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Journal of Nuclear Materials 249 (1997) 103–115

journal of nuclear materials

Radiation hardening revisited: role of intracascade clustering

B.N. Singh [a,*], A.J.E. Foreman [b], H. Trinkaus [c]

[a] *Materials Research Department, Risø National Laboratory, DK-4000 Roskilde, Denmark*
[b] *Materials Performance Department, Harwell Laboratory, Didcot, Oxon. OX11 0RA, UK*
[c] *Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany*

Received 25 March 1997; accepted 7 July 1997

Abstract

Experimental observations related to the initiation of plastic deformation in metals and alloys irradiated with fission neutrons have been analyzed. The experimental results, showing irradiation-induced increase in the upper yield stress followed by a yield drop and plastic instability, cannot be explained in terms of conventional dispersed-barrier hardening because (a) the grown-in dislocations are not free, and (b) irradiation-induced defect clusters are not rigid indestructible Orowan obstacles. A new model called 'cascade-induced source hardening' is presented where glissile loops produced directly in cascades are envisaged to decorate the grown-in dislocations so that they cannot act as dislocation sources. The upper yield stress is related to the breakaway stress which is necessary to pull the dislocation away from the clusters/loops decorating it. The magnitude of the breakaway stress has been estimated and is found to be in good agreement with the measured increase in the initial yield stress in neutron irradiated copper. © 1997 Elsevier Science B.V.

1. Introduction

It is well established that neutron irradiation causes a substantial amount of hardening and alters significantly the deformation behaviour of metals and alloys, particularly at relatively low irradiation temperatures (i.e., below recovery stage V). The literature on this topic clearly demonstrates that various aspects of the problem of radiation hardening were investigated both experimentally and theoretically in considerable depths already in the 1950s and 1960s. Since the early investigations of irradiation hardening by McReynolds et al. [1] and Blewitt et al. [2], a large number of papers have been published on this topic. A vast amount of results have been reported illustrating, for example, the effects of variables such as irradiation dose and temperature, impurities and alloying, cold-work and prestraining and crystal structure on the increase in the critical resolved shear stress (CSS) or flow stress caused by neutron irradiation (see Refs. [3,4] for reviews).

* Corresponding author. Tel.: +45-46 775 709; fax: +45-46 775 758; e-mail: afm@risoe.dk.

In an attempt to explain the experimental observations, two main hardening mechanisms have been proposed. The most popular one is the 'Zone Theory of Radiation Hardening' proposed by Seeger [5] and is commonly known as a dispersed barrier hardening (DBH) model. The 'Zone' refers to the 'vacancy-rich zone' in the center of a displacement spike [6]. These zones are expected to collapse into vacancy loops or stacking fault tetrahedra (SFT_s) during the cooling down phase of the cascades. In Seeger's hardening model, the vacancy clusters/loops produced in the cascades are assumed to act as barriers to gliding dislocations in the slip plane and therefore are taken to be the main source of radiation hardening. It should be noted here that the hardening mechanism considered in Seeger's model is essentially the mechanisms proposed by Orowan [7–9] for hardening due to precipitates and dispersed particles.

Another model for the radiation hardening has been considered in terms of formation of 'defect clouds' along the length of the grown-in dislocations [2] (i.e., dislocations present already before irradiation) such that these dislocations cannot act as dislocation sources. In other words, plastic deformation cannot be initiated until these dislocations are pulled away from the clouds of defects

PII S0022-3115(97)00231-6

[2,10–12]. The stress necessary to unlock these dislocations so that they can act as dislocation sources again is then likely to determine the yield strength of the irradiated crystals. This mechanism of radiation hardening is commonly known as the 'source' hardening (see later, for discussion).

In spite of the fact that a large number of investigations have been devoted to this topic, the controversy as to which of these two mechanisms is the correct one has still not been resolved. From the point of view of establishing a proper understanding of the deformation behaviour during irradiation, it is imperative, on the other hand, to resolve this controversy. In the past, one of the major problems in resolving this issue has been the lack of a detailed knowledge of the damage morphology produced within cascades and subcascades generated by energetic neutrons. It has been shown only recently, for example, that in copper neutrons are likely to produce displacement subcascades (e.g., [13–15]) and that several clusters of self-interstitial atoms (SIAs) are likely to be produced directly in a cascade or subcascade [16,17]. Furthermore, some of these SIA clusters may perform one-dimensional glide [16]. The possibility that the one-dimensional glide may play a decisive role in decorating the grown-in dislocations by small loops has been investigated by the present authors and the results are described in the accompanying paper [18]. Thus, in reality the structure of the damage produced by neutrons is far more complicated than the production of one vacancy loop (i.e., an obstacle) per collision event commonly assumed while considering the problem of radiation hardening.

Recognizing this improvement in our understanding of the primary damage production in displacement cascades and the fact that a number of experimental observations on deformation behaviour of irradiated materials are not consistent with the DBH model, we decided to 'revisit' the controversial topic of radiation hardening. There are three landmark observations on the plastic deformation behaviour of irradiated materials that cannot be explained within the framework of the DBH model, and these are as follows.

(i) A large increase occurs in the flow stress of materials irradiated with neutrons at temperatures below the recovery stage V but without generation of dislocations in a homogeneous fashion.

(ii) Even fcc metals and alloys irradiated beyond a certain dose level at temperatures below stage V exhibit a sharp and prominent yield drop as in unirradiated bcc iron (see Section 2).

(iii) Beyond the yield drop, plastic deformation occurs in a localized and heterogeneous fashion and practically all of the plastic deformation is concentrated in narrow bands ('cleared channels') representing only a small fraction of the specimen volume. During deformation, the gliding dislocations sweep out practically all defect clusters from these cleared channels, making them almost completely free of irradiation-induced defects and their clusters (see Section 4 for more details). Beyond a certain dose level, polycrystalline metals and alloys exhibit no work hardening and suffer from plastic instability (see Section 4).

In the present paper we assess the role of the pre-irradiation microstructure and damage production characteristics (e.g., intracascade clustering of SIAs) particularly in the initiation of plastic deformation in materials irradiated at temperatures below the recovery stage V. To facilitate the discussion of irradiation-induced hardening, first of all, some of the basic features of dislocation generation and plastic flow initiation processes in unirradiated materials are briefly outlined in Section 2. The general nature and the evolution of irradiation-induced microstructure are described in Section 3. This is followed by the experimental evidence for the irradiation hardening (Section 4). A summary of the conventional dispersed-barrier hardening model is presented in Section 5. The analysis of the microstructure and mechanical performance of metals and alloys with a low density of grown-in dislocations and a high density of irradiation-induced very small defect clusters or loops shows that it may not be appropriate to use Orowan type of mechanism to describe the observed radiation hardening. An alternative hardening mechanism, cascade-induced source hardening (CISH), is described in Section 6. In order to test the validity of the CISH model, the upper yield stress has been estimated by calculating the stress necessary to unlock the dislocations (decorated with small SIA loops) so that they may act as Frank–Read sources. The summary and conclusions of the present calculations are given in Section 7.

2. Dislocation generation and plastic flow initiation

In order to identify the role of irradiation-induced defects in modifying the plastic deformation behaviour of irradiated materials, it is, in our view, necessary to distinguish between the initiation (yielding) and continuation (i.e., work hardening or softening) stages of the plastic flow. The initiation of the plastic flow, commonly referred to as yielding, occurs when a significant number of grown-in dislocations are made free (i.e., unlocked) to move and act as dislocation sources. The multiplication and movement of these free dislocations and the resulting dislocation–dislocation interactions leads to work hardening as plastic deformation continues. For the sake of simplicity, in the following we shall consider only the initiation stage of the plastic flow.

It is well known that in a perfect (i.e., dislocation free) crystal, plastic deformation is not initiated until a strain of about 0.1 is reached [19]. However, in a large and soft single crystal plastic deformation begins already at a strain level of $\approx 10^{-5}$. According to Cottrell [20], the slip is nucleated heterogeneously and the grown-in dislocations (present before the application of load) provide the nucleation sites for the slip. A certain fraction of the grown-in dislocations (which are 'clean' and mobile) may be able to generate a large number of new dislocations by acting as Frank–Read (F–R) sources [21]. The 'clean' and 'atmosphere'-free dislocations will start operating as F–R sources when the applied stress is sufficient to overcome the restoring force on the dislocations due to their line tension, i.e., when the applied stress is about Gb/l where G is the shear modulus, b the Burgers vector and l the length of the dislocation segment (i.e., F–R source). According to this relationship, the F–R sources in pure single crystal of copper will be expected to operate at a stress level of about 4 MPa (for ρ (dislocation density) $\approx 10^{11}$ m^{-2}). This is about one half of the critical resolved shear stress (CRSS) value for single crystal copper reported recently by Dai [22]. In the extreme case of heavily ($\approx 85\%$) cold-worked copper, the stress necessary to operate F–R sources is estimated to be ≈ 590 MPa (with $\rho = 2 \times 10^{15}$ m^{-2} [23]) which is only about 30% higher than the experimentally observed value of the yield stress (see Section 4). Thus, it is the creation of new dislocations by the applied stress which determines the initiation of the plastic flow.

The formation of an 'atmosphere' of solute atoms around dislocations (i.e., Cottrell atmosphere [24]) is well established. This means that in order to initiate the plastic flow (i.e., to make the dislocation segments with 'atmosphere' act as F–R. sources), a higher stress will have to be applied, so that the dislocations could be pulled out of their atmosphere. Hence, an increase in the yield stress would be expected. This is also the condition which gives rise to the appearance of the upper yield stress (σ_{UY}) (Fig. 1) [25–27]. Thus, at the upper yield stress the dislocations are unlocked from their atmosphere and are free to move through the lattice. This transition from the 'locked' to free (i.e., from immobile to mobile) state leads to a sudden drop in the applied stress (i.e., the yield drop) since the stress necessary to unlock the dislocation is higher than the stress needed to keep the dislocations moving once they have been unlocked.

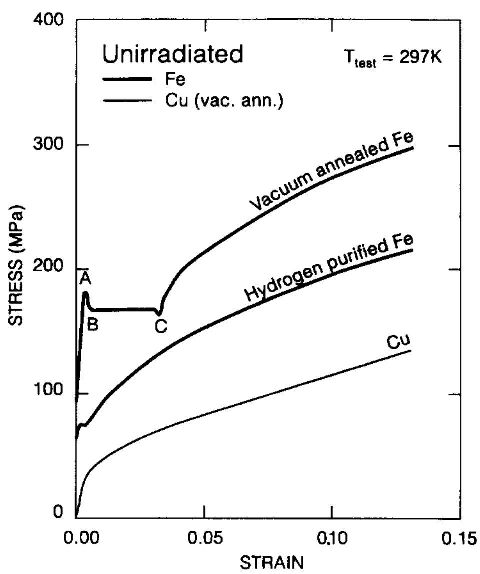

Fig. 1. Stress–strain curves for iron [25] and OFHC-copper [26,27] tensile tested in the annealed and unirradiated condition.

The abrupt yield drop has been treated also in terms of rapid multiplication of dislocation and the stress dependence of dislocation velocity [28]. The model predicts a prominent yield drop when the density of unlocked dislocations is very low (10^{6}–10^{8} m^{-2}), e.g., when most of the grown-in dislocations are locked. On the contrary, essentially no yield drop should occur when most of the grown-in dislocations (10^{10}–10^{12} m^{-2}) are not locked and are free to move. The values of σ_{UY} and the magnitude of the yield drop ($\sigma_{UY} - \sigma_{LY}$) depend on the density of unlocked (mobile) dislocations and the rate of their multiplication [28]. After the yield drop, the plastic deformation may proceed in the form of 'uniform' or 'non-uniform' yielding. The phenomena of the upper yield point and yielding uniformly or non-uniformly are observed commonly in iron and iron alloys whereas pure fcc metals (e.g., copper) normally do not show the upper yield point. However, the upper yield point has been observed in copper crystals containing zinc [29]. The locking of the edge component of the partial dislocations is assumed to occur by elastic ('hydrostatic') interactions [30].

3. Irradiation-induced defect microstructure

Before considering the response of irradiated materials to an applied stress, it is very useful to know the main features of the microstructure which evolve during irradiation. This is an important issue, since it is the composition of the microstructure which will determine the nature of the materials response. In recent years, a large number of investigations have been carried out to determine the evolution of the defect microstructure, particularly in fcc metals following neutron irradiations. Various aspects of defect production [31] as well as accumulation [32] under cascade damage conditions have been recently reviewed. In the following, therefore, we shall briefly summarize only those microstructural features which are relevant to the problem of irradiation hardening.

As demonstrated by molecular dynamics (MD) simulations [16,17], the defect production under cascade damage conditions (e.g., during neutron irradiation) has the following distinct features (at the end of the cooling-down phase of the cascade):

(i) intracascade clustering produces clusters of both vacancies and SIAs directly in the cascade volume,

(ii) several SIA clusters of different sizes are produced in each cascade,

(iii) small SIA clusters are mobile; they glide one-dimensionally as a group of crowdions.

Recently, MD simulations have further demonstrated that the clustering behaviour of SIAs in bcc iron may be significantly different from that in fcc copper [33]. For instance, the ratio of small (mobile) to large (immobile) SIA clusters produced directly in the cascades (at the end of the cooling-down phase) may be significantly greater in iron than that in copper.

The dose dependence of radiation damage accumulation in the form of defect clusters (loops and stacking fault tetrahedra (SFT_s) has been determined for neutron irradiated fcc metals (e.g., Cu, Ni) in a number of studies using transmission electron microscopy (TEM) (see Refs. [22,32] for review). An example of the dose dependence of cluster density in copper irradiated at temperatures in the range of 298–363 K is shown in Fig. 2 [32]. The main conclusion of these investigations is that at doses below $\approx 10^{-2}$ dpa, the increase in cluster density is proportional to the neutron fluence (or displacement dose). It should be noted that at these irradiation temperatures, about one half of the clusters formed during neutron irradiation of copper are of vacancy type, resolved in TEM as SFT_s; the other half are likely to be of interstitial type composed of SIAs. The fact that the cluster density is proportional to the neutron fluence would suggest that the observed clusters are produced directly in the cascades or subcascades. There are, however, some other TEM results for copper irradiated to intermediate doses (i.e., $> 2 \times 10^{-4}$ dpa) (Fig. 2) which show square root dependence of cluster density on the displacement dose, indicating interaction among adjoining cascades. The cluster density in copper irradiated at temperatures up to ≈ 360 K reaches a saturation level at doses somewhat below 10^{-2} dpa.

It is interesting to note that the dose dependence of cluster density in nickel irradiated with neutrons is similar

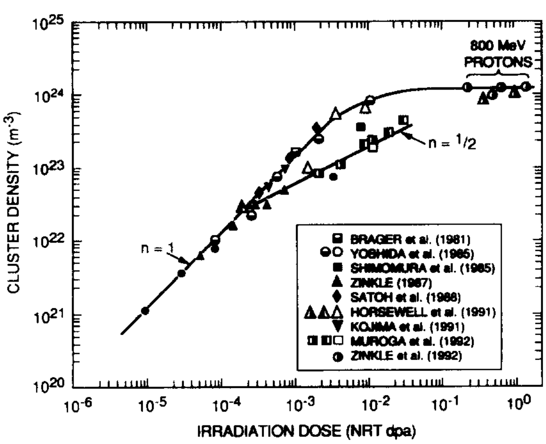

Fig. 2. Dose dependence of cluster density in copper irradiated with fission and fusion neutrons and 800 MeV protons at temperatures in the range of 298–363 K [32].

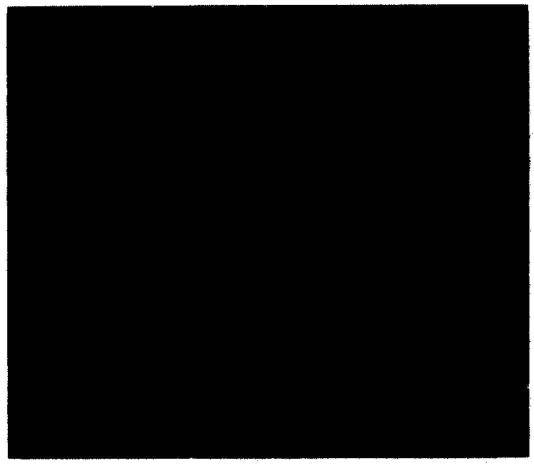

Fig. 3. A TEM micrograph showing a high density (5×10^{23} m^{-3}) of small clusters in copper irradiated at ~ 320 K to a dose level of 0.01 dpa. Note the lack of grown-in dislocations.

to that in copper and the cluster density in nickel also reaches the saturation level at doses below 10^{-2} dpa. However, the cluster density in nickel at a given dose level and at a similar homologous irradiation temperature is almost an order of magnitude lower than that in copper. It has been demonstrated recently that the cluster density in single crystal molybdenum irradiated with neutrons at ≈ 320 K saturates at a level almost two orders of magnitude lower than that in copper [34].

It needs to be emphasized here that the production of small but glissile clusters of SIAs in cascades and subcascades leads to decoration of grown-in dislocations by small loops. An extreme example of the localized segregation of loops is the formation of rafts of small loops in polycrystalline molybdenum first reported by Brimhall and Mastel [35]. Recently, similar results have been obtained also in single crystals of molybdenum [34]. The details of dislocation decoration is described in an accompanying paper [18].

Finally, it should be mentioned that apart from the presence of a few decorated dislocations, the spatial distribution of the defect clusters produced in copper during neutron irradiation at around room temperature is rather homogeneous. An example is shown in Fig. 3 [26,27]. It is important to note here that the microstructure is dominated by a high density of homogeneously distributed clusters/loops and SFT_s, but does not contain many dislocation segments.

4. Experimental evidence for radiation hardening

While discussing the deformation behaviour of irradiated materials, traditionally the attention has been focused mainly on the analyses of the influence of irradiation induced defect clusters only on the increase in the flow stress. The description given in Sections 2 and 3 makes it quite clear, on the other hand, that the traditional treatment of the radiation hardening is not at all adequate. What needs to be considered is the general plastic deformation behaviour of the irradiated materials. In order to facilitate such considerations, in the following we first describe some typical examples of experimental observations illustrating the salient features of the effect of irradiation on the general deformation behaviour of metals and alloys. This is followed by the description of the hardening models in Sections 5 and 6.

The deformation behaviour of unirradiated as well as irradiated single crystals and polycrystals of pure copper during tensile testing at room temperature (295 K) is illustrated in Fig. 4. Single crystals (Fig. 4a) were irradiated with 600 MeV protons [22] at 305–315 K, whereas the polycrystalline specimens were irradiated with fission neutrons at 320 K [26,27] (Fig. 4b) and 523 K [36] (Fig. 4c). The stress–strain curves shown in Fig. 4 demonstrate the following.

(a) Both single and polycrystal specimens irradiated at ≈ 300 K and tested at 295 K exhibit a prominent yield drop at displacement doses higher than 0.01 dpa. This illustrates the fact that the initiation of the plastic flow is localized and inhomogeneous.

(b) The occurrence of the yield drop is dose dependent.

(c) In the case of single crystals (Fig. 4a), after the yield drop the plastic flow continues in the form of propagation of Lüders band and produces some work hardening beyond the Lüders strain. In the polycrystalline specimens, on the other hand, no work hardening is observed after the yield drop (Fig. 4b).

(d) Polycrystalline copper irradiated at 523 K to dose levels of 0.1 and 0.2 dpa and tensile tested at 523 K exhibits no yield drop. The specimens irradiated at 523 K and tested at 295 K did not show yield drop either.

(e) In all cases, the yield stress increases markedly with the irradiation dose level.

The plastic deformation characteristics such as yield drop, lack of work hardening and increase in yield stress with increasing dose levels observed in irradiated copper (Fig. 4) are not isolated observations. In fact, these characteristics have been observed in many irradiated metals and alloys. Fig. 5 shows examples of such behaviour reported for 316 L (N) stainless steel [37] irradiated at 500 K ($0.29T_M$ where T_M is the melting temperature) and zirconium alloys [38] irradiated at 558 K ($0.26T_M$). In the case of stainless steel (Fig. 5a), the yield drop (and the plastic instability) is found to occur at a much higher dose level than that in copper, whereas in Zr-2 (Fig. 5b) the yield drop is observed already at a dose level of ≈ 0.5 dpa. It is interesting to note here that the presence of Y_2O_3 particles in Zr-2 prevents the occurrence of yield drop. Furthermore, the addition of 10% Y_2O_3 particles makes it possible for the alloy even to work harden. Strong yield drop and

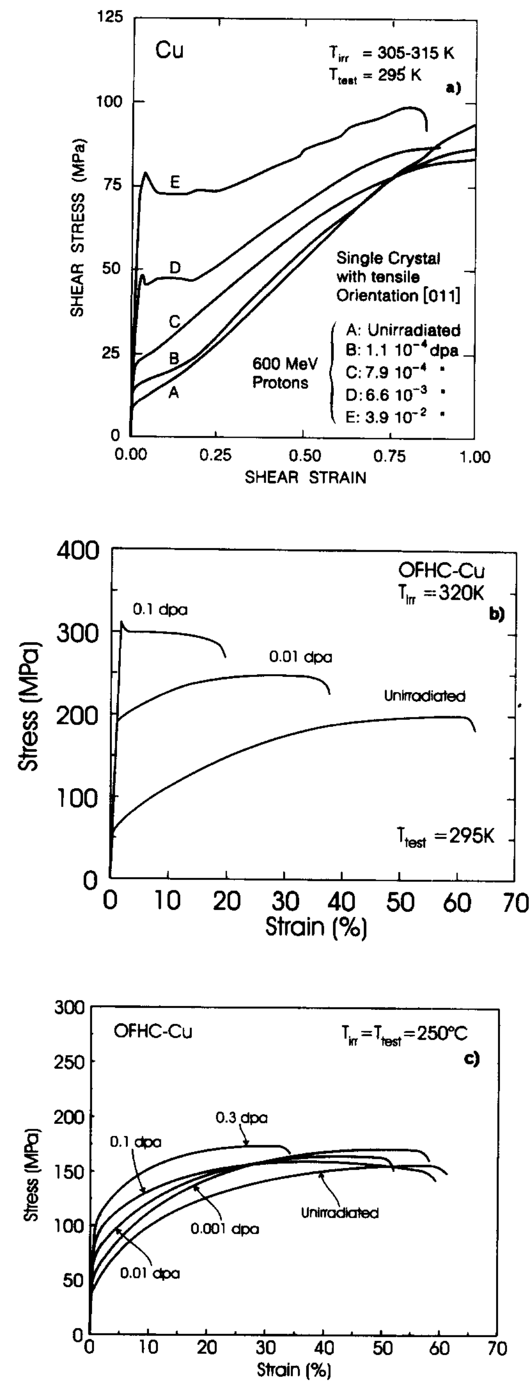

Fig. 4. Stress–strain curves for copper irradiated at various temperatures and displacement doses with 600 MeV protons or fission neutrons: (a) Single crystal irradiated with 600 MeV protons at T_{irr} = 305–315 K [22], (b) polycrystalline copper irradiated with fission neutrons at 320 K [26,27], and (c) polycrystalline copper irradiated with fission neutrons at 523 K [36]. Note the yield drop at $T_{irr} \leq 320$ K (below stage V) and its absence at 523 K (above stage V).

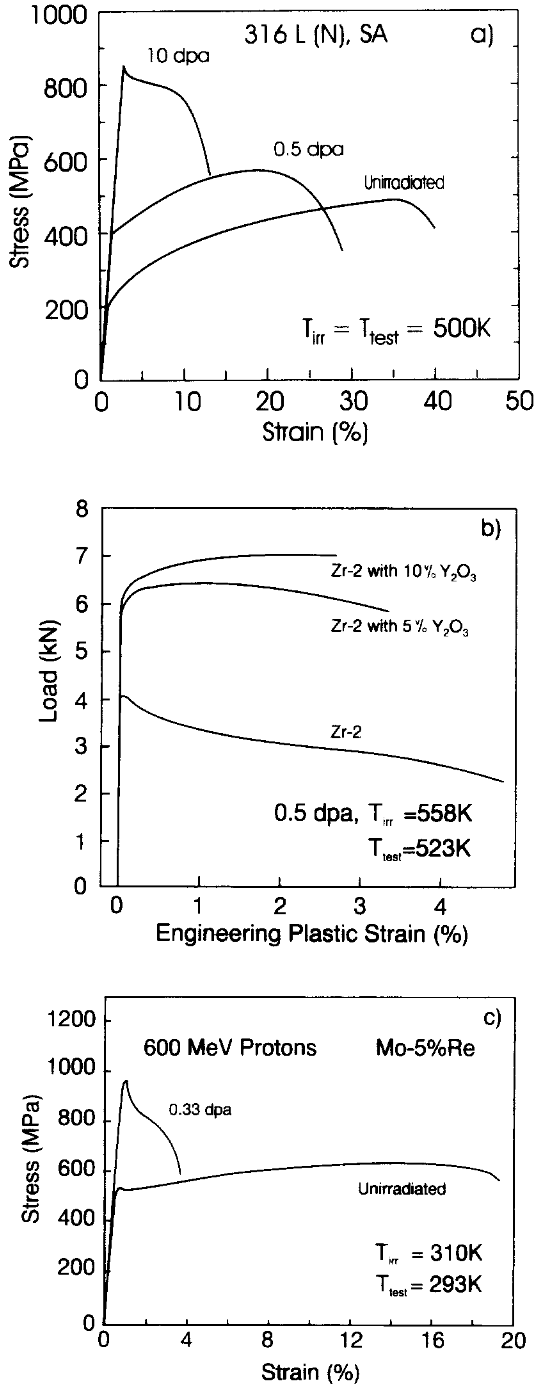

Fig. 5. Stress–strain curves for fcc, hcp and bcc alloys irradiated at temperatures below the recovery stage V: (a) Solution annealed 316 (LN) austenitic stainless steel irradiated with fission neutrons [37], (b) Zirconium alloy (Zr-2) with and without dispersion of Y_2O_3 particles, irradiated with fission neutrons [38], and (c) Mo–5% Re alloy irradiated with 600 MeV protons [39]. Note the occurrence of a yield drop in all cases except in the case of Zr-2 with dispersions of Y_2O_3 particles that may act as Orowan obstacles to dislocation motion.

plastic instability have also been observed in molybdenum and its alloys irradiated with 600 MeV protons [39] and fission neutrons [40] at ≈ 300 K and tensile tested at ≈ 300 K; an example is shown in Fig. 5c [39].

The dose dependence of irradiation hardening particularly in copper has been widely studied. Generally, the irradiation-induced hardening is found to vary as the square-root of the neutron fluence [3,5,22,41]. However, experiments have also shown the irradiation hardening to be proportional to a cube-root of the neutron fluence [2,10,42,43]. The increase in the yield stress due to irradiation has been found to be proportional even to the one-fourth power of the displacement dose [44–46].

Before considering the irradiation hardening models, it is of interest to examine and understand the post-deformation microstructures of irradiated materials. As indicated in Section 3, the pre-deformation microstructure of the irradiated copper samples is composed of a rather low density of grown-in dislocations [$< 10^{11}$ m^{-2}) and a high density of small loops/clusters and tetrahedra distributed homogeneously within the grains. The TEM investigations of deformed copper specimens irradiated at 320 K to a dose level of 0.01 dpa show that during deformation a large number of dislocations are generated throughout the whole crystal [26,27]. These newly generated mobile dislocations interact with each other and form dislocation networks. The interaction of these newly generated dislocations with the irradiation-induced defect clusters lead to a reduction in the density and a change in the spatial distribution of the irradiation induced clusters from homogeneous (in the as-irradiated and undeformed) to heterogeneous (in the irradiated and deformed) one [26,27]. It should be noted that these low dose specimens do not exhibit yield drop and plastic instability. On the contrary, they deform homogeneously and work harden in a normal fashion. (Fig. 4b).

In contrast, the high-dose (e.g., > 0.1 dpa) specimens irradiated also at 320 K and tensile tested at 295 K show practically no evidence of dislocation generation during deformation [26,27]. In fact, the pre- and post-deformation microstructures appear to be very much alike. Furthermore, the density and spatial distribution of the irradiation-induced defect clusters in the irradiated and deformed specimens were almost identical to those in the undeformed specimens [26,27]. Most of the dislocation segments in these high-dose specimens appear to be decorated with small defect clusters. It should be noted that precisely these are the specimens which, during deformation, exhibit a sudden yield drop and plastic instability (Fig. 4b). A similar lack of dislocation generation and interaction (during post-irradiation tensile deformation) has been observed in TZM and Mo–5% Re alloys irradiated at 320 K to a dose level of 0.16 dpa and tensile tested at 295 K [47].

Another well known and significant feature of the post-deformation microstructure observed in irradiated metals and alloys is the formation of 'cleared channels'.

These channels are rather narrow and long bands of materials from which practically all irradiation-induced defect clusters have been removed, presumably by gliding dislocations. The material between the cleared channels, on the other hand, remains completely undeformed, i.e., there are no indications of dislocation generation and motion in the material between the channels. Characteristically, the formation of these channels is observed in specimens irradiated to relatively high doses which also give rise to yield drop and plastic instability. Thus, the occurrence of yield drop, plastic instability and formation of clear channels strongly suggest that in specimens irradiated to relatively higher doses (i.e., $> 10^{-2}$ dpa) plastic deformation occurs in a heterogeneous and localized fashion. The cleared channels have been observed in irradiated and deformed copper [22,48–51], iron [51], molybdenum and its alloys [47,52–54] and niobium [55]. It is interesting to note that these observations are consistent with a deformation mechanism originally proposed by Cottrell [56]. According to Cottrell, plastic deformation in irradiated materials would be expected to occur by the initial slip dislocations sweeping away the irradiation-induced obstacles (i.e., clusters), making the passage of subsequent dislocations easier and creating localized 'run-away' slip. Later, it was established by Sharp [51] that the coarse slip-markings observed on polished surfaces after deformation of neutron-irradiated copper are a direct consequence of the channelling mode of deformation.

Thus, both the mechanical behaviour and the post-deformation microstructure of specimens irradiated at temperatures below the recovery stage V to doses in excess of $\approx 10^{-2}$ dpa demonstrate the following.

(a) The initiation of plastic deformation is controlled by the generation of mobile dislocations,

(b) The newly generated and rapidly gliding dislocations are responsible for the formation of 'cleared channels',

(c) The lack of defect clusters in the cleared channels would suggest that these small clusters are not very strong obstacles to dislocation motion,

(d) Practically all plastic deformation occurs in the cleared channels representing only a small fraction of the specimens' volume.

5. Dispersed barrier hardening (DBH) model

In an effort to explain the observed increase in the critical shear stress for plastic deformation due to neutron irradiation, Seeger proposed the so-called 'Zone Theory' of radiation hardening [5]. He considered that the increase in the critical shear stress "comes about by the thermally and stress activated cutting of dislocations through a forest of obstacles [57]". He assumed that in the irradiated materials these obstacles must be some sort of 'zones that are formed during neutron irradiation'. These zones are assumed to act as barriers to the motion of dislocations 'hung up' (held up) against these obstacles [5]. Clearly, this is a modified form of Orowan theory [7–9] for the 'by-passing' of impenetrable obstacles by the bowing of dislocation segments around them and yields a simple expression for the shear stress τ (in the absence of thermal activation) of the form

$$\tau = \alpha G b / l, \tag{1}$$

where G is the shear modulus, l is the average separation of obstacles (i.e., the length of dislocation segment held up against the obstacles in the slip plane) and α is a parameter representing the obstacle strength. For the case of a random array of obstacles (clusters, loops, 'zones') of diameter d and volume density N, $l = (Nd)^{-1/2}$. Thus Eq. (1) becomes

$$\tau = \alpha G b (Nd)^{1/2}. \tag{2}$$

Assuming that each neutron produces a defect cluster (i.e., 'zone') (i.e., N is proportional to the neutron fluence), the dose dependence of the shear stress increase due to irradiation can be written as

$$\tau = \text{const.}(\varphi t)^{1/2}, \tag{3}$$

where φ is the neutron flux and t is the irradiation time.

It is important to note here that Eqs. (2) and (3) implicitly assume that a sufficient number of mobile dislocation segments are available (prior to the application of load during tensile tests) to give an average segment length of l held up against the obstacles in the slip plane (Eq. (1)). In the absence of grown-in dislocation, the Orowan mechanism is no longer applicable to such a system since there would be no dislocation segments held up against the obstacles that can bow out between the obstacles in response to the externally applied force (stress). In such cases, the increase in the shear stress due to irradiation cannot be calculated from Eqs. (1)–(3) regardless of the number of obstacles present in the system, particularly when the irradiation-induced clusters/loops are very small (2–3 nm) in size.

While discussing the DBH model, the irradiation-induced defect clusters/loops are taken to represent 'soft' (i.e., $\alpha < 1$ in Eqs. (1)–(3) obstacles in the Orowan hardening mechanism. It should be recognized, however, that these small defect clusters would interact with the approaching dislocation segments and finally get 'absorbed' into them (see Section 6). This means that the mobile dislocations cannot get held up against or bow out between these small clusters, leaving a loop around them after by-passing. In other words, the irradiation-induced small clusters may not be appropriate obstacles to mobile dislocations as required for the Orowan mechanism to operate efficiently. The computer simulation of the movement of dislocations through random arrays of obstacles [58,59] is not then relevant to the calculation of the upper yield stress.

Furthermore, the occurrence of yield drop during tensile testing of irradiated materials (Section 4) would suggest that at the point of yielding there is, in fact, a severe lack of mobile dislocations (Section 2). This is consistent with the post-deformation microstructure showing the absence of deformation generated dislocation (Section 4). Thus, neither the mechanical performance nor the pre- and post-deformation microstructures provide the necessary evidence to show that the initiation of plastic deformation in irradiated materials is controlled by the Orowan type of hardening mechanism.

The DBH model assumes that the irradiation-induced clusters/loops act as obstacles to the dislocation motion. The hardening effect has been calculated in terms of specific interactions between gliding dislocations and (a) the 'tetragonal' distortions by the strain field of loops [60] and (b) relatively large (30–50 nm) loops produced during quenching [59,61–63]. These calculations show that both kinds of interactions can produce a significant level of hardening, always provided that the loops/clusters are rigid obstacles to the dislocation motion.

However, plastic deformation in specimens irradiated to an appreciable dose is highly localized and is not consistent with the Orowan mechanism. The deformation is concentrated in narrow bands, which are observed to be swept almost completely clear of defect clusters produced during irradiation. Thus, the small defect clusters are not behaving as rigid indestructible obstacles and are unlikely to be providing a strong impediment to the motion of the gliding dislocations, as envisaged in the DBH model. This implies that the presence of sessile vacancy and SIA clusters in the regions between the decorated dislocations is unlikely to play any significant role in the irradiation-induced increase in the upper yield stress. The presence of these clusters may, on the other hand, play an important role in the evolution of the dislocation decoration by affecting the arrival rate of glissile SIA clusters to the decoration region in the vicinity of grown-in dislocations.

In the case of quenching, the calculations are based on the attractive junction reaction model [64] and assume that each loop intersecting the glide plane provides a pair of forest dislocations. These junctions are then considered to be effective locks to gliding dislocations and hence potent sources of hardening. In the case of the low temperature neutron irradiated pure metals (e.g., copper), this mechanism is unlikely to dominate since the clusters/loops produced during irradiation are too small to provide forest dislocations necessary for forming junctions with the gliding dislocations. As shown later (Section 6.2), instead of forming junctions, these small clusters/loops are likely to get absorbed in the gliding dislocations (see also Ref. [65]). In fact, Foreman and Sharp [66] have shown that the dislocation loops even in the quenched metals can be swept out by the gliding dislocations during deformation.

It is reasonable to conclude, therefore, that the attractive junction reaction model does not appear to be an appropriate model for the calculation of radiation hardening where the clusters/loops are too small to form junctions. In addition, neither of these two mechanisms (i.e., tetragonal distortion and junction reaction) can explain the experimental observations of a sharp yield drop, the absence of deformation-induced dislocations and the lack of homogeneous plastic deformation in the irradiated and tensile tested specimens (see Section 4). On the contrary, these evidences suggest that the increase in the upper yield stress due to irradiation is caused by the difficulty in the generation of dislocations and not in their movement.

6. Cascade-induced source hardening (CISH)

In the present study, we consider an alternative proposition that the irradiation-induced increase in the yield stress (i.e., the stress necessary to initiate the plastic flow) may occur simply because most of the F–R sources (i.e., grown-in dislocations) are 'locked' during irradiation. This possibility was, in fact, first pointed out by Blewit et al. already in 1960 [2]. Because of the lack of detailed information regarding the damage production in cascades, Blewit et al. [2] were not able to specify as to why and how F–R sources may be blocked.

We, on the other hand, submit that most of the F–R sources (i.e., grown-in dislocations) get decorated by small SIA clusters/loops during neutron irradiation. These clusters are assumed to be produced directly in the multidisplacement cascades and small SIA clusters are assumed to be glissile. The main impact of the decoration is that the grown-in dislocations become unable to act as dislocation sources until a stress level much higher than Gb/l is reached. The level of this stress depends on details of the loop distribution in the vicinity of the locked dislocation and particularly on the minimum distance between the loops and the dislocation (see Section 6.2). In the following, we first summarize the main theoretical ideas about the decoration process and then estimate the 'stand-off distance'. On the basis of this knowledge, we proceed to estimate (in Section 6.3) the relation between the loop distribution and the stresses necessary to unlock the dislocation from its decoration so that it can act as a F–R source. This is the stress level which determines the upper yield stress.

6.1. Dislocation decoration process

Small glissile dislocation loops ('coupled crowdions') produced directly in displacement cascades perform a thermally activated one-dimensional motion until they get trapped in the strain field of another defect such as a grown-in dislocation. For low dislocation densities such a dislocation would have a relatively large collection area for accumulating glissile loops from its neighbourhood. At temperatures around $0.4T_M$ the extension of the trapping regions can reach a value as high as about 100 nm for favourable dislocation/loop configurations. Except for a direct encounter of a glissile loop with a dislocation, the loop will generally be trapped in a metastable state from which it may be detrapped by thermal activation. Absorption of such a loop by the dislocation, before it is detrapped, requires a change in its direction of motion, either by a thermally activated Burgers vector (BV) change or conservative climb, or by a mutual approach of the loop and the dislocation in a joint motion in the case of non-parallel BVs. Close to the dislocation, the barrier against Burgers vector changes or climb will vanish and the loop will spontaneously be absorbed into the dislocation. Loop accumulation can only occur outside this region (see Section 6.2). Here, for high loop arrival rates (high loop production rate/low dislocation density), a second loop may arrive in the interaction range of the first loop before this is detrapped or absorbed by the dislocation. The interaction between these two loops, possibly resulting in coalescence, reduces their detrapping and absorption probabilities. The problem of accumulation of glissile loops near grown-in dislocations has been treated in [18] while the influence of the damage rate and irradiation temperature on dislocation decoration has been considered in Ref. [67].

Under such conditions, loops accumulate in the neighbourhood of dislocations. This process is generally accompanied by some loop coarsening. Furthermore, the flux of single SIAs may contribute to the growth of these loops. The loop trapping in the primary trapping region ceases when the attractive stress field of the leading dislocation is compensated by the existing loops. The whole dislocation/loop configuration is then equivalent to a (sessile) dislocation shifted by the extension of the primary region of loop trapping. The loop trapping occurs now only ahead of the existing structure where the elastic interaction remains attractive. Accordingly, the structure grows in the direction where the interaction is strongest and begins to form a dislocation wall there. At a later stage, such walls may even form cell structures with diameters of several micrometers (μm). The formation of such walls has been observed in copper irradiated with fission neutrons [68].

6.2. *Stand-off distance between a loop and an edge dislocation*

Small glissile loops approaching the dislocation to a distance smaller than the stand-off distance get absorbed into the dislocation. Thus, the loop accumulation can occur only at a distance equal to or greater than the stand-off distance (see [18] for details). A simple estimate has been made of this distance of approach at which a small loop will change its Burgers vector on the basis of the line tension approximation. A transformation dislocation is assumed to propagate across the face of the loop in order to change the Burgers vector, which must occur with the assistance of the stress field of the edge dislocation. The energy required to produce this transformation dislocation must be balanced by the work done in shearing the loop behind the transformed zone.

A preliminary estimate suggests that the stress required to change the Burgers vector, τ_{bv}, of a loop of radius r_l is given by

$$\tau_{bv} = Gb/\beta r_l, \tag{4}$$

where β is a correction factor for extreme smallness of the loop and is estimated to have a value of ≈ 3 for tiny loops. The value of β depends on the core energy and may be even smaller than 3. In estimating τ_{bv} the line tension of a normal straight edge dislocation is taken to be $\approx Gb^2/2$, whereas the line tension for the loop component is taken to be $Gb^2/2\beta$. τ_{bv} is the magnitude of the stress that would be produced at a distance of about r_l from a straight edge dislocation (un-dissociated), which is quite a close distance of approach. It should be noted that this represents a lower bound of stand-off distance since it does not include the effect of thermal fluctuation. Thus, the lower bound to the stand-off distance is likely to be when the loop is close to touching the edge dislocation (i.e., of the order of the loop radius, r_l). At elevated temperatures, thermally activated BV changes or conservative climb accelerated due to reduced activation barrier close to the dislocation are expected to cause an increase in the effective stand-off distance.

An alternative mechanism of the loop absorption at a grown-in dislocation could be the break-up of the small SIA loops under the combined influence of a large hydrostatic tension from the dislocation and the ambient temperature. Once a self-interstitial atom breaks loose from the loop, it can escape from the loop and get absorbed into the core of a grown-in dislocation. This is a very simple but a clean mechanism by which self-interstitial atoms created by irradiation can be put back into the lattice.

We examine the feasibility of this mechanism by calculating the escape time, τ_{es}, for a self-interstitial atom from an interstitial loop as a function of distance y (of the loop) from an edge dislocation. The escape time τ_{es} can be shown to be given by

$$\tau_{es} = \left\{\omega_i \exp\left[-\left(E_b^i + E_m^i + p\Delta V\right)/kT\right]\right\}^{-1}, \tag{5}$$

where ω_i is the jump frequency, E_b^i and E_m^i are the binding and migration energy, respectively, for self-interstitial atoms. The pressure, p, exerted by the neighbouring dislocation on the loop at a distance y from the dislocation (at 90° to glide plane) is given by

$$p \approx Gb/2\pi y. \tag{6}$$

ΔV in Eq. (5) is the increase in volume when an atom leaves a loop/cluster and equals $\approx 0.3V_a$ (V_a is the atomic volume $\approx 10^{-29}$ m^3) since the volume occupied by a self-interstitial atom is $\approx 1.3V_a$.

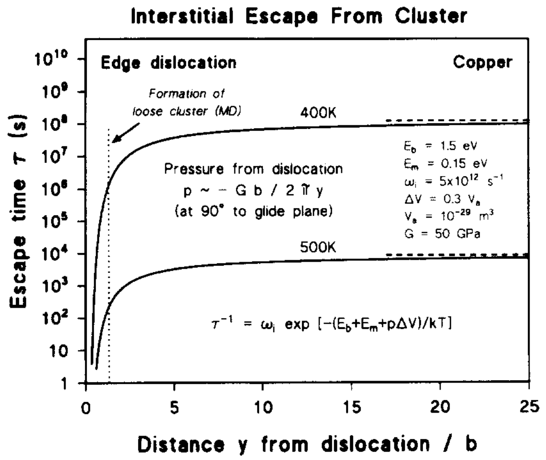

Fig. 6. The dependence of the escape time for an interstitial atom from a small (7 atom) SIA loop on the distance of the loop from an edge dislocation in copper under the influence of the ambient temperature and hydrostatic tension. Note that a small SIA loop when in a close proximity to an edge dislocation may break-up into single SIAs. MD simulations lead to a similar conclusion. The broken lines represent the asymptotic values of the escape time.

The dependence of the escape time on the distance of a 7 atom loop from an edge dislocation in copper has been calculated for 400 and 500 K with $E_b^i = 1.5$ eV, $E_m^i = 0.15$ eV, $\omega_i = 5 \times 10^{12}$ s^{-1} and $G = 50$ GPa; the results are shown in Fig. 6. The results suggest that small SIA loop/cluster may break-up under the combined influence of temperature and a high hydrostatic tension from neighbouring edge dislocation, but only when the loops are very close to the lead dislocation. This is very similar to the conclusion reached earlier in the case of loop absorption by Burgers vector change due to the elastic force field of a neighbouring dislocation.

The possibility of loop break-up was further investigated using molecular dynamic (MD) simulations. Very large hydrostatic tensile stresses were applied to a cluster of 7 interstitial atoms in the form of a symmetrical 'loop' contained in a block of 4000 fcc copper atoms equilibrated at 450 K. At a pressure of −6 GPa corresponding to a loop distance of ≈ 2 b from a pure edge dislocation, the loop was found to have transformed into a loose cluster of self-interstitial atoms (Fig. 6). These loose atoms did not leave the loose cluster most probably because of the absence of a dislocation line and its stress gradient in the vicinity of the cluster in the MD simulation experiment. In any case, the result suggests that the loop/cluster has to be very close (almost touching) to the lead dislocation before break-up may occur. More detailed MD simulations are necessary for a proper evaluation of the break-up and absorption of small loops/clusters into a neighbouring dislocation.

6.3. Relation between microstructure and upper yield stress

As regards the nature of loop/dislocation associations for which the stress necessary to unlock a dislocation from the loop ensemble is to be estimated, two cases may be distinguished: (i) the loops are clearly separated by distances similar to their size (a row of loops), and (ii) the loops are no longer well separated but form a network in which they have, at least partly, lost their individuality.

In the first case, we may neglect the effect of dislocation bowing out between neighbouring loops. For this case, we consider a straight row of sessile edge type loops of Burgers vector, b, diameter d, and spacing l at a distance y (stand-off distance) parallel to a straight glissile edge dislocation of Burgers vector, b, in an elastically isotropic medium of shear modulus G and Poisson's ratio ν. Using the 'infinitesimal loop approximation' we find that the force acting between one loop and the dislocation is maximum ($\partial^2 E/\partial x^2 = 0$) at an angle of about 40° between the distance vector and the glide plane of the dislocation where it assumes a value

$$F_{\mathrm{dl}} \approx 0.069(G/(1-\nu))(bd/y)^2. \tag{7a}$$

This force must be compensated by the force on the dislocation due to the external shear stress τ (resolved shear stress)

$$F_{\tau\mathrm{d}} = \tau b l. \tag{7b}$$

$F_{\mathrm{dl}} = F_{\tau\mathrm{d}}$ yields for the stress necessary to unlock the dislocation (at $\nu = 1/3$)

$$\tau \approx 0.1G(b/l)(d/y)^2. \tag{8}$$

According to Eq. (8), an upper yield stress of the order of 300 MPa for the neutron irradiated copper at ≈ 325 K [26,27] ($G = 55$ GPa) would be consistent with $(b/l)(d/y)^2 \approx 5 \times 10^{-2}$ obtained, for instance, by taking $l = 35b$ (≈ 9 nm) and $y = 0.75d$ which are reasonably consistent with the observed microstructure. The presence of loops with Burgers vectors different from that of the lead dislocation would require lower values of l and y to yield the same upper yield stress of 300 Mpa. More detailed microstructural investigations are required to determine l and y accurately and to establish the relation between the upper yield stress and the microstructure. Furthermore, an accurate value of y needs to be determined using molecular dynamics experiments since the elasticity based calculations become inaccurate close to dislocations.

For the second case, we approximate the loop ensemble accumulated near the dislocation by a sessile dislocation dipole of Burgers vector, b, and diameter, a, separated from the leading dislocation by a stand-off distance y. For $a \gg y$, which is generally fulfilled, the externally applied shear stress has to overcome the maximum attractive shear stress exerted by the close dislocation component of the dipole on the primary dislocation occurring at 22.5° between the distance vector and the glide plane. The corresponding condition yields an estimate for the upper yield stress given by

$$\tau \approx Gb/8\pi(1-\nu)y. \qquad (9)$$

According to Eq. (9), an upper yield stress of the order of 300 MPa for neutron irradiated copper at ≈ 320°C would be consistent with $y = 10b$. This would suggest once again that the stand-off distance, y, is only of the order of a few nanometers which is in a reasonable agreement with the observed spatial distribution of loops in the vicinity of decorated dislocations. A lateral smearing out of the dislocation dipole representing the loop ensemble would require lower values of y. Independent of the precise details, our estimates show that the locking mechanism proposed in the present paper provides a possible explanation for the observed radiation hardening.

It is important to recognize at this juncture that within the framework of the CISH model, the main parameters determining the unlocking stress (i.e., the upper yield stress) are y, l and d (Eqs. (8) and (9)). Therefore, in order to understand the dependence of the upper yield stress on experimental variables such as recoil energy, damage rate, displacement dose and irradiation temperature, the effect of these parameters on y, l and d will have to be investigated. For this, first of all the kinetics of the dislocation decoration phenomenon will have to be established. This has been done in the accompanying paper [18]. The conditions for the dislocation decoration has been further investigated and the results are reported in Ref. [67]. The physical basis for the dose and temperature dependencies of the dislocation decoration as well as the upper yield stress have been also considered in Ref. [67]. It is clear, however, that still further calculations are necessary before these dependencies can be predicted quantitatively and compared with the available experimental results.

7. Summary and conclusions

Experimental results on pre- and post-deformation microstructure, irradiation-induced yield drop and the lack of work hardening and homogeneous plastic deformation in metals and alloys irradiated with fission neutrons at temperatures below stage V have been summarized. A close examination of these results has revealed that several fundamental features of the plastic flow observed in irradiated metals and alloys have been overlooked in the past. In fact, earlier treatments of radiation hardening have considered mainly one aspect of the effect of irradiation, namely the irradiation-induced increase in the yield strength. Practically no effort has been made to understand other important features such as yield drop, lack of work hardening, lack of dislocation generation during deformation and the localized nature of plastic deformation that are commonly observed during deformation of materials irradiated with fission neutrons. Furthermore, while relating the observed increase in the yield strength to irradiation-induced (and pre-deformation) microstructure, only the presence of defect clusters has been taken into account. The role of two other important features of the pre-deformation microstructure such as the presence of dislocations decorated with very small loops/clusters and a very low density of grown-in dislocations, on the other hand, have not been examined.

An analysis of the deformation behaviour observed in neutron irradiated metals and alloys in terms of dislocation generation and plastic flow initiation shows that the experimental results cannot be rationalized in terms of the conventional DBH model. Furthermore, it seems completely inappropriate to use Orowan type of hardening mechanism to explain the increase in yield strength due to irradiation-induced very small (≈ 2 nm in diameter) clusters or loops since these clusters/loops cannot act as rigid and indestructible obstacles to dislocation motion as envisaged in the Orowan mechanism. In fact, these small clusters/loops would get absorbed (see Section 6.2) into a dislocation even before the dislocation comes in physical contact with them. The fact that practically all defect clusters in the cleared channels are swept away by gliding dislocations would also indicate that these small loops/clusters do not act as effective obstacles to dislocation motion. Finally, the observations of yield drops and the lack of dislocation generation and plastic deformation in a homogeneous fashion would suggest that the increase in the yield strength is not because of an increased difficulty in dislocation motion due to the presence of defect clusters. It has been argued that these observations cannot be rationalized in terms of models based on interactions of glide dislocations with tetragonal distortions produced by single SIAs and loops of SIAs [60] or with the forest dislocation junctions produced by the loops intersecting the glide plane [59,61–63].

On the contrary, all the available evidence suggests that the increase in the upper yield stress (i.e., difficulty in plastic flow initiation) may, in fact, be due to the difficulty in dislocation generation. Thus, the problem of radiation hardening should be treated in terms of the Cottrell atmosphere type and not the Orowan type of hardening mechanism. This is the main hypothesis of the cascade-induced source hardening (CISH) model proposed in the present paper. Like the impurity atoms in the Cottrell atmosphere, the 'atmosphere' of tiny SIA loops in the immediate vicinity of grown-in dislocations is assumed to make it difficult for the grown-in dislocations to act as F–R sources. In order to initiate plastic deformation in materials containing these dislocations decorated with small loops, we need to unlock these dislocations so that they can act as F–R sources. Hence, the stress necessary to unlock the dislocations represents the upper yield stress.

The experimental results presented in Section 4 (Fig. 5) clearly suggest that the CISH mechanism must be operating even in the complicated materials such as stainless steel, Zr-2 and Mo–Re alloys. It is quite possible that, in these alloys, dislocations are prevented from acting as dislocation sources not only by small SIA loops decorating them, but also by the irradiation-induced segregation of impurity atoms on the grown-in dislocations and on the SIA loops decorating the grown-in dislocations.

The upper yield stress has been estimated by calculating the stress necessary to pull the dislocations away from the atmosphere of loops decorating them. The results show that the increase in the upper yield stress of neutron irradiated copper can be understood in terms of the breakaway stress.

The formation of the atmosphere of loops decorating the grown-in dislocations has been attributed to the glide and trapping of sessile loops created by the cascades. In this treatment it is assumed that very small loops are absorbed into the grown-in dislocations and only larger ones form the atmosphere at a distance defining the 'stand-off' distance. The mechanisms of very small loops into the grown-in dislocations have been considered in terms of changes in Burgers vector and break-up of very small SIA loops (into single SIAs) under the influence of large stresses in the vicinity of an edge dislocation.

Acknowledgements

The work reported here was partially funded by the European Fusion Technology Program. The authors would like to thank Dr B. van der Schaaf for providing the data on 316 stanless steel.

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