In-situ TEM Observation of Nanoscale Stacking Fault Tetrahedra in A Ni Based Superalloy Inconel X-750

Zhongwen Yao and He Ken Zhang
Department of Mechanical and Materials Engineering, Queen's University, Kingston, Ontario, Canada, K7L 3N6

Nanoscale stacking fault tetrahedra (SFTs) were rarely observed in irradiated alloys, whereas they were frequently found in pure face center cubic (fcc) metals, such as Ni, Cu and Au. In this chapter, SFTs were introduced to the fcc Ni based superalloy Inconel X-750 by heavy ion (1 MeV Kr\(^{2+}\)) irradiation. Irradiation and transmission electron microscopy (TEM) in-situ observations were performed at the intermediate voltage electron microscope (IVEM) Tandem facility at Argonne National Laboratory. The SFTs were mainly characterized using weak beam dark field microscopy. Evolution and annihilation of the SFTs were dynamically observed and captured on video. Formation of the SFTs was initiated by creation of small vacancy type Frank loops from the core of cascades, followed by a loop-SFT transformation process. Vacancy accumulation at the Frank loops stage was observed, and size variations of the SFTs were correlated with the irradiation temperatures and systematically assessed. Two types of SFT annihilation were observed; including 1) transient destruction following cascade damage and 2) gradual shrinkage through absorbing SIAs or emitting vacancies intermediated by an inverse transformation into Frank loops. Types of SFTs, either perfect or imperfect, were statistically analyzed. A size limitation to the SFTs of 6 ~ 8nm were observed. Vacancy Frank loops approximately this size can preferentially collapse into imperfect SFTs, and loop-SFT transformation was not observed in loops any larger. Nucleation, growth, shrinkage and annihilation of SFTs were discussed from the perspectives of atomic displacement as well as energy.

Keywords: transmission electron microscopy, stacking fault tetrahedra, in-situ dynamical observation, weak beam dark field

1. Introduction

Irradiation with energetic particles can cause atomic displacements to produce point defects (self-interstitial atoms and vacancies) in metals within cascades. These point defects may then recombine to annihilate; escape from cascade to become free migrating defects; or accumulate to form clusters during cooling of the cascades. Both experimental survey and theoretical modeling have convincingly demonstrated that the generation and accumulation of point defects can lead to the formation of either planar defects, such as dislocation loops, or 3-dimensional agglomerations, such as voids and stacking fault tetrahedra (SFTs) [1–5]. During the past decades, cascade induced SFTs were evidenced in most of face centered cubic (fcc) pure metals, such as Cu [3,6], Ni [7–9], Pd [10,11] and Au [12], as well as in some fcc alloys [13]. TEM is often used to observe small SFTs, and TEM diffraction condition is a crucial factor to image the SFTs. Based on previous work, Yao et al. [12] analyzed the nature of the SFTs in pure Ni and suggested the best imaging condition to be weak beam dark field condition with \(g(5g) g=020\) close to zone axis <101>. The method is illustrated in fig. 1.

Fig. 1 TEM dark field weak beam, \(g(5g) g=020\), micrographs in irradiated Ni under two different orientations showing the contrast of SFTs. (a) Close to a zone axis <101>, (c) wire frame of a tetrahedron oriented in the same way. (b) Close to a zone axis <001>, and (d) wire frame of a tetrahedron oriented in the same way [1].
Research on irradiation induced SFTs are of great interest due to their ability to hinder dislocation motion, and consequently confer significant hardening to the material. Experimentally, interactions between the SFTs and mobile dislocations have been investigated in various metals, including Cu [6] and Ni [8], etc. Specifically, molecular dynamical (MD) simulations have been utilized to systematically study such interaction in Cu [14]. All these studies clearly presented the pinning effect of the sessile SFTs.

Considerable research has indicated that during irradiation, the SFTs formed directly from cascades or subcascades, and an abundance of experimental data on damage accumulation in fcc metal is available [3,7,10,15–17]. However, the mechanism of SFT evolution from cascades remains to be fully elucidated. Similar to those formed by quenching, electron irradiation and high speed deformation, the cascade induced SFTs were proven to be of the vacancy type in nature [18]. All these techniques can create vacancy supersaturation or condensed vacancy clusters, which are believed to be prerequisite to the SFT evolution [17,18]. The most widely accepted model as proposed by Silcox and Hirsch postulates that the SFTs are formed by collapsing from triangular vacancy type Frank loops [19]. The SFTs with tetrahedron shape, described as 6 ½<110> type stair-rod partial dislocations bounded with four triangular {111} stacking faults, are then formed [2]. This geometric feature is convincing and can be reproduced by MD simulation [17,20]; however, no direct experimental observation has been reported yet. Several parameters may contribute to this transforming process. Schäublin et al. reported that the stacking fault energy (SFE) and shear modulus play key roles, through comparison amongst different fcc materials [10]. The SFTs in Ni and Cu can be easily neglected due to their minuscule size in the nanometer range; the maximum reported by many is ~ 6–8 nm and no SFTs were observed to be any larger [8,10]. The mechanism behind this size limitation is still unknown. It is recognized that in Cu and Ni, recoil energy, dose, and irradiation temperature all impact SFT evolution [21], and these factors will be discussed further in this report. Post formation but prior to destruction, changes in SFTs result from interactions with vacancies, SIAs and their clusters [21]. Apart from the perfect SFTs, Schäublin et al. found that about 36% of SFTs are in fact imperfect in high energy (590 MeV) proton irradiated Cu [10]. These imperfect SFTs are assumed to act more effectively as a sink. Shrinkage and annihilation of SFTs have been studied through thermal annealing of the perfect SFTs [22], irradiation and post irradiation characterizations [23], as well as MD simulations [24]. Nonetheless, there lacks dynamical observation of irradiation instigated annihilation occurring under stress-free state.

Dynamical observation of these nanometric SFTs can be best achieved by TEM coupled with in-situ irradiation. In the current work, dynamical observations of the evolution and development of SFTs in the fcc Ni-based alloy Inconel X-750 are presented and discussed whereas SFTs in Ni-based superalloys were barely reported. Two different types of SFT annihilation are documented and the mechanism is discussed. As well, effects of dose and irradiation temperature are systematically studied.

2. Experimental Setup

The chemical composition of Inconel X-750 is listed in Table 1. The received material was subjected to AMS 5698g standard heat treatment (solution treatment at 1120 °C for 15 mins followed by air cool; 15% cold work reduction and aging treatment at 732 °C for 16 h followed by air cool). Specimens were mechanically thinned to 50 ~ 100 µm and then twin-jet electropolished into TEM samples using a Tenopol-5 apparatus with 5% perchloric acid in methanol electrolyte at -40°C. The microstructure prior to irradiation was characterized in TEM and shown in fig. 2. L11 ordered spherical γ′ precipitates Ni3(Al, Ti) with sizes about 15-25 nm were distributed homogenously within the matrix under two beam bright field observation (fig. 2 (a)). γ′ precipitates are coherent with the γ matrix with a lattice misfit of less than 0.5%, thus the weak contrast around them due to lattice strain variation can be suppressed in weak beam dark field condition (fig. A-1 (b)), and is therefore suitable for the observation of SFT evolution. The polished sample contained negligible amount of SFTs, which were assumed to be formed either by fast cool from high temperature or by the following deformation.

<table>
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<tr>
<th>Table A-1</th>
<th>Chemical composition of the Inconel X-750 (by wt%)</th>
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<tbody>
<tr>
<td>Ni</td>
<td>Cr</td>
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<td>Bal.</td>
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TEM in-situ experiments were conducted at the intermediate voltage electron microscope (IVEM) Tandem facility at Argonne National Laboratory. The facility houses a Hitachi 9000 NAR TEM interfaced to one Tandem ion accelerators [25]. All TEM observations were conducted at an operating voltage of 200 kV. It has been reported that if the tetrahedron is viewed along one of the six <101> directions with \( g = \{020\} \), the structure will project to 2D image as a triangular shape, rather than a trapezoidal shape given by viewing through the <001> direction [1]. Thus \( g(5g) \) weak beam dark field using \( g = 020 \) close to the [101] zone axis were used predominantly for SFT characterization. Edge lengths of the SFTs’ 2-dimensional triangular shapes were measured and regarded as the size of the 3-dimensional
shapes. Beam alignment and stigmatism were carefully adjust to minimize the blurring of the micrographs, and as such some small defects with sizes ~ 0.8 nm which were previously determined as ‘black dots’ can now be characterized.

![Fig. 2 TEM micrographs showing microstructures prior to irradiation. a) Two beam bright field, b) Weak beam dark field. Arrowed defect is an SFT formed during material processing.](image)

### 3. Evolution of the SFTs

Evolution of the SFTs was observed right after the initiation of irradiation and video recorded. Fig. 3 and fig. 4 present snapshots from the video, which captured the formation and annihilation of two SFTs, both recorded during irradiation at 200°C. The time indicated in the micrographs corresponds to real time during the irradiation process. For both SFTs, sudden yields of small loop-like features were observed prior to formation of the perfect SFTs. At the initial stage of evolution of the loop-like features, they are of triangular shape as shown in fig. 3 and 4 at 4s. The loop features then became blurred, which is assumed to indicate their accumulation of free migrating vacancies. More obvious size change of the loops can be spotted in fig. 4 from 4s to 28s, because this loop exists longer than the one shown in fig. 3, allowing more vacancy absorption. The duration of the defects existing as loops varied and showed no obvious regularity. The loops then collapsed into perfect SFTs and the transformation occurred instantly. Conversion from loop to SFT occurred at 10s in fig. 3, and at 32s at fig. 4. After formation, the SFTs were sessile and no obvious size change was observed. The life time of SFTs also varied during the irradiation; some existed for only 1 or 2 seconds before annihilation. Longer lifetime SFTs were illustrated here in fig. 3 and 4 for an easier analyses purpose. It should be noted that, during irradiation, two different types of annihilation were observed. The first SFT shown in fig. 3 was observed to have disappeared suddenly at 22s and a new defect formed adjacent to it. This SFT is assumed to be destructed by another cascade forming in its vicinity. The second type is shown in fig. 4. Between 60 and 64 second, an inverse transformation from SFT to loop was observed. The loop then shrank gradually into disappearance by absorbing free migrating SIAs, and no second defect formed adjacently.

![Fig. 3 Snapshots of dynamical observation of evolution of a SFT formed at 200 °C. Annihilation occurred at 22s suddenly.](image)
Fig. 4  Snapshots of dynamical observation of evolution of a SFT formed at 200 °C. Annihilation occurred from 60s to 79s gradually.

Fig. 5  $g(5g)$ weak beam dark field micrographs, with $g = 020$ close to the zone axis [101], showing the irradiation induced SFTs after irradiated to 0.06 dpa. a) 400 °C, b) 600 °C.

4. Temperature Effects

Similar SFT structure was observed throughout the temperature range of 60°C to 600°C. General observations of the defect structure after irradiated to 0.06 dpa were shown in fig. 5, where (a) is at 400°C and (b) at 600°C. SFTs formed at other temperatures can be found in previous reports. Apart from SFTs, small $\frac{1}{2}<110>$ type perfect loops were also noted in the entire temperature range, and large faulted and unfaulted loops were spotted at temperatures > 500°C[26,27]. The large $\frac{1}{2}<111>$ edge-on faulted loops are shown in fig. 5 (b), most of which were determined as
interstitial in nature and discussed previously [27]. Apparently, irradiation temperature does affect SFT evolution. With increasing temperature, increase in mean sizes of the SFTs can be noted (fig. 5). However, very small (<1 nm) SFTs exist at all temperature, indicating that increase in irradiation temperature did not change the size of the entire SFT population but only the mean size. Size distribution of SFTs in samples irradiated at different temperature but same dose (0.06 dpa) is plotted in fig. 6. For each temperature, approximate 100 SFTs were measured. On the other hand, number density of SFTs is also greatly affected by temperature, as can be generally observed in fig. 5, as consistent with previous literature [26,27].

Similar to Schäublin and Yao et al.’s results on pure metal (Ni, Cu, Au and Pd) [10], SFTs yielding from the current irradiation in X-750 also appear as perfect SFTs or imperfect SFTs. Identification of the SFTs followed Schäublin et al.’s criteria [10], which is achieved by determining the degree of asymmetry of the triangular contrast. Simply put, if one clear edge of the SFT is shorter than the other the SFT is recognized as imperfect. A group of illustrations are shown in fig. 7, where the first row shows experimentally observed perfect SFTs and the second row imperfect SFTs.

Previously we reported the fraction of the SFTs in the total defects to be around 50% ~ 60% and shows no significant change with irradiation dose and temperature from 60°C to 400°C [26]. It should be noted that at low temperature, a large population of defects are smaller and easily tangled together due to a higher number density, easily classified as black dots. At elevated temperatures, although very small defects still presented, but the fraction decreased (fig. 6). A more accurate measurement at 600°C shows 70% percent of the defects to be SFTs. It is the same case when determining the imperfect SFTs. Characterization at 600°C shows about 25% of the SFTs are imperfect.
Large SFTs were observed at elevated temperatures; however, they do not exceed the limit size of 7 nm. Observations of these large SFTs were paid special attention during irradiation and are illustrated in fig. 8 in weak beam dark field or two beam bright field condition respectively. Probably it is more difficult for a very large vacancy Frank loop (~8 nm) to collapse completely to form a perfect SFT. For most of them, as arrowed in fig. 8, the collapsing process does occur but reaction not completed. As such, they would be recognized as imperfect SFTs. No loop-SFT collapsing process was observed for defects larger than 6~8 nm.

5. Collapse Mechanism

Characterization of SFTs in fcc metals has been a subject of research over half a century, however, there are still a few mysteries to be resolved. For instance, the nucleation process proposed by Silcox and Hirsch [19] has been widely accepted but support by direct experimental observation is still lacking. MD simulation has been performed by Yao et al. [17] to study the formation of SFTs in Ni, which has an SFE of 125 mJ·m⁻². In the SFE aspect, this is the closest to the Ni-based superalloy X-750 discussed in this current study. Silcox and Hirsch’s model was successfully reproduced by Yao et al., and a conclusion was reached, which postulates that the nucleation of vacancy clusters performs as a prerequisite for SFT formation. By trying different interatomic potentials, they also suggested that apart from the SFE, some other factors such as mobility of the vacancies and SIAs would also affect the formation of the SFTs [17]. The current experimental observations show good consistency with the MD simulation. In the SFT formation process shown in fig. 3 and 4, vacancy loops are clearly formed prior to their collapse into SFTs. Triangular shape of the loop spotted at 4s in fig. 3 implies this is likely a Frank type loops lying on the {111} plane, facing the observation direction z. As shown in fig. 4 from 4s to 28s, the obvious size change of this Frank loop indicates that the accumulating rate of vacancies, which is determined by the mobility of vacancies, affects the final size of the SFT. From the video, we can see the initial Frank loop or the vacancy platelet formed instantly, indicating they arose directly from quenching of the cascades. It has been experimentally confirmed that the completed SFT would grow by absorbing vacancies during annealing [1]. No obvious size change of the formed SFTs observed here is suggested due to their short life time before destruction. However, comparing to the Frank loop stage, we assume the sink strength of loops for vacancies is greater than the SFTs. The mechanism is still currently unclear. It could be, that due to sum of line energy and SFE, the loops are energetically more favourable than the SFTs [21]. For the same reason, Singh et al. also assumed the imperfect SFTs, which can be treated as an intermediate state of SFT and loop, are more effective as a sink than the perfect ones [21].

6. Annihilation of SFTs

Fig. 3 and 4 also illustrate two different types of SFT annihilation. The first shown in fig. 3 is a direct destruction of the SFT by cascades. Once a new cascade formed adjacent to the pre-existing SFT, the impingement of the cascade would destroy the SFT rapidly, as shown in fig. 3 at 22s. Vacancies of the old SFTs would redistribute and a new defect would likely form close to core of the new cascade. Vacancy loss during the redistribution may make new vacancy type defects smaller and imperfect or even disappear. In the earlier years, Howe and McGurn performed cold irradiations (below 30K) with 100keV O⁺ into Au containing quench induced SFTs and collapse of the SFTs was observed [12]. Vacancies and SIAs are obviously immobile at this temperature. Annihilation of the SFTs thus was suggested to be due to the collision events, although no direct observation was available. MD simulation in Cu has illustrated a cascade exerting to a pre-existing SFT would cause its dissolution and the formation of a new defect [24]. This is consistent with
the annihilation process shown in fig. 3. Singh et al. also noted that under this mechanism, the density of the SFTs saturates when the distance spacing between them equals the size of the cascades [21]. This first type of annihilation depends only on the damage production efficiency, that is, when the next ion will shoot adjacent to the existing SFT, which is determined by the ion flux. If the SFT is ‘lucky’ enough as to have no cascade nucleated close to it for a long time, the second type of annihilation would likely occur. As shown in fig. 4, transformation of the SFT to a Frank loop was spotted and then the loop shrank until disappeared. Basically, this is an annihilation process due to thermal annealing effect, but accelerated by irradiation since free migrating SIAs are produced continuously. The SFT cannot shrink by direct vacancy emission during annealing but will transform to Frank loop first [21], which can then shrink by emitting vacancies. However, unlike single thermal annealing, the vacancy type defects may shrink not only by emitting vacancies but also by absorbing SIAs during the irradiation. Singh et al. stated that the accumulation of SIAs on faces of a SFT may cause its transformation into Frank loops [21]. Kuhlmann-Wilsdorf has proposed an annihilation mechanism of the SFTs which transform first to triangular Frank loops [28]. This current investigation provides solid evidence to the above assumptions. Now we can see, the evolution of the SFTs from the cascade cores, the annihilation by new cascades, the thermal annealing growth and the shrinkage all contribute to the observed fast dynamical construction-destruction-reconstruction process of SFTs. The hardening effect conferred by irradiation is thus not based on the pinning effect of stable SFTs, but by a macro effect of the rapidly refreshing SFTs.

SFE has been reported by many authors through either experimental observation or theoretical modeling to be a key parameter affecting the formation of the SFTs, albeit not the only parameter [10,17]. Schäublin et al. found ~90% of the total defects in Cu to be SFTs but in Ni the percentage is lower at ~50% [10]. In the current investigation, snapshots taken at 600°C is shown in fig. 8, and approximate 70% of the defects are characterized as SFTs. SFE for Cu is 70 mJ m⁻² and is 125 mJ m⁻² for Ni. Information on SFE is unavailable for the X-750 alloy due to its complexities parameter affecting the formation of the SFTs, albeit not the only parameter [10,17]. Schäublin et al. found ~90% of the total defects in Cu to be SFTs but in Ni the percentage is lower at ~50% [10]. In the current investigation, snapshots taken at 600°C is shown in fig. 8, and approximate 70% of the defects are characterized as SFTs. SFE for Cu is 70 mJ m⁻² and is 125 mJ m⁻² for Ni. Information on SFE is unavailable for the X-750 alloy due to its complexities regarding chemical composition, precipitation level, and temperature for measurement. However, the SFE for X-750 should theoretically be lower than pure Ni due to addition of alloying elements, but it should be close to pure Ni. Fraction of SFTs is higher in X-750 than Ni but lower than Cu. A rough assumption thus can be made, that the SFE would affect the loop-SFT transformation which causes the variation in SFT fraction, because SFTs are easier to be produced at low SFE material such as Cu.

7. Conclusion

Inconel X-750 was irradiated with 1 MeV Kr²⁺ in-situ coupled with IVEM observation. The experimental findings are summarized as follows:

The SFT formation was dynamically observed, which demonstrated that the small loops formed directly by collapsing from agglomerated vacancy platelets arising in the core of the cascade, and followed by their instantaneous transformation to SFTs.

Triangular Frank loops prior to collapsing to SFTs can absorb vacancies more effectively than formed SFTs. The temperature-dependent vacancy accumulating rate can cause size increase of a SFT with an increment of irradiation temperature. No obvious growth of formed perfect SFTs was spotted simply due to their rapid annihilation before any changes are noticeable.

Measurements conducted in the 600°C irradiated sample show approximately 70% of the total defects to be SFTs; about 25% of which are imperfect SFTs. Loop-SFT collapsing process would likely be affected by SFE of the material.

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