reversals to failure $(2N_f)_t$, at which the elastic and plastic strain components are equal. From equations (1) and (2), transition life is obtained as:

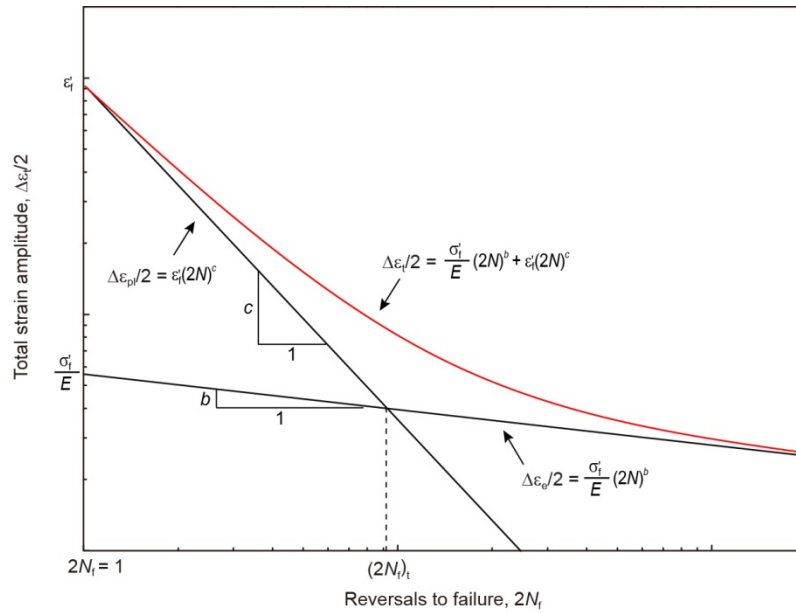
$$(2N_f)_t = \left(\frac{\varepsilon'_f E}{\sigma'_f} \right)^{1/(b-c)} \quad (7)$$

where σ'_f is the fatigue strength coefficient (which approximately equals the true fracture strength) and b is the fatigue strength exponent which ranges from -0.05 to $-$

0.12 for most metals. The LCF regime corresponds to $(2N_f) < (2N_f)_t$ while the HCF corresponds to $(2N_f) > (2N_f)_t$.

The exponent in Eq. (7), $1/(b-c)$, typically ranges from 1.54 to 2.63. From Equation (7), it can be found that the transition life $(2N_f)_t$ is also positively correlated with the fatigue ductility coefficient. Thus, the transition life $(2N_f)_t$ can also be used to assess the resistance to LCF under strain control, similar to the role of fatigue limit played in high-cycle fatigue.

Since the transition life is defined as the number of reversals to failure $(2N_f)_t$, at a “critical” $\Delta\varepsilon/2$ where elastic and plastic strain components are equal, we have performed strain-controlled fatigue tests of GNG metals for different $\Delta\varepsilon/2$ to explore their “critical” $\Delta\varepsilon/2$ and $(2N_f)_t$ experimentally, instead of employing the parameters of Basquin and Coffin-Manson equations. The “critical” total strain amplitudes of GNG/CG, GUF/CG and homogeneous CG Cu are 0.29%, 0.28% and 0.2%, respectively. Note that when comparing low-cycle fatigue properties of GNG/CG and homogeneous CG Cu using transition life, the low-cycle fatigue property of GNG/CG Cu is somewhat underestimated, because its $(2N_f)_t$ is acquired at higher $\Delta\varepsilon_{pl}/2$, due to its high strength and higher elastic strain component, as seen in Fig. 2b.



Supplementary Fig. 1. The variation of the elastic, plastic and total strain amplitude as functions of the number of load reversals to failure ($2N_f$) (from Reference [1]).

References:

[57] Z. Zeng, X.Y. Li, D.S. Xu, L. Lu, H.J. Gao, T. Zhu, Gradient plasticity in gradient nano-grained metals, *Extreme Mech.Lett.* 8 (2015) 213-219.

[58] Y. Wang, G.X. Yang, W.J. Wang, X. Wang, Q. Li, Y.J. Wei, Optimal stress and deformation partition in gradient materials for better strength and tensile ductility: A numerical investigation, *Sci. Rep.* 7 (2017) 10954.