Mechanical properties for irradiated face-centred cubic nanocrystalline metals

X. Z. Xiao$^{1,2}$, D. K. Song$^1$, H. J. Chu$^{4,5}$, J. M. Xue$^{2,3}$ and H. L. Duan$^{1,2}$

$^1$State Key Laboratory for Turbulence and Complex System, Department of Mechanics and Engineering Science, College of Engineering, $^2$CAPT, HEDPS and IFSA Collaborative Innovation Center of MoE, and $^3$State Key Laboratory of Nuclear Physics and Technology, School of Physics, Peking University, Beijing 100871, People’s Republic of China
$^4$Department of Mechanics and Shanghai Institute of Applied Mathematics and Mechanics, Shanghai University, Shanghai 200444, People’s Republic of China
$^5$State Key Laboratory for Mechanical Behavior of Materials, Xi’an 710049, People’s Republic of China

In this paper, a self-consistent plasticity theory is proposed to model the mechanical behaviours of irradiated face-centred cubic nanocrystalline metals. At the grain level, a tensorial crystal model with both irradiation and grain size effects is applied for the grain interior (GI), whereas both grain boundary (GB) sliding with irradiation effect and GB diffusion are considered in modelling the behaviours of GBs. The elastic-viscoplastic self-consistent method with considering grain size distribution is developed to transit the microscopic behaviour of individual grains to the macroscopic properties of nanocrystals (NCs). The proposed theory is applied to model the mechanical properties of irradiated NC copper, and the feasibility and efficiency have been validated by comparing with experimental data. Numerical results show that: (i) irradiation-induced defects can lead to irradiation hardening in the GIs, but the hardening effect decreases with the grain size due to the increasing absorption of defects by GBs. Meanwhile, the absorbed defects would make the GBs softer than the unirradiated case. (ii) There exists a critical grain size for irradiated NC metals, which separates the grain size into the irradiation...
hardening dominant region (above the critical size) and irradiation softening dominant region (below the critical size). (iii) The distribution of grain size has a significant influence on the mechanical behaviours of both irradiated and unirradiated NCs. The proposed model can offer a valid theoretical foundation to study the irradiation effect on NC materials.

1. Introduction

Designing materials that can withstand severe irradiation environments is a great challenge for next generation nuclear reactors. High-energy particle impact can remove atoms from their original lattice sites during the collision cascade and result in the formation of irradiation-induced defects, which include interstitials, vacancies, defect loops and stacking fault tetrahedrons (SFTs), etc. These immovable defects can lead to irradiation hardening and embrittlement, which are the main reasons of material failure [1–3]. It has been well known that grain boundaries (GBs) are significant sinks for irradiation-induced defects [4–7], and nanocrystal (NC) metals with a comparatively large volume of GBs are thought to be more irradiation resistant than conventional polycrystalline metals [8,9]. Therefore, the study of the macroscopic behaviours of NC metals in severe irradiation environments has been a topic of increasing research interest. Some prominent reviews can be found in the papers by Wurster & Pippan [10], Andrievski et al. [11] and Beyerlein et al. [12].

Recently, it has been indicated through several irradiation experiments of NC metals that irradiation-induced defects can be largely affected by GBs. For example, Rose et al. [13] examined the evolution of microstructural defects in NC zirconia and palladium with heavy ion irradiation, and found that the defect density decreases as the grain size gets small, and no defects could be detected when the grain size reduces to approximately 30 nm. Nita et al. [14,15] investigated the proton and ion irradiation impact on NC copper and nickel, and pointed out that the irradiation-induced defects in the grains are mainly SFTs. However, the defect density appears much lower than that of the corresponding coarse grain (CG) counterpart, which indicates good irradiation resistance for NC materials. Sharma et al. [16,17] studied the effect of proton irradiation on the mechanical behaviours and microstructures of NC nickel, and found that GBs play significant roles in the formation and distribution of irradiation-induced defects in NC metals. Matsuoka et al. [18] performed neutron irradiation on ultra-fine grain (UFG) SUS316L steel, and defect-free zones have been observed near GBs, which suggests that GBs can act as effective sinks for irradiation-induced defects. Through neutron irradiation experiments on UFG low-carbon steel, Alsabbagh et al. [19] found that because of the large fraction of GBs, the irradiation effect on both strength and ductility of UFG steel can be reduced, which indicates better irradiation tolerance than the CG counterpart. It can be concluded from these experiments that GBs can act as available sinks for irradiation-induced defects, which effectively reduce the irradiation effect on NC metals. However, available experimental data about the irradiation effect on the mechanical behaviours of NC metals are still quite limited [11] and the corresponding atomistic mechanisms about the effect of GBs on irradiation-induced defects are far from understood [6].

To figure out the underlying microscopic mechanisms of how GBs affect the production of irradiation-induced defects, first-principle calculation and molecular dynamics (MD) have been applied [20,21]. For instance, Liu et al. [21] calculated the formation energies and diffusion barriers near GB in copper by first principle to study the interaction between defects and GBs, and found that interstitials are more active than vacancies to diffuse to GBs. The first MD simulation of displacement damage cascade formation near GBs was performed by Sugio et al. [22] for face-centred cubic (FCC) Ag and it was concluded that GBs preferentially absorb interstitials over vacancies during the defect production stage. The GB-defect interaction in NC copper was studied [6] and it was found that GBs can act as not only the sinks for irradiation-induced interstitials but also the source to emit interstitials to annihilate vacancies in the GI. Samaras et al. [23] performed MD simulations of displacement cascades in NC nickel, and indicated that GBs may absorb
more interstitials than vacancies, which leads to a vacancy dominant defect structure after the
displacement cascades. Borovikhov et al. [24] studied the influence of interstitials or vacancies
on the GB sliding process in tungsten, and found that the introduced defects can make the
average sliding-friction resistance decrease by more than an order of magnitude. Although MD
simulations have offered detailed atomic descriptions about the interaction between GBs and
defects, which depends on both material properties and atomic structures, there are still some
fundamental mechanisms that remain unknown [25], e.g. the effect of absorbed defects on the
mechanical behaviours of GBs.

Besides experiments and computational simulations, theoretical modelling has always been an
important way to study the irradiation effect on metallic behaviours [26–31]. For instance, Krishna
et al. [27] formulated a continuum crystal plasticity model to account for defect annihilation
for polycrystalline copper under neutron irradiation, and can capture the inhomogeneous
plasticity deformation. Patra et al. [29] presented a micromechanics-based model to simulate
the plasticity behaviours of irradiated body-centred cubic (BCC) metals, and the evolutions
of both immobile, mobile dislocations and irradiation-induced defects were all involved. To
simulate the plastic flow localization in defect-free channels, Xiao et al. [31] proposed a tensorial
plasticity model for irradiated FCC metals, and the evolutions of both immobile, mobile dislocations and irradiation-induced SFTs could be effectively considered. It should be noted that
all these theoretical models mentioned above are proposed for irradiated polycrystalline metals.
However, to the author’s knowledge, a constitutive framework to incorporate the irradiation
effect in NC metals has not been reported in the literature.

To study the irradiation effect on mechanical behaviours of NC materials, two major questions
should be considered. Firstly, how to model the deformation behaviour of individual grains,
which includes the properties of GIs and GBs with irradiation effect? Regarding the plastic
deformation inside GIs, partial or full dislocations are expected to emit from the GBs and
consequently slide on their corresponding slip systems. Meanwhile, the remanent irradiation-
induced defects acting as immovable obstacles will impede these dislocations gliding. Therefore,
the spatial dependent dislocation–defect interaction with GB effect should be considered in
modelling the properties of GIs. Regarding the plastic deformation of GBs, there are many
theoretical models to capture the different deformations. Both GB diffusion [32,33] and GB sliding
[34–37] become the dominant mechanisms when the grain size decreases to tens of nanometers.
On the one hand, GBs can act as fast diffusion paths for atom migration, which corresponds the
creep in NC materials [38]. On the other hand, GBs are usually viewed as amorphous materials
and the study of GB sliding can be based on the amorphous metallic theory [39,40], in which
the absorption of irradiation-induced defects by GBs would like to make the average sliding-
friction resistance of GBs decrease as indicated by MD simulations [24]. Therefore, the influence
of irradiation on GIs and GBs should be considered separately at the grain level. Secondly, how
to model the macroscopic behaviours of NC materials? The mixture rules are usually used as the
scale transition from the grain level to the macroscopic level [41]. Although simple expressions
can be obtained from the mixture methods, stress concentrations between individual grains can
hardly be predicted because the strain/stress fields are treated to be homogeneous. Considering
the temporal and spatial coupling among different grains, an elasto-viscoplastic self-consistent
(EVPSC) theory has been proposed recently [42,43], and was developed to take into account the
imperfect interfaces [44] and the strain-rate sensitive behaviours [45] of NC materials. However,
in this EVPSC method, the volume fraction of each individual grain is assumed to be the same
without considering the grain size distribution. While, it should be noted that as the size effect
becomes important at nanoscale, the grain size distribution of NC materials may have a significant
influence on the macroscopic mechanical behaviours [46,47]. Therefore, in this work, a modified
EVPSC method considering grain size distribution for different volume fractions of individual
grains will be proposed for the scale transition.

In summary, both experiments and computational simulations have indicated that the
irradiation effect on NC metals can be largely different from their CG counterpart, which mainly
originate from the absorption of irradiation-induced defects by GBs. However, there is a lack
of corresponding theoretical studies for the mechanical behaviours of irradiated NC metals. Therefore, it would be necessary to build a theoretical model to analyse the irradiation effect on the mechanical behaviours of NC metals.

The purpose of this work is to develop a self-consistent theory to model the mechanical behaviours of irradiated FCC NC metals, which includes (i) reasonable depiction and formulation of the mechanical behaviours of both GIs and GBs with irradiation effect at the grain level, and (ii) an appropriate scale transition from the grain level to the macroscopic level to obtain the mechanical properties of irradiated NC metals. This paper is organized as follows. In §2, we propose the constitutive relation for the GI with irradiation effect. In §3, the GB diffusion and GB sliding with irradiation effect are considered for GBs. In §4, a modified EVPSC with grain size distribution is developed for the scale transition. Applications of the model in irradiated NC copper are performed in §5. Finally, we close with some conclusions in §6.

2. Constitutive model of grain interiors

For irradiated NC metals as shown in figure 1a, two major mechanisms will dominate the inelastic deformation of GIs, which include the spatial-dependent interaction between dislocations and irradiation-induced defects, as well as the emission of full or partial dislocations from GBs. Recently, Xiao et al. [31] have proposed a tensorial crystal model to consider the spatial-dependent dislocation-defect interaction for irradiated FCC single crystals. Although the dislocation-defect interaction mechanism seems similar, it should be noted that (i) the defect density in GIs of NC is greatly affected by GBs, which is different from the free surface of single crystals; (ii) it would be preferable for dislocations to be emitted from GBs rather than nucleated inside the GIs when the grain size decreases to a few nanometers.

The continuum-mechanical behaviours for irradiated GIs of FCC NC metals are modelled under the standard rate-dependent crystal plasticity framework [48,49]. The plastic deformation takes place through the slipping of dislocations on the corresponding slip system \( \alpha \), which is defined by the normal direction \( n^\alpha \) of the slip plane and the slip direction \( s^\alpha \). The rate of the plastic deformation gradient of the \( i \)th GI can be expressed as

\[
\dot{F}^p_i = \left( \sum_{\alpha=1}^{N_\alpha} \gamma^{1,\alpha}_i s^\alpha \otimes n^\alpha \right) \cdot \dot{F}^p_i
\]  

\[(2.1)\]
where \( \dot{\gamma}_i^{L,\alpha} \) is the shearing slip rate on slip system \( \alpha \) of the GI. \( N_s \) is the number of slip systems, and there are 12 slip systems characterized by the Miller indices \((111)\) for FCC materials.

In this paper, the capitals 'I' and 'B' in superscript or subscript are used to distinguish the related variables corresponding to the GI and GB, respectively. The shearing slip rate \( \dot{\gamma}_i^{L,\alpha} \) can be expressed as [50]

\[
\dot{\gamma}_i^{L,\alpha} = \dot{\gamma}_0^I \left( \frac{|\tau^{L,\alpha}|}{\tau^{L,\alpha}_{c,i}} \right)^{1/m_I} \text{sign}(\tau^{L,\alpha}), \tag{2.2}
\]

where \( \dot{\gamma}_0^I \) and \( m_I \) are the reference shearing rate and the strain rate sensitivity for the GI, respectively. \( \tau^{L,\alpha} \) is the resolved shear stress (RSS) and \( \tau^{L,\alpha}_{c,i} \) is the critical resolved shear stress (CRSS). RSS is related to \( \tau^{L,\alpha} = T^{(1)} : R^{L,\alpha} \), where \( R^{L,\alpha} \) is the Schmid tensor defined as \( R^{L,\alpha} = \frac{1}{2} (s^\alpha \otimes n^\alpha + n^\alpha \otimes s^\alpha) \) for the GI, and \( T^{(1)} \) is the second Piola–Kirchhoff stress. The viscoplastic strain rate tensor for the GI is given as

\[
\dot{\varepsilon}_I^{vp} = \sum_{\alpha=1}^{N_s} \dot{\gamma}_i^{L,\alpha} R^{L,\alpha}. \tag{2.3}
\]

For irradiated NC metals, both dislocations emitted from GBs and the spatial-dependent interaction between dislocations and defects inside the GI would play important roles in determining the behaviours of the GI. Thus, the CRSS of the GI should involve the dislocation-defect interaction within GIs just vanishes. For irradiated FCC metals, SFTs are the main irradiation-induced defects at a low irradiation dose (dpa < 0.1) [16]. \( \beta \) gives the defect habit plane and there are four different habit planes for SFTs, i.e. \( N_d = 4 \) for SFTs on \((111)\) habit planes.

(a) Irradiation-induced defect hardening

Under irradiation, numerous irradiation-induced defects may be formed in the GI, and the defect density would decrease with the increasing proportions of GBs [14,15], which makes the irradiation effect grain-size dependent. To reveal the spatial-dependent dislocation–defect interaction with GB effect, a grain size-dependent tensorial plasticity model [31] is considered for \( \tau_{\text{def},i}^{\alpha} \), i.e.

\[
\tau_{\text{def},i}^{L,\alpha} = \tau_{\text{dis},i}^{L,\alpha} + \tau_{\text{dis},i}^{L,\alpha} + \tau_0^I, \tag{2.4}
\]

It should be noted that for FCC metals, \( \tau_0^I \) is always quite small and often neglected in the numerical analysis [31]. In the following, details of the first two terms (\( \tau_{\text{def},i}^{L,\alpha} \) and \( \tau_{\text{dis},i}^{L,\alpha} \)) of CRSS in equation (2.4) will be presented.

\[
\tau_{\text{def},i}^{L,\alpha} = \frac{b \mu}{h_d(d_i)} \sum_{\beta=1}^{N_d} N_{\alpha}^\beta : H^\beta, \tag{2.5}
\]

where \( b \) and \( \mu \) denote the magnitude of the Burgers vector and the shear modulus, respectively. \( h_d(d_i) = h_{d_0}((d_i - 2w_s)/d_i)^2 \) is the dislocation–defect hardening coefficient, which is dependent on the grain size \( d_i \) and the thickness \( w_s \) of the shell region where it is free of defects near the GBs as shown in figure 1b. \( h_{d_0} \) is the dislocation–defect hardening coefficient without the influence of GBs and \( \xi \) is a geometrical parameter. When \( d_i < 2w_s \), no defects will exist in the GI, and the dislocation–defect interaction within GIs just vanishes. For irradiated FCC metals, SFTs are the main irradiation-induced defects at a low irradiation dose (dpa < 0.1) [16]. \( \beta \) gives the defect habit plane and there are four different habit planes for SFTs, i.e. \( N_d = 4 \) for SFTs on \((111)\) habit planes.

The dislocation and defect descriptor tensors \( N^\alpha \) and \( H^\beta \) are defined as

\[
N^\alpha = n^\alpha \otimes n^\alpha \tag{2.6}
\]

and

\[
H^\beta = N_{\text{def}} N_{\text{def}} (I^{(2)} - n^\beta \otimes n^\beta + P_{\text{ann}}^\alpha \delta_\alpha^\beta n^\beta \otimes n^\beta), \tag{2.7}
\]
where $I^{(2)}$ is the second-order identity tensor. $N_{\text{def}}$ and $d_{\text{def}}$ are the volume density and size of the SFT, respectively. $\delta_{\alpha}^\beta$ is the Kronecker delta and $P_{\text{ann}}^\alpha$ is the annihilation probability proposed by Krishna et al. [27], i.e.

$$P_{\text{ann}}^\alpha = A_{\alpha}^{\text{ann}} \rho_{\alpha}^n,$$  

(2.8)

where $\rho_{\alpha}^n$ is the network dislocation density and $A_{\alpha}^{\text{ann}}$ is the annihilation area of defects given by $A_{\alpha}^{\text{ann}} = 2d_s S_{\alpha} + \pi d_s^2$. $d_s$ represents the stand-off distance, while $S_{\alpha}$ is the moving distance of the dislocation before it meets a forest of dislocations.

The evolution of defect density in the GIs can be governed by [30]

$$\dot{H}_\beta = -\eta \sum_{\alpha=1}^{N_s} (N_{\alpha} : H_{\beta}) N_{\alpha} |\dot{\gamma}_{I_{\alpha}}|,$$  

(2.9)

where $\eta$ is the annihilation efficiency. According to equation (2.9), the evolution of the defect density on habit plane $\beta$ is induced by the interaction between the defects on plane $\beta$ and dislocations on all slip systems.

(b) Slip resistance to dislocation emission

For NC metals, dislocations (full or partial) emitted from GBs will play an important role in the plastic deformation when the grain size gets down to tens of nanometers. Actually, there exists a critical grain size $d_p = 2\mu b b_p / 3\gamma_{sf}$, where $b_p$ is the magnitude of the Burgers vector of partial dislocations and $\gamma_{sf}$ is the stacking fault energy [51]. Above $d_p$, full dislocations will move along \{111\}\langle110\rangle slip systems; below $d_p$, partial dislocations will move along \{111\}\langle11\bar{2}\rangle faulting systems for FCC metals [52]. When full dislocations are emitted from GBs, the slip resistance $\tau_{\text{dis},I_{\alpha}}$ can be expressed as [34,35]

$$\tau_{\text{dis},I_{\alpha}} = \frac{\mu b}{d_i}. $$  

(2.10)

When the grain size $d_i$ decreases below $d_p$, the slip resistance will be dominated by partial dislocations [34,35], i.e.

$$\tau_{\text{dis},I_{\alpha}} = \frac{\mu b}{3d_i}. $$  

(2.11)

It can be seen from equations (2.4), (2.10) and (2.11) that both $\tau_{\text{def},I_{\alpha}}$ and $\tau_{\text{dis},I_{\alpha}}$ are grain size dependent. Different from the coarse grains in conventional polycrystals, the grains in NCs have little space for dislocation tangle. Therefore, the dislocation network hardening effect can be neglected.

3. Constitutive model of grain boundaries

For NC materials, there exists a high volume fraction of GBs in the inter-crystalline regions, which may greatly affect the mechanical behaviours of NC when the grain size gets small. Typically, there are two major inelastic deformation mechanisms that will determine the behaviours of GBs, i.e. GB sliding [34,35] and GB diffusion [32]. Therefore, the total inelastic strain rate of GBs can be expressed as

$$\dot{\epsilon}_{B_{\text{VP}}} = \dot{\epsilon}_{\text{BS}_{\text{VP}}} + \dot{\epsilon}_{\text{BC}_{\text{VP}}}, $$  

(3.1)

where $\dot{\epsilon}_{\text{BS}_{\text{VP}}}$ and $\dot{\epsilon}_{\text{BC}_{\text{VP}}}$ denote the inelastic strain rate of GB slip and GB creep due to GB diffusion, respectively. In the following, the detailed description of the GB behaviours with irradiation effect will be given below.

(a) Grain boundary sliding

It is well known that there are no long-range ordered crystal structures for GBs, which is similar to the amorphous isotropic material, e.g. metallic glass. Therefore, based on the amorphous theory of
Anand & Su [40], the constitutive relations for GB sliding with irradiation effect are considered as follows. For amorphous isotropic metals, it has been indicated that no preferred slip system exists other than the principal directions of stress. Then, the plastic deformation of GBs is assumed to occur through shearing relative to six potential slip systems, which are defined as,

\[
\begin{align*}
\varepsilon^{(1)}_i &= \cos \theta e_1 + \sin \theta e_3, \\
\varepsilon^{(2)}_i &= \cos \theta e_1 - \sin \theta e_3, \\
\varepsilon^{(3)}_i &= \cos \theta e_1 + \cos \theta e_2, \\
\varepsilon^{(4)}_i &= \cos \theta e_1 - \cos \theta e_2, \\
\varepsilon^{(5)}_i &= \sin \theta e_2 - \cos \theta e_3, \\
\varepsilon^{(6)}_i &= \sin \theta e_2 + \cos \theta e_3,
\end{align*}
\]

where \( \theta = \pi/4 + \varphi/2 \), with \( \varphi \) the angle of internal friction, which is defined according to the friction coefficient \( f_s \). For the case without irradiation, \( f_s \) is close to the internal frictional coefficient of metallic glass [39]. While, for the case of irradiation, large numbers of interstitials and vacancies will be absorbed into GBs, and these irradiation-induced defects will make the sliding friction resistance decrease as indicated through MD simulations [24]. Therefore, the internal friction angle can be expressed as,

\[
\varphi = \begin{cases} 
\arctan f_s, & \text{without irradiation} \\
\arctan \left( \frac{f_s}{\kappa} \right), & \text{with irradiation},
\end{cases}
\]

where \( \kappa \) is the decay coefficient of internal friction due to the influence of irradiation. Then, the inelastic strain rate of GB slip takes the form,

\[
\dot{\varepsilon}^{(\text{BS,VP})}_i = \sum_{\alpha=1}^{N_B} \dot{\varepsilon}^{(\alpha)}_i \mathbf{R}^{B,\alpha},
\]

where \( N_B = 6 \) and \( \mathbf{R}^{B,\alpha} \) is the Schmid tensor for GBs defined as \( \mathbf{R}^{B,\alpha} = \frac{1}{2} (s^{\alpha}_0 \otimes n^{\alpha}_0 + n^{\alpha}_0 \otimes s^{\alpha}_0) \).

The plastic shear rate of GBs gives

\[
\dot{\varepsilon}^{B,\alpha}_i = \dot{\gamma}^{B}_0 \left( \frac{\tau^{B,\alpha}}{c_0 + \tan \varphi \cdot \sigma^{B,\alpha}} \right)^{1/n_B},
\]

where \( \dot{\gamma}^{B}_0 \) is a reference shear strain rate, \( c_0 \) is the cohesion and \( n_B \) is the strain rate sensitivity of GBs. \( \tau^{B,\alpha} \) and \( \sigma^{B,\alpha} \) are the RSS and normal compression on each slip system, respectively.

(b) Grain boundary diffusion

The GBs can act as diffusion paths for atom migrations, which is controlled by creep even at low temperatures [32]. The plastic strain rate contribution \( \dot{\varepsilon}^{(\text{BC,VP})}_i \) of GB creep [38] can be expressed as

\[
\dot{\varepsilon}^{(\text{BC,VP})}_i = \frac{150 \Omega_B \delta D_{gb}}{k_B T d_i^3} \exp \left( - \frac{Q_{gb}}{RT} \right) T^{(1)},
\]

where \( \Omega_B \), \( \delta \) and \( D_{gb} \) are the atomic volume, the layer thickness and the GB diffusivity, respectively. \( k_B \) is the Boltzman constant and \( R \) is the ideal gas constant. \( Q_{gb} \) and \( T \) are the activation energy for GB diffusion and the temperature, respectively. The influence of irradiation on GBs’ diffusivity can be studied by considering the change of the activation energy \( Q_{gb} \) and stress \( T^{(1)} \) in equation (3.6). The plastic strain rate contribution \( \dot{\varepsilon}^{(\text{BC,VP})}_i \) would vary with \( T^{(1)} \) under...
irradiation effect. The activation energy for GB diffusion might be changed due to the effect of irradiation-induced defects. However, few available results from experiments and numerical simulations are reported. For brevity, the effect of irradiation on the activation energy is neglected in this work.

In §§2 and 3, the constitutive relations for the GIs and GBs with irradiation effect are proposed at the grain level. In order to obtain the overall behaviours of irradiated NC metals at the macroscopic level, the scale transition method is required and will be given below in details.

4. Elasto-viscoplastic self-consistent method with grain size distribution

As mentioned in the introduction, both grain size and grain size distribution could play important roles in determining the responses of NC metals [47]. In this part, we will present a modified EVPSC method with considering grain size distribution to bridge the gap between the microscopic mechanical behaviour of individual grains and macroscopic mechanical behaviour of NC. First, based on the classical self-consistent theory, a representative element volume (RVE) is chosen as shown in figure 1a, which contains $N$ GIs with different grain sizes and between them are the GBs [53]. The grain size distribution follows the log-normal distribution as represented by Zhu et al. [47], i.e.

$$P(d_i) = \frac{1}{(2\pi)^{1/2}d_i \phi} \exp \left(-\frac{1}{2} \left(\frac{1}{\phi} \ln \frac{d_i}{d_0}\right)^2\right),$$

(4.1)

where $d_0$ and $\phi$ are the parameters describing the median and shape parameters, respectively. $d_0$ and $\phi$ can be obtained by

$$d_0 = \sqrt{\frac{\bar{d}^4}{\hat{\phi} + \bar{d}^2}},$$

(4.2)

and

$$\phi = \sqrt{\ln \left(\frac{\phi}{\bar{d}^2} + 1\right)},$$

(4.3)

where the arithmetic mean size $\bar{d}$ can be measured experimentally, and $\hat{\phi}$ is the variance of $d_i$. Assuming that each grain in the RVE is of the same shape, then the total volume of the RVE is

$$V = \int_0^\infty kd_i^3 P(d_i) \, dd_i,$$

(4.4)

where $k$ is a constant representing the shape of the grains (e.g. $k = \pi/6$ for a sphere). Then, one can have the volume weighted grain size distribution $f_{i,V}$ as

$$f_{i,V} = \frac{kd_i^3}{V} P(d_i).$$

(4.5)

It should be noted that the irradiation dose considered in this work is low (less than 0.1 dpa) and the main defects are SFTs. Therefore, the effects of irradiation on the grain size and its distribution are small and neglected. When the proper external load is applied on the RVE, the macroscopic strain rate tensor $\dot{\mathbf{E}}$ and stress rate tensor $\dot{\mathbf{S}}$ of heterogeneous materials can be obtained according to the classical homogenization progress,

$$\dot{\mathbf{E}} = \frac{1}{V} \int_V \dot{\mathbf{e}}_{i}^{\text{HI}} \, dV = \sum_{i=1}^{N} f_{i,V} \dot{\mathbf{e}}_{i}^{\text{HI}},$$

(4.6)

and

$$\dot{\mathbf{S}} = \frac{1}{V} \int_V \dot{\mathbf{\sigma}}_{i}^{\text{HI}} \, dV = \sum_{i=1}^{N} f_{i,V} \dot{\mathbf{\sigma}}_{i}^{\text{HI}},$$

(4.7)
Figure 2. Schematic of the elastic-viscoplastic self-consistent method with interface effect. (a) A representative individual GI with a coated GB embedded in an infinite homogeneous effective medium, which is subjected to a uniform strain rate $\dot{E}$. (b) A homogenized inclusion embedded in an infinite homogeneous effective medium, subjected to a uniform strain rate $\dot{E}$. (Online version in colour.)

where $\dot{\varepsilon}_i^{HI}$ and $\dot{\varepsilon}_i^{HI}$ denote the rate of the stress tensor $\sigma_i^{HI}$ and the strain tensor $\varepsilon_i^{HI}$ of the $i$th homogeneous inclusion (HI) as shown in figure 2a, respectively. One HI consists of one GI and its coated GB as shown in figure 2a.

Second, the $i$th GI with its coated GB is represented as a two-phase material embedded in an infinite medium as shown in figure 2a and one can have

$$\dot{\varepsilon}_i^{VP} = A_i^{IB,VP} : \dot{\varepsilon}_i^{B,VP},$$

(4.8)

where $A_i^{IB,VP}$ is the viscoplastic concentration linking the viscoplastic strain rate tensor $\dot{\varepsilon}_i^{VP}$ of the GI and the viscoplastic strain rate tensor $\dot{\varepsilon}_i^{B,VP}$ of the GB, i.e.

$$A_i^{IB,VP} = [I^{(4)} - S^E : M_i^B : (B_i^B - B_i^I)]^{-1},$$

(4.9)

where $B_i^B$ and $B_i^I$ are the viscoplastic stiffness tensor of the GB and GI, respectively. $M_i^B$ is the viscoplastic compliance tensor of the GB, $S^E$ is the well-known Eshelby tensor [54] and $I^{(4)}$ is the fourth-order identity tensor.

Then, the viscoplastic stiffness tensor $B_i^{HI}$ of the HI (i.e. the GI with its coated GB) can be obtained by using the Mori–Tanaka homogenization scheme [55]

$$B_i^{HI} = [(1 - f_i^I)B_i^B + f_i^I B_i^I : A_i^{IB,VP}] : [(1 - f_i^I)I^{(4)} + f_i^I I^{(4)} : A_i^{IB,VP}]^{-1},$$

(4.10)

where $f_i^I$ is the volume fraction for the GI. For the case of a sphere,

$$f_i^I = \left( \frac{d_i}{d_i + w_{gb}} \right)^3,$$

(4.11)

here $w_{gb}$ is the width of the GB. Therefore, the viscoplastic strain rate tensor of the $i$th HI is,

$$\dot{\varepsilon}_i^{HI,VP} = f_i^I \dot{\varepsilon}_i^{VP} + (1 - f_i^I) \dot{\varepsilon}_i^{B,VP}.$$

(4.12)

Third, the $i$th HI is embedded in an effective medium representing the equivalent material as shown in figure 2b and the local mechanical behaviours of the $i$th HI can be derived from the
secant elasto-viscoplastic self-consistent scheme [42], i.e.

$$\dot{\varepsilon}_i^{HI} = A_i^{CE} : [\dot{\varepsilon} - \dot{\varepsilon}^{VP} + (I^{(4)} - S^E) : B_i^{vP} + S^E : C_i^{HI} : \dot{\varepsilon}_i^{HI, vp}] \quad (4.13)$$

and

$$\sigma_i^{HI} = C_i^{HI} : A_i^{CE} : [(\dot{\varepsilon} - \dot{\varepsilon}^{VP}) + (S^E - I^{(4)}) : (\dot{\varepsilon}_i^{HI, vp} - B_i^{vP} : \dot{\varepsilon}^{VP})] \quad (4.14)$$

where $\dot{\varepsilon}^{VP}$ is the rate of macroscopic viscoplastic strain tensor and can be expressed as

$$\dot{\varepsilon}_i^{HI, vp} = \sum_{j=1}^{N} f_{i,j}^{VP} B_j^{CE} : \dot{\varepsilon}_j^{HI, vp}, \quad (4.15)$$

where $\dot{\varepsilon}_i^{HI, vp}$ denotes the viscoplastic strain rate tensor of the $i$th HI. The superscript ‘t’ means the transposition of the concentration tensor $B_j^{CE}$, which is defined by

$$B_j^{CE} = C_j^{HI} : A_j^{CE} : S^E. \quad (4.16)$$

$A_i^{CE}$ denotes the elastic strain concentration tensor, and $A_i^{vP}$ is the viscoplastic strain concentration tensor, which are defined as

$$A_i^{CE} = [I^{(4)} + S^E : C_i^{HI} - C^E]^{-1} \quad (4.17)$$

and

$$A_i^{vP} = [I^{(4)} + S^E : M^E : (B_i^{HI} - B^E)]^{-1}, \quad (4.18)$$

where $C^E$ and $B^E$ are the macroscopic effective elastic stiffness tensor corresponding to the elastic compliance tensor $S^E$ and viscoplastic stiffness tensor corresponding to the viscoplastic compliance tensor $M^E$, respectively. $C_i^{HI}$ is the elastic stiffness tensor of the HI [56–58].

5. Mechanical property of irradiated NC copper

To evaluate the feasibility and efficiency of the proposed framework, the theory is applied to model the mechanical properties of irradiated NC copper and compared with experimental results. In order to obtain the macroscopic behaviours of NC copper, 500 grains with random orientations are selected in the RVE and the corresponding calculation results are convergent. The time increment method is applied to obtain the macroscopic strain $\varepsilon$ and stress $\Sigma$. The major calculation progress is given as follows [59]:

1. According to equations (4.12)–(4.15), calculate the value of $\dot{\varepsilon}_i^{HI, vp}(t_n)$, $\dot{\varepsilon}_i^{HI}(t_n)$, $\dot{\varepsilon}_i^{HI}(t_n)$ and $\dot{\varepsilon}^{VP}(t_n)$ at a given external load, and obtain the values at a time step $t_n$ ($n = 1, 2, \ldots$).
2. According to equations (4.6) and (4.7), calculate $E(t_{n+1})$ and $\Sigma(t_{n+1})$ at the next time step, i.e. $E(t_{n+1}) = E(t_n) + \Delta \varepsilon \Delta t$ and $\Sigma(t_{n+1}) = \Sigma(t_n) + \Delta \Sigma \Delta t$.
3. Repeat the calculation steps (1) and (2), the macroscopic stress–strain curve is obtained.

The parameters for the GI are given as follows: $\gamma_0^I = 10^{-4} \text{s}^{-1}$, $m_I = 0.05$, the magnitude of Burgers vector $b = 0.256 \text{nm}$, shear modulus $\mu = 40 \text{GPa}$, $\omega_s = 14 \text{nm}$, $\xi = 3$ and $\eta_{d0} = 1$ [31]. The initial SFT volume density at 0.1 dpa is $N_{def} = 4.5 \times 10^{23} \text{m}^{-3}$ and the size of SFT $d_{def} = 2.5 \text{nm}$ [60], the stand-off distance $d_s = 2.4 \text{nm}$ [27] and $\eta = 25$. The magnitude of Burgers vector of partial dislocations $b_p = 0.148 \text{nm}$ and the stacking fault energy $\gamma_{sf} = 45 \text{mJm}^{-2}$ [35].

The following parameters are used to describe the irradiated behaviours of GBs. The friction coefficient $f_k = 0.06$, which is close to that of metallic glass [39]. The decay coefficient of internal friction $\kappa = 10$ [24] at 0.1 dpa. $c_0 = 550 \text{MPa}$, $\gamma_0^B = 10^{-3} \text{s}^{-1}$ and $m_B = 0.1$. For GB diffusion, the atomic volume $\Omega_a = 1.18 \times 10^{-29} \text{m}^3$, $\delta \cdot D_{gb} = 5 \times 10^{-15} \text{m}^3 \text{s}^{-1}$, the Boltzmann constant $k_B = 1.38 \times 10^{-23} \text{JK}^{-1}$, the gas constant $R = 8.311 \text{JK}^{-1} \text{mol}^{-1}$, the activation energy $Q_{gb} = 104 \text{kJmol}^{-1}$ and $T = 300 \text{K}$ [38]. The width of the GB $w_{gb} = 1 \text{nm}$ [34].
Figure 3. Size effect on the tensile response of a homogenized inclusion (HI) without irradiation effect and the plastic strain rates due to different mechanisms. The load strain rate is $10^{-5}$ s$^{-1}$. (a) Stress–strain curves of the HI regarding different grain sizes; (b) plastic slip rate of the GI regarding different grain sizes; (c) plastic slip rate of the GB regarding different grain sizes; (d) plastic creep rate of the GB regarding different grain sizes. (Online version in colour.)

Individual GIs with their coated GBs, i.e. the HIs, as shown in figure 2a, are the basic units of NC and their deformations will play an important role in the mechanical behaviours of the whole NC. Therefore, we first investigate the size effect on the tensile responses of an individual HI and the relative plastic strain rates of different mechanisms without irradiation effect. The load strain rate is fixed at $10^{-5}$ s$^{-1}$ along [100]. It can be seen that the yield stress of the HI will firstly increase with the decrease of grain size from 100 nm, whereas below about 30 nm, the yield stress will decrease as shown in figure 3a. The reasons are (i) the slip resistance for dislocations emitted from GBs increases when the grain size decreases as shown in equations (2.10) and (2.11). (ii) With the decrease of grain size, the volume ratio of the GBs gradually increases. Therefore, the contribution of the GB plastic deformation on the total deformation becomes important. Consequently, its corresponding deformation mechanisms would play the dominant role. As illustrated in figure 3b–d, the plastic slip rate of GI gradually decreases with the grain size, whereas the plastic slip and creep rates inside the GB increase, which implies that the GB deformation tends to be the dominant deformation mechanism when the grain size decreases to a few nanometers.

The irradiation effect on the tensile behaviours of the HI and its corresponding plastic strain rates of different mechanisms are given in figure 4 for various grain sizes. The load strain rate is fixed at $10^{-5}$ s$^{-1}$ and the irradiation dose dpa = 0.1. As shown in figure 4a, irradiation will lead to the increase of yield stress because of the impediment of slip dislocations by irradiation-induced defects. After the yield point, the immobile irradiation-induced defects will be annihilated by the corresponding dislocation slipping, which results in the decrease of CRSS and the plastic flow localization. However, the irradiation effect gets weak with the decrease of grain size. It
Figure 4. Size effect on the tensile response of a homogenized inclusion (HI) with irradiation effect and the plastic strain rates due to different mechanisms. The load strain rate is $10^{-5}$ s$^{-1}$ and the dpa = 0.1. (a) Stress–strain curves of the HI regarding different grain sizes; (b) plastic slip rate of the GI regarding different grain sizes; (c) plastic slip rate of the GB regarding different grain sizes; (d) plastic creep rate of the GB regarding different grain sizes. (Online version in colour.)

is understandable that the increasing proportion of GBs can absorb irradiation-induced defects inside the GI, which results in the decrease of defect density in GIs and its related irradiation hardening effect. Comparing figure 4b–d, it can be seen that (i) when the grain size is above 40 nm, the plastic deformation of the GI is dominant for the deformation of NC, while the contributions of GB slip and creep can be ignored because of the small volume fraction of GBs. (ii) The plastic deformations of GB slip and creep are affected by both irradiation and grain size. It should be noted that the phenomenon of strain softening for irradiated nanocrystals as observed in figure 4a is related to the annihilation of defects due to the dislocation–defect interaction, which is different from that for unirradiated nanocrystals related to the formation of shear banding.

To study the effect of grain size distribution on the mechanical behaviours of nanocrystals, NC copper under uniaxial tension is performed at the strain rate of $10^{-5}$ s$^{-1}$ with different variances. For a fixed average grain size $d = 30$ nm and different variances (10, 50 and 200), the corresponding number weighted probability and volume weighted probability are depicted in figure 5a,b. For both the unirradiated and irradiated cases, there exists a few hundreds MPa drop of the flow stress with the increase of variance as shown in figure 5c,d. In fact, it can be seen in figure 5b that a higher percentage of the grains in NCs will consist of large grains with low strength when the variance = 200, which results in the decrease of the whole strength of NC materials. Comparing figure 5c,d, the irradiation effect on the mechanical behaviours for grain size $d = 30$ nm is not obvious because of the dramatically decrease of irradiation-induced defects, which are absorbed by the large volume fraction of GBs.

The macroscopic responses of unirradiated NC copper based on the EVPSC method for different average grain sizes are presented in figure 6a. The variance of the grain size distribution
is fixed at 20. The load strain rate is $10^{-5}$ s$^{-1}$ and the dpa = 0.1. An average grain size is fixed at 30 nm. When the average grain size decreases from 100 nm to 28 nm, the yield strength will increase because of the increasing slip resistance of dislocations emitted from GBs as shown in equations (2.10) and (2.11). Meanwhile, the mechanical responses are close to that of the GI, which is owing to the high volume fraction of the GI relative to that of GBs. With the further decrease of grain size from 28 to 4 nm, the strength decreases due to the high volume fraction of GBs and the increasing influences of GB sliding and GB diffusion on the whole deformation. The irradiation hardening phenomenon is observed in figure 6b when the irradiation dose dpa = 0.1. The relative increase of the yield stress due to irradiation drops with the decrease of grain size, i.e. from approximately 41% at 100 nm to approximately 12% at 50 nm, which is mainly ascribed to the reduction of irradiation-induced defects absorbed by the large volume fraction of GBs. With the onset of yield stress, the irradiation-induced defects would be annihilated by slipping dislocations, which may induce the post-yield softening phenomena as observed in experiments [3].

The size effect without irradiation is demonstrated through the Hall–Petch plot as presented in figure 7. Modelling results are illustrated in solid lines, while experimental data are represented with symbols [61–64]. It can be seen that (i) the yield strength almost increases linearly with the inverse square root grain size until it decreases to approximately 27 nm. (ii) The theoretical results match well with the experimental data [61,62], which is consistent with the Hall–Petch law. (iii) Below the grain size of about 27 nm, the classic Hall–Petch law is broken that the strength decreases with the grain size, which is the inverse Hall–Petch effect as observed in the
Figure 6. Macroscopic stress–strain relations of NC copper under uniaxial tension regarding different grain sizes (a) without irradiation and (b) with irradiation. The load strain rate is $10^{-5}$ s$^{-1}$ and the dpa = 0.1. The variance of grain size distribution is fixed at 20. (Online version in colour.)

Figure 7. Hall–Petch plot for NC copper without irradiation. The simulation results are compared with experimental data (star [61]; triangle [62]; inverse triangle [63]; square [64]). Above 27 nm, the ultimate strength increases with the inverse square root of grain size. Below 27 nm, the inverse Hall–Petch law is broken due to the softening effect of GBs as observed by experiments [65–67]. (Online version in colour.)

Experiments [65–67]. (iv) Besides the difficulty to form the pile-up mechanism in nanograins, the break-down of the Hall–Petch law is due to the softening effect of the high ratio of GBs when the grain size gets small, which includes the GB slipping and GB diffusing mechanisms. (v) The grain size of about 27 nm with the maximum strength can be well predicted by the present model.

Figure 8 shows the comparison of the yield strength between the irradiated (dpa = 0.1) and unirradiated cases. It is indicated that (i) there exists a critical size of approximately 28 nm that separates the grain size region into the irradiation hardening region and the irradiation softening region. Noted that this critical size is different from the critical grain size $d_{p}$ to distinguish the partial and full dislocation emission. (ii) In the irradiation hardening region, the yield stress increases because of the impediment of slip dislocations by irradiation-induced defects. While the hardening effect decreases with grain size because of the absorption of defects by GBs. (iii) In the irradiation softening region, GB sliding and GB diffusion gradually dominate the macroscopic
behaviours of NC. Under irradiation, numbers of irradiation-induced defects near the GBs will be absorbed into GBs. These absorbed defects will lead to the decrease of the sliding friction coefficient in GBs [24] as shown in equation (3.3), which makes the macroscopic behaviours of the irradiated NC softer than that of the unirradiated case.

Figure 9 gives a schematic of the proposed deformation map with/without irradiation effects, in which the contributions of different mechanisms (i.e. GI deformation, GB sliding and GB diffusion) to the plastic deformation of NCs are illustrated. Regions I, II and III, respectively, indicate the contributions of GI deformation ($\dot{\varepsilon}^{I,vp}$ in equation (2.3)), GB sliding ($\dot{\varepsilon}^{BS,vp}$ in equation (3.4)) and GB diffusion ($\dot{\varepsilon}^{BC,vp}$ in equation (3.6)). The contribution is calculated by the formula $\dot{\varepsilon}^{j,vp}/\dot{\varepsilon}^{HI,vp}$ ($j = I, BS$ and $BC$) when the plastic deformation happens. The red dot-dashed lines are the boundaries of different mechanisms without the irradiation effect, while the blue dashed lines are the boundaries with the irradiation effect. Regarding the unirradiated polycrystal with a large grain size, the predominant deformation mechanism originates from the slip of full or partial dislocations in the GIs. With the decrease of grain size, the contribution

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**Figure 8.** Comparison of the yield strength with respect to the inverse square root grain size for both the irradiated (dpa = 0.1) and unirradiated cases. There exists a critical grain size, above which is the irradiation hardening region; below which is the irradiation softening region. (Online version in colour.)

**Figure 9.** A schematic of the deformation map for nanocrystals with/without irradiation effect. Region I: GI deformation; region II: GB sliding; region III: GB diffusion. (Online version in colour.)
(owing to GBs including GB sliding and GB diffusion) to the crystalline plastic deformation behaviours increases. When the grain size decreases to a few nanometers, GB sliding and diffusion become the dominant.

Regarding the irradiated polycrystal, the boundaries of different mechanisms are changed. As shown in figures 8 and 9, when the grain size is above 28 nm, the irradiation-induced defects can lead to irradiation hardening, which increases the plastic strain rate of GB sliding and GB diffusion according to equations (3.5) and (3.6). Therefore, the contributions of GB sliding and GB diffusion to the plastic deformation increase. When the grain size is below 28 nm, the flow stress would decrease due to the irradiation softening effect as shown in figure 8. Therefore, the contributions of GI deformation and GB diffusion to the plastic deformation decrease. However, the contribution of GB sliding increases because of the decreased slip friction among GBs.

6. Conclusion

In this paper, we present a micromechanical framework to study the mechanical behaviours of FCC NC metals with irradiation effect. At the grain level, a core-shell model is proposed to consider the influence of irradiation on the mechanical behaviours of individual grains. The plastic deformation of the GI is dominated by the dislocation–defect interaction and the slip resistance of partial or full dislocations. The effect of irradiation on the properties of GBs is taken into account through the GB sliding friction, which may lead to irradiation softening. The EVPSC method with the influence of grain size distribution is applied to obtain the macroscopic behaviour of NCs with irradiation effect. Numerical results of the irradiated NC copper show that

1. Without irradiation, as the average grain size decreases from tens of nanometers to a few nanometers, the dominant deformation mechanism for unirradiated NC metals transforms from GI sliding to GB diffusion and sliding.

2. With irradiation, the irradiation-induced defect hardening and the slip resistance of dislocations emitted from GBs will dominate the GI behaviours. With the decrease of grain size, due to the increasing absorption of defects by GBs, irradiation hardening would be weakened, which indicates the irradiation hardening is dependent on grain size. The absorption of irradiation-induced defects will reduce the average sliding-friction resistance of GBs, which makes the GBs softer than the unirradiated case.

3. The model shows that there exists a critical size for irradiated NC metals, which separates the grain size region into the irradiation hardening region and the irradiation softening region.

4. Grain size distribution has a significant influence on the macroscopic behaviours of NC materials for both the irradiated and the unirradiated cases. Increasing the variance of grain size distribution leads to the decreasing flow stress of NCs, which is ascribed by the effect of large grain with a low strength.

Acknowledgements. The authors thank the reviewers’ for their helpful suggestions.

Funding statement. This work has been funded by National Natural Science Foundation of China (NSFC) under Grants 11225208, 11172001, 91226202, and the key subject ‘computational Solid Mechanics’ of China Academy of Engineering Physics.

Author contributions. The authors are all responsible for the establishment of the theoretical model.

Conflict of interests. We have no competing interests.

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