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1 **Free surface impact on radiation damage in pure nickel by in-situ self-ion**
2 **irradiation: can it be avoided?**

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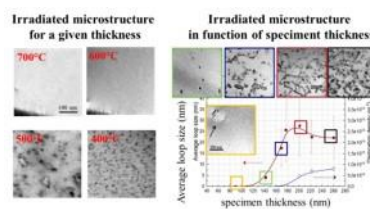
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18

1 **Abstract**

2 A major issue of in-situ radiation damage studies is related to the influence of free
3 surfaces. Free surfaces in thin foils are indeed strong sinks for radiation-induced defects.
4 Nevertheless, in-situ irradiation is a powerful tool to study real-time microstructural
5 evolution and obtain insight into dynamic mechanisms of radiation damage. Thus, a
6 detailed evaluation of surface effects is essential to validate existing results and provide
7 guideline for future comparative experiments. In this work, nickel is chosen as model
8 material to conduct systematic studies on surface effects due to the high mobility of its
9 self-interstitials. Ultra-high purity Ni thin foils are in-situ irradiated by 2 MeV Ni²⁺ ions
10 at high temperatures (400-700°C). Microstructural evolution analysis and detailed
11 characterization of dislocation loops are performed in function of specimen thickness.

12 The present work shows: (i) a drastic influence of thickness on the microstructural
13 evolution and irradiated microstructure with the existence of a critical thickness
14 depending on temperature; (ii) a good prediction of an adequate irradiation thickness with
15 a vacancy concentration calculation model; (iii) an impact of free surfaces on the fine
16 distribution of loop Burgers vectors even above the critical thickness; (iv) a first
17 determination of migration energy of vacancies in Ni considering the temperature
18 dependence of the loop-depleted zones; (v) a production bias model showing that a loss
19 of 10% interstitials favors the growth of vacancy loops observed for the first time in Ni
20 at 510°C.



21

22 **5 keywords**

- 1 in-situ ion irradiation, influence of surfaces, dislocation loops, vacancy concentration
- 2 calculations, production bias model

1 **1 Introduction**

2 In-situ transmission electron microscopy (TEM) is a powerful tool to study the real-
3 time microstructural evolution under irradiation and provides reliable details for atomic-
4 scale modeling in order to obtain valuable insight into dynamic mechanisms of cascade
5 damage and defect evolution [1–5]. Despite significant advantages of in-situ irradiations,
6 some issues still remain in the evaluation of the effects of free surfaces in thin foils. Such
7 effects are discussed in some studies in face-centered cubic (fcc) materials (nickel, copper,
8 tungsten and austenitic stainless steels (ASSs)) [4,6–9]. As strong sinks for point defects,
9 free surfaces affect significantly the defect formation and evolution in thin-foil samples.
10 Thus, surface effects need to be fully characterized and understood in each system, which
11 is useful to better understand the fundamental mechanism of radiation damage and to
12 provide an indispensable guideline for comparative experiments.

13 The irradiation behavior of pure nickel (Ni) and Ni-based alloys as fcc model
14 materials is widely studied to better understand solute effects in fcc structure [10–15].
15 When irradiated specimens are thin foils, the influence of surfaces in Ni is quite
16 pronounced as reported in the literature [6,14] due to the high mobility of self-interstitials
17 (low migration energy calculated as $E_m^i \sim 0.15\text{eV}$ [16,17]). Nevertheless, surface effects
18 in Ni have still not been unambiguously described. Therefore, it is essential to investigate
19 in detail the irradiation behavior of Ni thin foils and to illustrate unequivocally surface
20 effects as necessary guidance for comparative studies of Ni and its alloys.

21 In literature, the investigation of the in-situ behavior of pure nickel in thin foils
22 focused mainly on two aspects: (i) the defect evolution under electron irradiation and (ii)
23 the cascade damage produced by ion irradiation.

1 (i) The first major aspect is the defect evolution under electron irradiation in High
2 Voltage Electronic Microscope (HVEM, voltage generally of 1 MV). Dislocation loops
3 are found to be characteristic in nickel under electron irradiation at low doses [2,18–20].
4 Most of the observed loops were pure edge faulted Frank loops with Burgers vector
5 $\mathbf{b}=1/3\langle 111 \rangle$ of interstitial nature. Vacancy loops were found only close to a dislocation
6 or on the compression side of interstitial loops [2]. Norris [18], Urban [2], Yoo and
7 Stiegler [19] observed a linear growth of faulted loops over a large range of temperatures.
8 The linear growth rate of Frank loops was found when the temperature exceeds 400°C
9 and was temperature-dependent [2]. The growth of interstitial loops under electron
10 irradiation is explained by the preferential absorption of interstitials by dislocations. The
11 development of a chemical rate theory model with the dislocation bias factor was applied
12 to electron irradiated Ni and allowed a good prediction of defect evolution by several
13 authors [18–23]. Norris interpreted the linear growth of interstitial loops as the case that
14 vacancies are immobile and surfaces dominate all other sinks for interstitials. However,
15 Urban and Wilkens [24] and Kiritani et al. [22] showed that this linear growth at high
16 temperature is actually related to the mobility of vacancies. They found that interstitial
17 loops can grow linearly when the temperature is sufficiently high at which vacancies are
18 mobile. Based on this model, they obtained a good agreement between experiments and
19 simulations of the linear growth rate of interstitial loops in fcc metals and its temperature-
20 dependency.

21 However, electron irradiations have their own limitations in representing the
22 radiation damage induced by neutrons. Electron bombardments create no cascades which
23 are a characteristic phenomenon of neutron and ion bombardments [25,26]. The defect
24 evolution during electron irradiation takes no account of the influence of cascades that

1 may drastically modify the nature of defects. The data of loop growth rate under ion with
2 the influence of cascades is unavailable in the literature.

3 (ii) The second aspect is the cascade damage produced by ion irradiation. Results
4 of in-situ ion irradiations in Ni are sparse. The group of Ishino conducted a series of in-
5 situ irradiations using heavy gaseous ions (argon, xenon, and krypton) to study the
6 cascade damage produced during irradiations in Ni [6,27,28]. Their in-situ experiments
7 using 300/400 keV argon ions at 300-773 K showed that clustering of point defects (PDs)
8 was strongly influenced by the presence of point defects sinks: surfaces, pre-existing
9 dislocations, loops and cavities [6]. Wedge-shaped specimens were used to study the
10 effects of surface sinks. It is of great interest to notice two phenomena. Firstly defect-free
11 zones were detected and the thickness of these zones increases with temperature. The
12 second one is the observation of small metastable vacancy defects in thin zones with a
13 very short lifetime during irradiation at 773 K. In thick zones, interstitial loops are formed
14 and grow. These two phenomena show that microstructural evolution depends strongly
15 on the thickness of the specimen, especially at high temperatures indicating a significant
16 influence of surfaces.

17 The in-situ gaseous ion irradiations of Ishino et al. indeed provide a better
18 understanding of cascade damage created in fcc metals. They point out that the surfaces
19 are strong sinks for mobile PDs and that damage distribution is strongly thickness-
20 dependent [1]. Another group reports that, in Ni, the density of dislocation loop induced
21 by 25 keV He⁺ ions irradiated at 400°C increases and then saturates with thickness [29].
22 However, some important effects are not considered. The fact that they used low energy
23 gaseous ions induces several issues: strong inhomogeneous damage through the thickness,

1 the influence of injected interstitials and influence of gaseous impurities [30–32], which
2 may radically change the defect nature and final microstructure.

3 In nickel, the data of in-situ high-energy self-ion irradiation including loop growth
4 rate and detailed final microstructure with the influence of surfaces is unavailable in the
5 literature. Moreover, our previous work shows that large independent vacancy Frank
6 loops can be formed in-depth in self-ion irradiated Ni at high temperature [33], which
7 was not reported before. It raises questions on the surface effects on the nature of defects
8 created in heavy-ion irradiated thin-foil Ni since a clear demonstration is unavailable.
9 Besides, a recent in-situ experiment shows that some loops in Ni can glide over long
10 distances under krypton ions (Kr^+) irradiation at 500°C [15]. In iron, mobile loops are
11 depleted in thin-foil samples compared to bulk samples due to image force [34,35]. The
12 experiment [15] may suggest a preferential absorption of mobile loops by surfaces.
13 However, experimental observations are rarely reported in thin-foil Ni in the literature.

14 To sum up, lack of systematical data on the influence of free surfaces in Ni thin
15 foils under self-ion irradiation raises questions about the representability of irradiated
16 microstructure in thin foils and mechanisms studied from previous in-situ experiments.
17 In recent years, developments in multi-scale modeling and improvements of in-situ
18 techniques are provided. In fcc [36] and bcc [37,38] materials, simulation of long term
19 microstructure under ion irradiation can be performed by rate equation cluster dynamics.
20 High reliability and quantitative in situ experiments are highly desired to validate
21 modeling results for a better understanding of the dynamic mechanisms of radiation
22 damage.

23 In this study, we describe the first systematical campaign of in-situ high energy self-
24 ion (2 MeV Ni^{2+}) irradiations in thin foils of ultra-high purity nickel to characterize the

1 influence of the free surface to provide guidelines for future experiments and modeling.
2 Microstructural evolution during irradiation is recorded and studied. After irradiations,
3 detailed characterizations of the microstructure, particularly dislocation loops (nature,
4 Burgers vector, size, density and spatial distribution) are performed in function of sample
5 thickness and irradiation temperature. Then, the influence of the free surfaces and
6 dynamic mechanisms of radiation damage are discussed in detail.

1 2 Method

2 2.1 Studied materials

3 Rods of 12 mm diameters of nickel (Ni) were manufactured by cold crucible
4 induction melting at the Ecole des Mines de Saint Etienne (EMSE) in France. The
5 measured impurities content in mass ppm was respectively C<8, S<2, O<3, N<2. 400 μm
6 thick slices are cut from the rod and mechanically polished to 50-80 μm thick. 3 mm
7 diameter foils are punched out and annealed at 1000°C for 2 hours in a vacuum of 10^{-7}
8 mbar followed by air-cooling to reduce the dislocation density. Before annealing, three
9 scans with argon atmosphere are conducted to purify the atmosphere in the furnace and
10 pure zirconium getter is used to absorb residual oxygen during the annealing. Finally,
11 TEM thin foils are prepared by twin-jet electro-polishing in a bath of methanol/nitric acid
12 at -30°C at 10V. In TEM, specimens are clean and hold a good surface state. A low
13 density of dislocations ($<10^{10} \text{ m}^{-2}$) is measured by TEM before irradiation.

14 2.2 Irradiation conditions

15 Self-ion in-situ irradiations are performed at the JANNUS-Orsay platform
16 Laboratoire de Physique des 2 infinis Irène Joliot-Curie (IJCLab) in France within the
17 framework of the French National network of accelerators for irradiation and analysis of
18 molecules and materials (EMIR&A) [39]. Thin foils are irradiated in a 200 kV FEI Tecnai
19 G2 TEM with 2 MeV Ni^{2+} ions using a rastered beam produced by a 2 Megavolts tandem
20 Van de Graaff accelerator (ARAMIS). The irradiation temperatures are from 400°C to
21 700°C controlled by a Gatan double-tilt heating holder with an estimated uncertainty of
22 25°C. The ion flux is $4 \pm 0.8 \times 10^{11} \text{ ions.cm}^{-2}.\text{s}^{-1}$. Final fluences are $9 \pm 1.8 \times 10^{13} \text{ ions.cm}^{-2}$
23 and $2.7 \pm 0.4 \times 10^{14} \text{ ions.cm}^{-2}$. Irradiations are performed under a two-beam condition
24 with diffraction vectors type of $\mathbf{g}=\langle 200 \rangle$ or $\langle 111 \rangle$ in kinematical bright-field (KBF)

1 mode ($s_g > 0$) [40]. During irradiation, the microstructure evolution is recorded on video
2 and analyzed after irradiation.

3 The damage profile is calculated by the Stopping Range of Ions in Matter (SRIM)
4 2013 code [41] using the Kinchin-Pease option with a displacement threshold energy of
5 40 eV [25,42]. This damage is also calculated by a recently developed molecular dynamic
6 code IRADINA [43] with the same input parameters as SRIM. Calculated profiles are
7 plotted in Appendix A (Fig. A. 1). The two codes are in good agreement for both
8 displacement damage and implantation profile. The thickness of zones of interest is less
9 than 300 nm. Despite a slight variation of dose over the thickness, the final dose in
10 samples is taken at 200 nm thickness and considered as constant and equal to 0.06 dpa
11 for the lower fluence and 0.18 dpa for the higher fluence. The injected interstitials are
12 very few (0.02 ppm/dpa) and can be neglected.

13 **2.3 Defect analysis**

14 From videos, the evolution of dislocation loops during the irradiation is analyzed.
15 Then, irradiation defects are characterized using a 200 kV FEI TECNAI G2 TEM with a
16 LaB₆ filament located in SRMP laboratory in CEA-Saclay. KBF mode is used to optimize
17 the defect contrast.

18 The size and density of dislocation loops are determined from images under two-
19 beam conditions in kinetic BF mode. The longest distance within the loops is measured
20 manually in ImageJ software and taken as the diameter. Statistic analysis is carried out
21 with an adequate number of loops to calculate their average size. The error bar of the
22 average loop size in figures is the standard deviation. The number density of loops (for
23 short, density of loops in the following) is calculated as the counted number of loops
24 divided by the volume of the analyzed zone. The volume is estimated by multiplying the

1 projected area with the sample thickness estimated by the Convergent Electron Beam
2 Diffraction (CBED) technique [44] along the [220] reflection. The CBED method gives
3 an estimation of the thickness and the extinction distance along the used reflection. Our
4 measurements lead always to an extinction distance of 55 ± 3 nm which is in good
5 agreement with a theoretical value between 57 and 58 nm using jems software [45] (lattice
6 parameter $a_0=0.352$ nm). The error of thickness measurements is always within 5%.
7 Invisible loops in the zone under each two-beam condition are not contained in the
8 calculated number density. The uncertainty of the density results from the error of the
9 count of loops and the measurement of sample thickness. In our case, the lack of precision
10 in the counted loop number is mainly due to small-size indistinguishable loops and large
11 size agglomerated loops. The error is assumed to be 10%.

12 The analysis of loop Burgers vectors is performed using the invisibility criterion
13 [41] and a statistic treatment [46] of at least 5 two-beam conditions images along several
14 zone axes. For each \mathbf{g} , visible loops are counted. A system of equations based on used \mathbf{g}
15 and corresponding visibility proportion is established and resolved by the least-square
16 method programmed in Scilab software with an error given by the standard deviation
17 from the least-squares.

18 The inside-outside method [40,47,48] is used under FS/RH convention to determine
19 the nature of pure edge Frank loops with $\mathbf{b}=1/3\langle 111 \rangle$. As perfect loops with $\mathbf{b}=1/2\langle 110 \rangle$
20 are not generally pure edge, safe/unsafe conditions are determined for studied loops and
21 the nature of loops under safe conditions is analyzed using inside-outside method [48]. A
22 detailed analysis is presented in Section 3. At last, the depth distribution of loops inside
23 the foil is analyzed by the stereo-imaging technique. A spot of contamination at one
24 surface is taken as a reference point. Two KBF images are taken at the same position

1 before and after tilting beta angle $\sim 17^\circ$ keeping the same diffraction conditions. The depth
2 of a loop h is calculated by [4]:

3
$$h = \left| \frac{d_1 \cos \theta_2 - d_2 \cos \theta_1}{\sin(\theta_1 - \theta_2)} \right|$$

4 where d_1 and d_2 are respectively the distances between the loop and the rotation axis
5 passing the surface mark point before and after tilting; θ_1 and θ_2 are beta angles before
6 and after tilting. Then, the thickness of the studied zone is equally divided into 5 nm
7 intervals and an analysis of loop frequency distribution along depth is performed. The
8 depth, at which the accumulated frequency reaches 5%, defines the loop-denuded
9 thickness.

1 **3 Results**

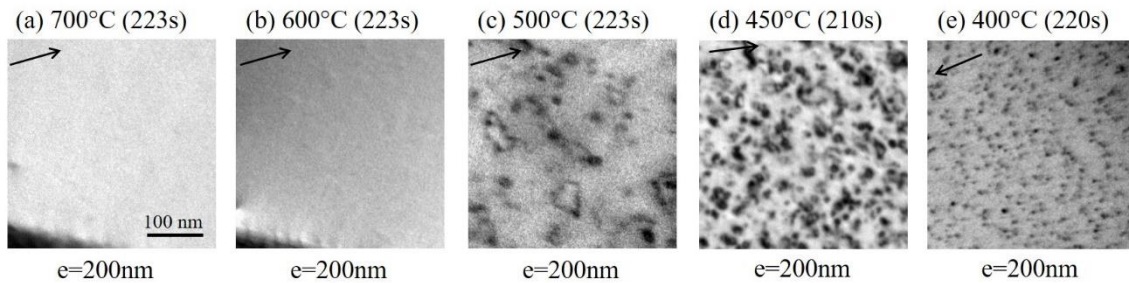
2 In this section, the influence of temperature on defect formation is firstly studied
3 for a thickness of around 200 nm as explained in Section 3.1. The maximum temperature
4 where defects are visible is chosen to evaluate the microstructural evolution and loop
5 growth under irradiation. Then, a post-characterization of the irradiated microstructure is
6 performed to investigate the influence of surfaces on dislocation loops. Finally, the
7 thickness-dependency of the irradiated microstructure is presented at lower temperatures
8 to study the role of temperature on surface effects.

9 **3.1 Determination of an adequate irradiation temperature to study surface effects**

10 General microstructures after irradiation in zones of 200 nm thickness are shown
11 by TEM micrographs in Fig. 1 over a large range of temperatures (from 700°C to 400°C).
12 The objective is to find the maximum temperature at which radiation-induced defects can
13 be formed. The frequently used 200 nm thickness for in-situ studies using a conventional
14 200 kV TEM is chosen for our experiments. The maximum observable thickness of Ni
15 samples in a 200 kV TEM is around 260 nm. If defects can be formed at 200 nm thickness,
16 the thickness marge will be large enough to study the thickness-dependency of the
17 microstructure.

18 Fig. 1(a-e) present the irradiated microstructure at the same dose (0.06 dpa – 220 s
19 of irradiation) at respectively 700/600/500/450/400°C. At 700°C and 600°C, pre-existing
20 dislocations move during the irradiation but newly-created defects are unstable (appear
21 and vanish). In the literature, dislocation loops have been observed in self-ion irradiated
22 bulk Ni at 575 °C [49] and in He⁺ irradiated thin foil Ni up to 700°C at unspecified
23 thickness [29]. The fact that no defect is observed in this work at 600°C and 700°C is

1 very likely attributed to the strong absorption of point defects by surfaces at such
2 thickness.



3
4 Fig. 1. TEM micrographs showing irradiated microstructures for 200 nm thickness for
5 220 seconds of irradiation (equivalent to 0.06 dpa) at various temperatures: (a) 700°C, (b)
6 600°C and (c) 500°C with $\mathbf{g}=\langle 200 \rangle$ close to zone axis $\langle 110 \rangle$; (d) 450°C with $\mathbf{g}=\langle 200 \rangle$
7 close to zone axis $\langle 103 \rangle$; (e) 400°C with $\mathbf{g}=\langle 111 \rangle$ close to zone axis $\langle 110 \rangle$.

8 When the temperature is reduced to 500°C, visible dislocation loops are formed
9 under the same diffraction conditions in the same zone irradiated at higher temperatures.
10 Loops grow during the irradiation. The final microstructure at 0.06 dpa (about 220
11 seconds of irradiation) is shown in Fig. 1(c). Fig. 1(d) and Fig. 1(e) present the final
12 microstructures of two other irradiations at the same dose at respectively 450°C and
13 400°C. Loops are smaller and show a higher density at lower temperatures. Based on
14 these observations, irradiations around 500°C have been chosen for the next parts of this
15 study.

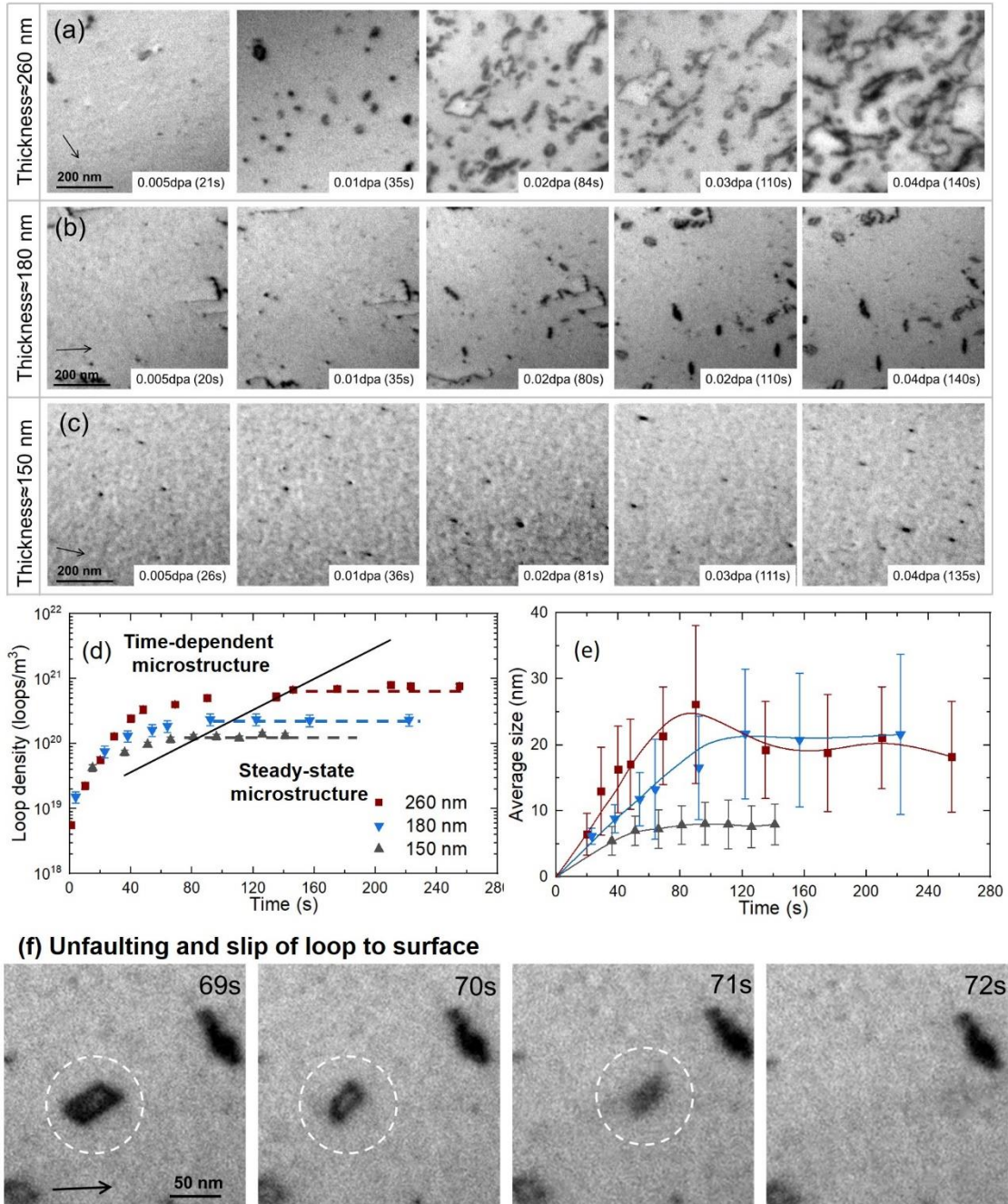
16 3.2 Influence of thickness at 510°C on defect formation and evolution (density, size 17 and growth rate)

18 Fig. 2(a-c) present the microstructural evolution at 510°C at different thicknesses
19 and Fig. 2(d-e) show respectively the evolution of loop density and the average loop size
20 in function of the irradiation duration t . Loop densities and average sizes stabilize at
21 different instants for each thickness. At 150 nm thickness, the loop density and average
22 size are approximately constant when $t > 80$ s, so microstructure appears to be in steady-
23 state. At 180 nm and 260 nm, the steady state starts respectively from around 100 s and

1 130 s. However, the steady state is achieved via different mechanisms. (i) At 150 nm,
2 loops are continuously created during the irradiation. Formed loops grow slightly and
3 most of them quickly disappear, probably to the surfaces. An equilibrium between the
4 loop creation and absorption is established and form a steady-state microstructure. (ii) At
5 150 nm and 180 nm, the phenomena are similar with larger loop growth at 180 nm. The
6 interaction mechanism of a dislocation loop with the surface is shown in Fig. 2(f): a grown
7 Frank loop is unfaulted into a perfect loop which then immediately glides and disappears.
8 (iii) For a thickness of 260 nm, the microstructural evolution is at first similar to the 180
9 nm one, but, from 80 s large loops are numerous and tangle with each other, leading to
10 the formation of a dislocation network. Thus, the average size is slightly reduced and
11 finally stabilized.

12

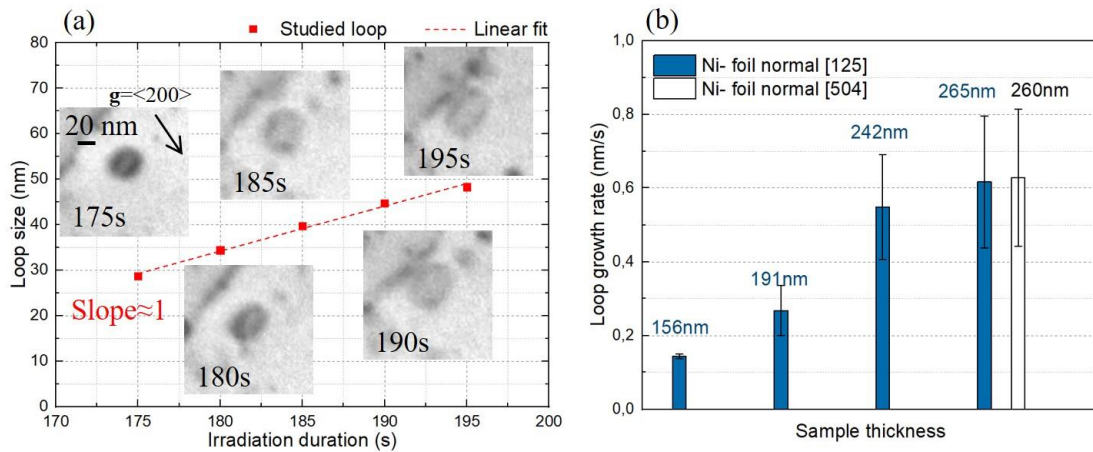
13



1
2 Fig. 2. Microstructural evolution during in-situ irradiation as a function of irradiation
3 thickness and dose at 510°C. (a-c) Selected micrographs illustrating the microstructure
4 recorded under two-beam KBF conditions using (a) $g = \langle 200 \rangle$, (b) and (c) $g = \langle 111 \rangle$;
5 (d-e) Evolution of loop density and average size in function of irradiation time; (f)
6 unfauling of a dislocation loop followed by its slip to surface.

7 Fig. 3(a) presents the typical growth of a Frank loop in function of irradiation time
8 at 510°C. Its growth can be approximately considered as linear with time. A similar
9 phenomenon of interstitial Frank loops was extensively studied under electron irradiation

1 experiments [2,18–20]. This linear growth rate is verified for many other loops, which
 2 allows us to calculate an average growth rate for linearly-growing loops. The results are
 3 indicated in Fig. 3(b). The loop growth rate increases with thickness as observed
 4 intuitively in Fig. 2.



5
 6 **Fig. 3. Determination of loop growth rate during irradiation at 510°C. (a) selected TEM**
 7 **micrographs showing size evolution of a Frank loop in function of time and the**
 8 **corresponding linear fitting; (b) Average loop growth rate in nm/s in function of thickness**
 9 **in two specimens.**

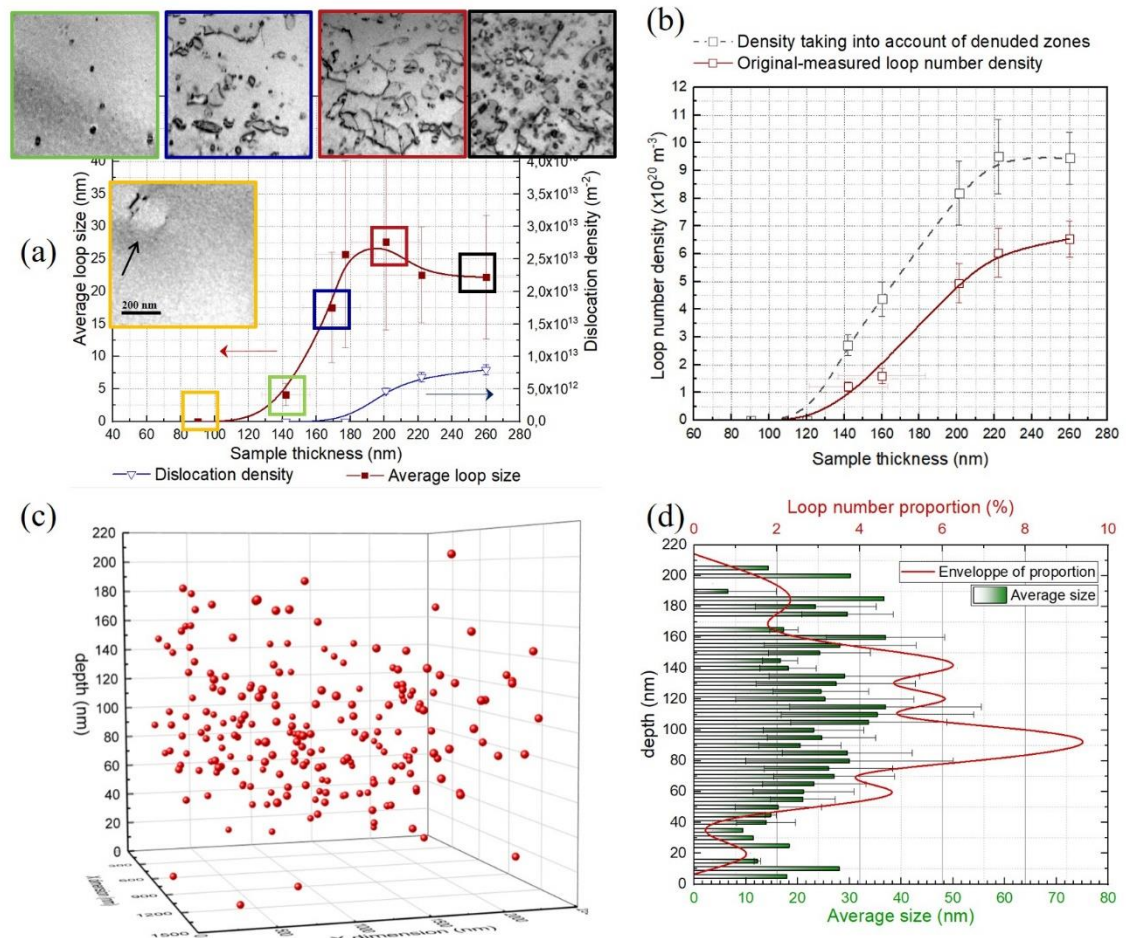
10 It is of great interest to notice that the growth rate is close to saturation taking into
 11 account the larger standard deviation when the thickness exceeds 240 nm. Moreover, the
 12 same growth rate is observed in another foil with different grain orientations (not shown
 13 here). The standard deviation of growth rate is quite important because some loops grow
 14 very fast while some grow slowly even in thick zones. This should be attributed to the
 15 fact that loops are located at different depths inside the foil where the influence of surfaces
 16 changes.

17 **3.3 Post characterization of final microstructure at 510°C (loop average size,**
 18 **density, spatial distribution)**

19 In the following part, we focus on the post-characterization of the final
 20 microstructure irradiated up to 0.06 dpa at 510°C. At this dose, the microstructure is in a

1 steady-state and the dislocation network is moderately developed allowing to study
 2 numerous individual loops and obtain good statistics.

3 Loop average size and dislocation line density in function of thickness are presented
 4 in Fig. 4(a). The density of loops measured directly from images is plotted by the red
 5 curve in Fig. 4(b). In Fig. 4(a), loop average size increases firstly up to a maximum around
 6 190 nm and then decreases and stabilizes to a saturation value. This variation is attributed
 7 to the formation of dislocation lines as analyzed above (Fig. 2). During the irradiation,
 8 the tangle of large loops contributes to the development of a dislocation network. The
 9 formation of such a network reduces the maximum size of loops and therefore their
 10 average size.



11
 12 Fig. 4. Final microstructure in function of foil thickness of Ni irradiated up to 0.06 dpa at
 13 510°C: (a) loop density and (b) average size from TEM micrographs taken with $g=\langle 220 \rangle$.

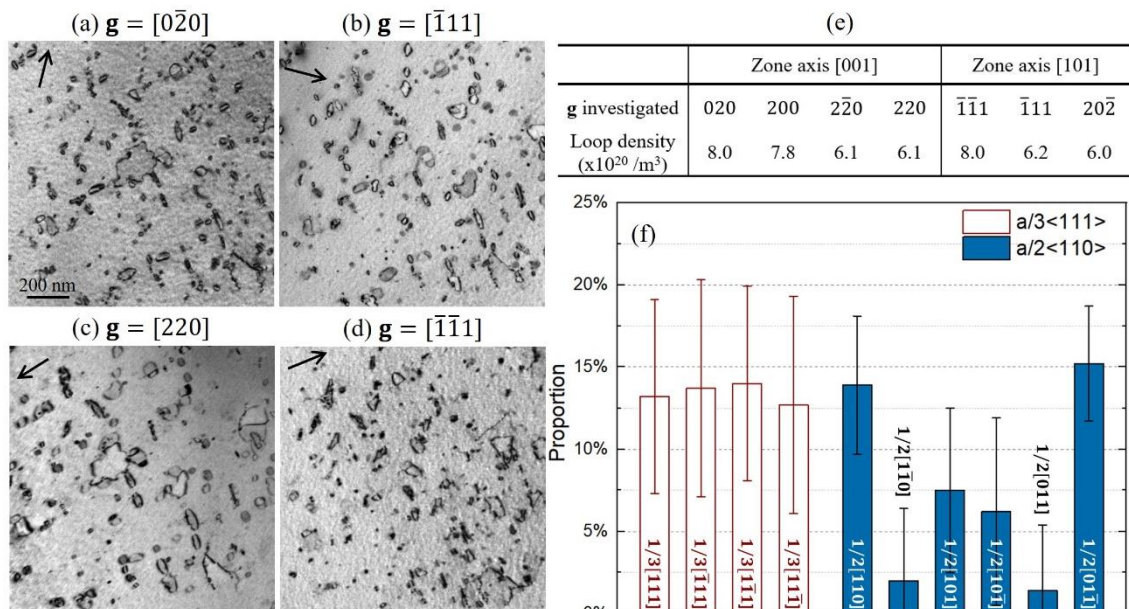
1 Spatial distribution of loops by stereo-imaging technique with (c) 3D visualization of loop
2 position in studied zone of 220 nm thickness and with (d) analysis of loop average size
3 and number proportion v.s. depth, the red curve plotted in Origin using B-spline.

4 However, in Fig. 4(b), the density of loops (red curve) does not show a saturation
5 even up to 260 nm thickness. Since the calculation of loop number density implicates the
6 total thickness of the specimen, homogeneity of loop spatial distribution may be
7 questioned. The localization of loops is studied using the stereo-imaging technique. Fig.
8 4(c) shows the 3D distribution at 220 nm thickness. Fig. 4(d) is the average loop size and
9 loop number proportion along the thickness. The number proportion is calculated by
10 dividing the counted number of loops in each layer along depth direction by the total loop
11 number in the studied zone. Loop-depleted zones near surfaces are observed. The strong
12 absorption of near-surface loops by surfaces is accounted for this depletion. The width of
13 loop-depleted zone thickness is approximately 40 ± 5 nm at each side at 220 nm thickness.
14 We choose 5 nm as uncertainty in this work as it is the error of measurement in the stereo-
15 imaging. Furthermore, at 220 nm thickness, loops in the middle of the foil are slightly
16 larger in terms of average size than those close to surfaces.

17 Taking into account of these loop-depleted zones near surfaces, a corrected loop
18 density using the thickness of loop-located zones is calculated and plotted in the black
19 pointed curve in Fig. 4(b). In this graphical representation, the saturation of loop density
20 is clearly observed. The dependency of these parameters suggests that the final
21 microstructure is thickness-independent above 220 nm. However, it should be noticed
22 that the density shown here takes no account of invisible loops and this is not the real
23 density of existing loops in the sample. Nevertheless, this correction will not change (i)
24 the shape of the black curve in Fig. 4(b) and (ii) the saturation thickness as demonstrated
25 in the next subsection.

1 **3.4 Post characterization of dislocation loops in thick zones (≥ 200 nm) at 510°C**
 2 **(loop type and nature)**

3 In thick zones, Frank loops and perfect loops are both detected. During the
 4 irradiation, the absorption of mobile loops by surfaces is observed, which may lead to the
 5 depletion of some perfect loops. A loop-type analysis by a statistic method [46] in the
 6 200 nm thickness zone is performed to study the surface effects on loop type. In the first
 7 step, microstructure of the same zone is imaged with different \mathbf{g} . For instance, Fig. 5(a-d)
 8 shows four of the seven studied TEM micrographs. According to the authors [46], at least
 9 four \mathbf{g} vectors should be considered (at least two of them non-coplanar) and the accuracy
 10 can statistically-speaking be improved with more \mathbf{g} . In this work, we selected in total
 11 seven different \mathbf{g} , four along the zone axis [001] and three others along [101], which
 12 satisfies completely the exigence and provides good accuracy. Then, loop densities are
 13 calculated from micrographs taking into account loop-depleted zones (excluding 40 nm
 14 for each side). The measured densities are listed in Fig. 5(e).



15 Fig. 5. Loop type analysis by statistic method [46] based on invisibility criterion [41] in
 16 Ni irradiated to 0.06 dpa at 510°C: (a-d) TEM micrographs of the same zone showing
 17 visibility of loops in function of diffraction vectors \mathbf{g} ; (f) loop number density under seven
 18

1 different \mathbf{g} used for statistic analysis; (e) Proportion of each family. Thickness of the zone
 2 is around 200 nm and the foil normal is close to [125].

3 The next step is to establish the system of equations to calculate the density of the
 4 aimed loop family. In fcc structure, there are four families of Frank loops and six families
 5 of perfect loops. Individual analyses for each family are conducted similarly. The method
 6 is illustrated in Equation (1) by the case of the Frank loop family with $\mathbf{b}=1/3[111]$. Based
 7 on the invisibility criteria, the system of equations for achieved diffraction conditions is:

$$8 \quad \begin{cases} N_{\mathbf{g}=020}^{visible} = \frac{1}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{3}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{4}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=200}^{visible} = \frac{1}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{3}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{4}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=2\bar{2}0}^{visible} = \frac{0}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{2}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{5}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=220}^{visible} = \frac{1}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{1}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{5}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=\bar{1}11}^{visible} = \frac{1}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{3}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{3}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=\bar{1}\bar{1}1}^{visible} = \frac{1}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{3}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{3}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \\ N_{\mathbf{g}=20\bar{2}}^{visible} = \frac{0}{1}N_{\mathbf{b}=\frac{1}{3}[111]} + \frac{2}{3}N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} + \frac{5}{6}N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \end{cases} \quad \text{Equation (1)}$$

9 Where $N_{\mathbf{g}}^{visible}$ is the density of visible loops for the used \mathbf{g} and summarized in Fig. 5(e),
 10 $N_{\mathbf{b}=\frac{1}{3}[111]}$ is the density of $1/3[111]$ Frank loops, $N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]}$ is the sum of three
 11 other families of Frank loops, $N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle}$ is the sum of six families of perfect loops. This
 12 system can be written in the following way:

$$13 \quad \begin{pmatrix} N_{\mathbf{g}=020}^{visible} \\ N_{\mathbf{g}=200}^{visible} \\ N_{\mathbf{g}=220}^{visible} \\ N_{\mathbf{g}=2\bar{2}0}^{visible} \\ N_{\mathbf{g}=\bar{1}11}^{visible} \\ N_{\mathbf{g}=\bar{1}\bar{1}1}^{visible} \\ N_{\mathbf{g}=20\bar{2}}^{visible} \end{pmatrix} = \begin{pmatrix} 8.0 \\ 7.8 \\ 6.1 \\ 6.1 \\ 8.0 \\ 6.2 \\ 6.0 \end{pmatrix} \times 10^{20} = \begin{pmatrix} 1 & 1 & 2/3 \\ 1 & 1 & 2/3 \\ 0 & 2/3 & 5/6 \\ 1 & 1/3 & 5/6 \\ 1 & 1 & 1/2 \\ 1 & 1 & 1/2 \\ 0 & 2/3 & 5/6 \end{pmatrix} \begin{pmatrix} N_{\mathbf{b}=\frac{1}{3}[111]} \\ N_{\mathbf{b}=\frac{1}{3}\langle 111 \rangle / \frac{1}{3}[111]} \\ N_{\mathbf{b}=\frac{1}{2}\langle 110 \rangle} \end{pmatrix} \quad \text{Equation (2)}$$

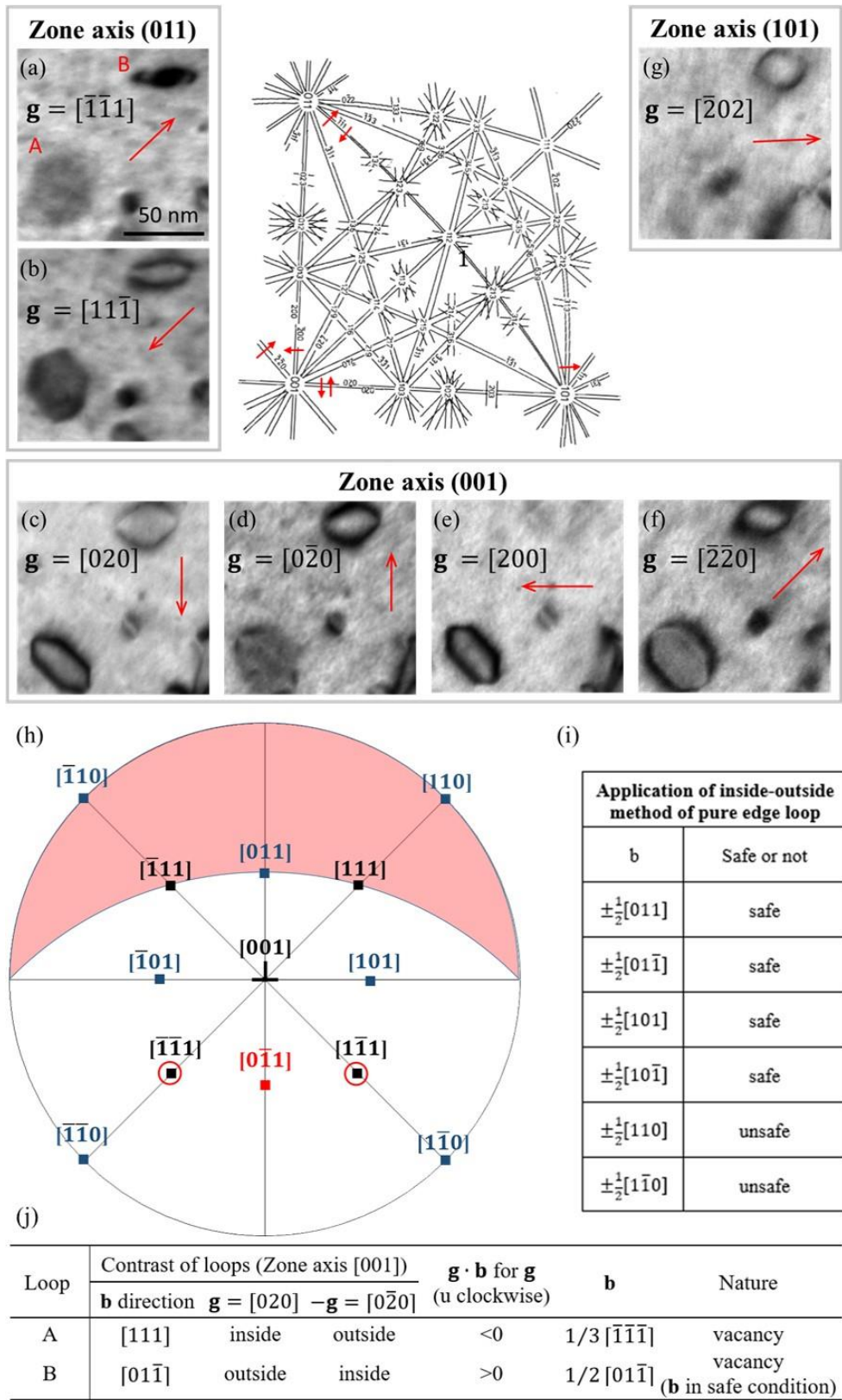
14 This equation is solved by the least-squares method with an error given by the standard
 15 deviation from the least-squares. The solution gives the density of three families in Eq.

1 (2), for instance, $N_{\mathbf{b}=\frac{1}{3}[111]} = 12 \pm 5 \times 10^{19} \text{ m}^{-3}$. By the same analysis, we calculate the
 2 density of other families. Finally, the density of all Frank loop families and perfect loop
 3 families is deduced by summing results of each family and are respectively $N_{\text{Frank}} = 50 \pm$
 4 $11 \times 10^{20} \text{ m}^{-3}$ and $N_{\text{perfect}} = 43 \pm 12 \times 10^{20} \text{ m}^{-3}$. Thus the total density taking into account
 5 invisible loops is $N_{\text{total}} = 91 \pm 16 \times 10^{20} \text{ m}^{-3}$. We obtain the proportion of each family by
 6 dividing the density of the considered family over the total density. The results are
 7 presented in Fig. 5(a). Frank loops are equally distributed among the 4 families with a
 8 total density of $50 \pm 10 \times 10^{20} \text{ m}^{-3}$. The proportion of each Frank loop family is around 13
 9 $\pm 1\%$ over all loops. On the contrary, a heterogeneous distribution of perfect loops is
 10 clearly shown. The family of perfect loops with $\mathbf{b}=\frac{1}{2}[011]$ shows the lowest proportion
 11 and is almost completely depleted while perfect loops with $\mathbf{b}=\frac{1}{2}[01\bar{1}]$ has the highest
 12 proportion, which may be related to the surface absorption and will be discussed in the
 13 following section.

14 With the Burgers vector analysis, a “surface-free” density of loops in the sample
 15 (without surface elimination) can be estimated. If we consider that loops are formed
 16 equally at the beginning and that the depletion of perfect loops results only from their
 17 movement to surfaces, the “surface-free” total density of perfect loops can be estimated
 18 by multiplying the highest density by six. The corrected density of perfect loops is $85 \pm$
 19 $20 \times 10^{20} \text{ m}^{-3}$. Thus the “surface-free” density of loops taking into account of escaped
 20 loops at 200 nm thickness is $N_{\text{total, corrected}} = 135 \pm 22 \times 10^{20} \text{ m}^{-3}$. We can also calculate the
 21 corrected proportion of Frank loops and perfect loops which is 37% Frank loops and 63%
 22 perfect loops. If the loop type analysis is conducted for each thickness, the curve of loop
 23 density in Fig. 4(b) can be again corrected. However, we tend to assume that the
 24 distribution of Burgers vectors is the same for the studied range of thickness (150-260

1 nm). With such an assumption, this correction will not change the profile of the curve. So
2 a new curve is not plotted.

3 In our previous work [33], the relation of the morphology of a Frank loop and its
4 nature was highlighted. For the first time, large Frank loops in Ni irradiated at 450°C were
5 found to be segmented and of vacancy nature. In this study, large segmented loops are
6 observed at 510°C so they must be vacancy-type. The nature of individual loops is
7 determined manually using the inside-outside method [40,47,48] in thick zones (thickness
8 ≥ 200 nm). For tens of characterized loops in each zone, the same vacancy nature for both
9 Frank loops and perfect loops was identified independently on their Burgers vectors. We
10 present here a detailed analysis of two representative loops as shown in Fig. 6.



1

2 Fig. 6. Determination of Burgers vector and nature of two representative loops in Ni
 3 irradiated to 0.06 dpa at 510°C. (a-g) TEM micrographs for analysis of Burgers vectors;
 4 Kikuchi map extracted from [40]; (h-i) Stereo-projection along zone axis [001] for

1 analysis of safe/unsafe conditions [48] for perfect loops $1/2[0\bar{1}1]$, plotted using CaRIne
2 software; (j) determination of loop nature by the inside-outside method [47,48].

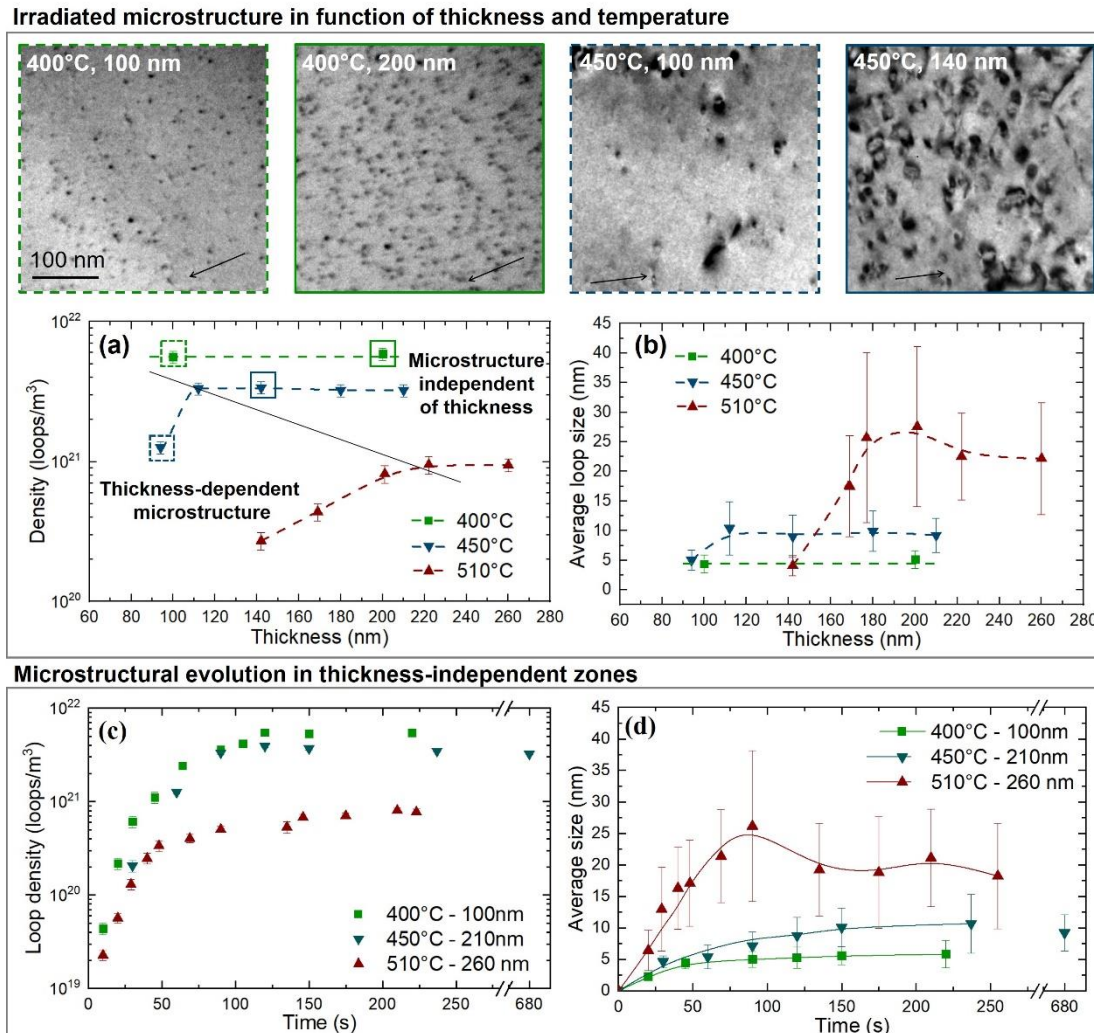
3 Fig. 6(a-g) allow the determination of Burgers vector of loop A and loop B,
4 resulting in $\mathbf{b}=\pm 1/3[111]$ for loop A and $\mathbf{b}=1/2[01\bar{1}]$ for loop B. Detailed analysis of \mathbf{b} is
5 listed in Appendix B (Table B. 1). Since A is a Frank loop so a pure edge loop, the inside-
6 outside method can be directly applied to determine its nature. Using the FS/RH
7 convention [50] by considering the sense of the dislocation line as clockwise, the inside-
8 outside behavior shown in Fig. 6(c-d) leads to $\mathbf{b}=1/3[\bar{1}\bar{1}\bar{1}]$ the vacancy nature as shown
9 in Fig. 6(j). The same nature can be deduced from another inside-outside pair in Fig. 6(a-
10 b) as well. For the perfect loop B, we should determine whether it is under safe conditions
11 before applying the inside-outside method [48], as illustrated in Fig. 6(i). Loop B can only
12 be formed from the unfauling of $1/3[\bar{1}\bar{1}1]$ or $1/3[1\bar{1}1]$ Frank loops by Shockley partials
13 with $\mathbf{b}=1/6\langle 112 \rangle$. The stereo-projection in Fig. 6(h) shows that these two families of
14 Frank loops are both out of red zones thus in safe conditions according to Fig. 6(i).
15 Therefore, the inside-outside behavior of loop B together with the analysis in Fig. 6(h)
16 confirm its vacancy nature ($\mathbf{b}=1/2[01\bar{1}]$).

17 **3.5 Role of temperature on surface effects (critical thickness of saturated** 18 **microstructure) and defect evolution**

19 The temperature-dependency of surface effects shown in Fig. 1 is investigated in
20 detail. Fig. 7(a-b) presents the thickness-dependency of loop number density and average
21 size respectively at 400, 450 and 510°C. The curves of 510°C has been presented in Fig.
22 3(a) and plotted here to compare with the lower temperatures. Microstructures at 400 and
23 450°C are shown by TEM micrographs in Fig. 7. At 400°C, loop number density and size
24 are the same at thicknesses of 100 nm and 200 nm. It suggests that saturation of
25 microstructure starts from a thickness lower than 100 nm. At 450°C, the two parameters

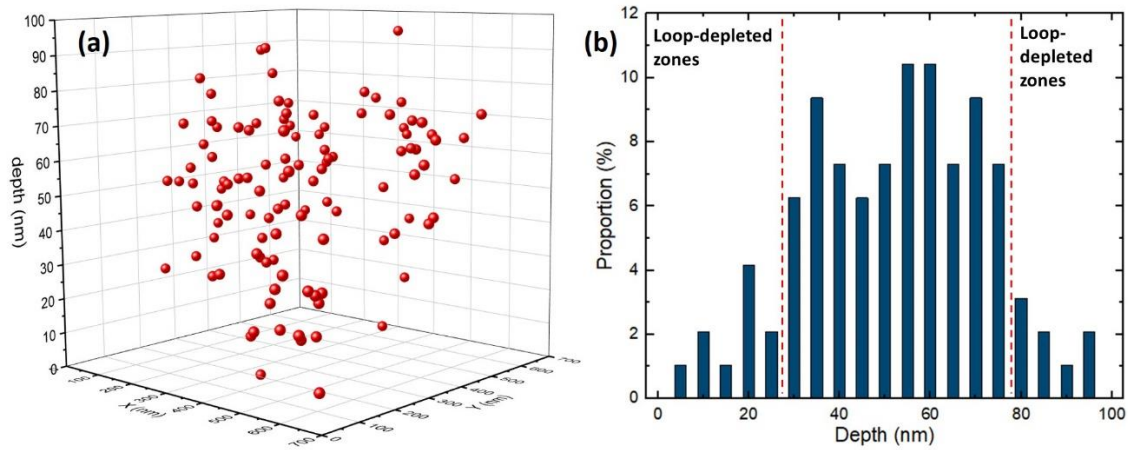
1 are lower at 80 nm than at 180 nm and 210 nm. When the thickness exceeds 180 nm, the
2 microstructure is certainly thickness-independent. Based on these observations, a black
3 line is drawn in Fig. 7(a) dividing approximately thickness-dependent zones and
4 thickness-independent zones. In thickness-independent zones, the difference of
5 microstructure is only caused by temperature without any influence of thickness.
6 Microstructural evolutions in saturated zones are recorded at each temperature, as shown
7 in Fig. 7(c-d). The number density at 400°C reaches a steady-state after 120 seconds of
8 irradiation while the average size still increases slowly with time. At 450°C, the steady-
9 state of number density is achieved slightly later than at 400°C. The average size
10 continuously increases with time after the steady state of loop density. However, this
11 steady state of loop density should be “temporary”. Loop density will finally decrease
12 due to the development of a dislocation network. The irradiation at 450°C is conducted
13 for a high dose in the same foil until 680 seconds (equivalent to 0.18 dpa). This tendency

- 1 is shown by the two points at $t=680$ s at 450°C in Fig. 7(c-d) where loop size and density
- 2 are both lower than the values at the so-called steady state.



3
4 Fig. 7. Influence of surface effects in function of temperature. Evolution of (a) Loop
5 density and (b) average loop size in function of thickness at 400/450/510°C with selected
6 TEM micrographs showing corresponding microstructure; evolution of (c) loop density
7 and (d) average size in function of time in thickness-independent zones at each
8 temperature with selected TEM micrographs showing the real-time microstructural
9 evolution.

10 The stereo-analysis was also performed at 450°C to study the influence of temperature on
11 the width of loop-depleted zones. Fig. 8 shows the loop-depleted zones in Ni irradiated at
12 450°C for a 100 nm thickness. Above 100 nm, loop density is too high to correctly
13 identify loops before and after tilting. The width of the zone is about 25 ± 5 nm at each
14 side of the foil.



1
 2 Fig. 8. Loop depth distribution in Ni irradiated at 450°C. (a) 3D visualization of loop
 3 position in studied zone of 100 nm thickness; (b) loop number proportion in function of
 4 depth by stereo-imaging in Ni irradiated at 450°C showing loop-depleted zones in each
 5 side of the foil.

6
 7 **4 Discussion**

8 **4.1 Absorption of point defects by surfaces**

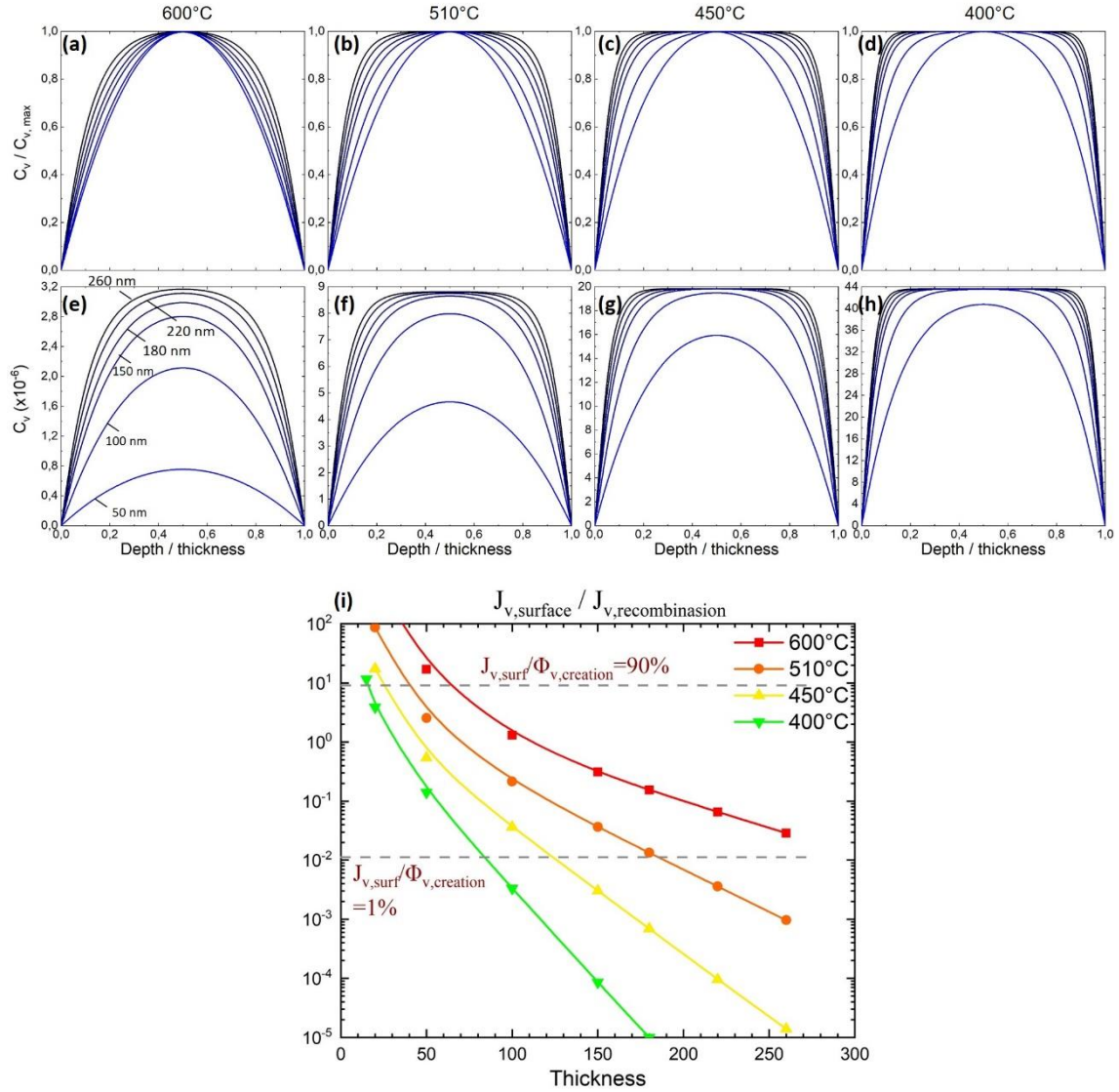
9 To evaluate surface effects, the absorption of point defects by surfaces should be
 10 considered. We consider here a system without any internal sinks (dislocation lines and
 11 loops) to maximize the influence of surfaces. Profiles of the vacancy concentration are
 12 calculated numerically by a one-dimensional steady-state rate diffusion equation based
 13 on [51] (see details in Appendix C) for the thickness of 50/100/150/180/220/260 nm at
 14 400/450/500/600°C. Based on this numerical solution, we can calculate the normalized
 15 profile (Y/Y_{\max} for each thickness) and the profile of the concentration of vacancies ($C_v =$
 16 $YC_{v,\infty}$). The results are presented in Fig. 9(a-h).

17 For a given temperature, for instance 510°C (see Fig. 9(b) and Fig. 9(f)), the
 18 profile expands from a typical surface-dominated one to a recombination-dominated one
 19 with increasing thickness. This could explain the thickness-dependency of the irradiated
 20 microstructure shown in Fig. 4(a) and Fig. 4(b). At 510°C, a recombination-dominated
 21 zone occurs in the center of the foil when $t=150$ nm. It corresponds to the beginning of

1 loop growth in thick zones experimentally observed. Obviously, the fraction of volume
2 which is less affected by surface increases with thickness. Finally, in thick zones (220
3 nm), the vacancy concentration is stabilized and reaches a thickness-independent stable
4 value, which could lead to the observed thickness-independent microstructure. The same
5 tendency is shown at lower temperatures. However, surface effects remain strong at
6 600°C in the studied thickness range. When the temperature decreases, the profile
7 expands more quickly with increasing thickness from surface-dominated profiles to
8 recombination ones. This could be related to the temperature dependency of critical
9 thickness for saturated microstructure.

10 For further quantitative evaluation of the absorption by surface, the ratio of
11 vacancy flux to surface $J_{v,surf}$ and to the recombination $J_{v,recomb}$ is calculated and shown
12 in Fig. 9(i). The lower dashed line corresponds to a ratio of 0.01 i.e. approximately 1% of
13 PDs are absorbed by surfaces. The corresponding thicknesses for each temperature are
14 80/120/190 nm respectively for 400/450/510°C. Considering the simplicity of the model
15 without effects of internal sinks (radiation-induced loops during the irradiation and
16 thermal vacancies as recombination site [51]), this estimation is fairly in good agreement
17 with our experiment observations (80/110/220 nm) in Fig. 7(a). The upper dashed line in
18 Fig. 9(i) corresponds to 90% absorption of PDs by surfaces. It is worth noting that the
19 results in Fig. 9(i) is for the very center of the foil, where the surface absorption is the
20 lowest across the entire depth. The surface absorption in the rest volume is higher than
21 90%. Thus, it suggests a critical thickness under which the sample will be completely free
22 of defects. It reads respectively 18/25/40/70 nm for 400/450/510/600°C. This is in fairly
23 good agreement with our observations at 510°C (under 40 nm thickness, no loop is
24 observed) and with those reported by other authors at lower temperatures (~20 nm at

1 400°C in Ni under Ar⁺ irradiation [1]). However, the critical thickness for the defect-free
 2 zone is usually larger than the calculated one because other factors should be taken into
 3 account, such as absorption of formed defects by surfaces.



4
 5 Fig. 9 Profiles of (a-d) normalized and real (e-f) vacancy concentrations and (i) ratio of
 6 vacancy flux towards surfaces and to recombination in the center of the foil in function
 7 of thickness (50/100/150/180/220/260 nm) and temperature: concentration profiles at (a,e)
 8 600°C; (b,d) 510°C; (c,g) 450°C; (d,h) 400°C; in (i), the upper dashed line corresponding
 9 to 90% absorption of PDs by surfaces and lower dashed line corresponding to 1%
 10 absorption by surfaces.

1 4.2 Surface denuded zones

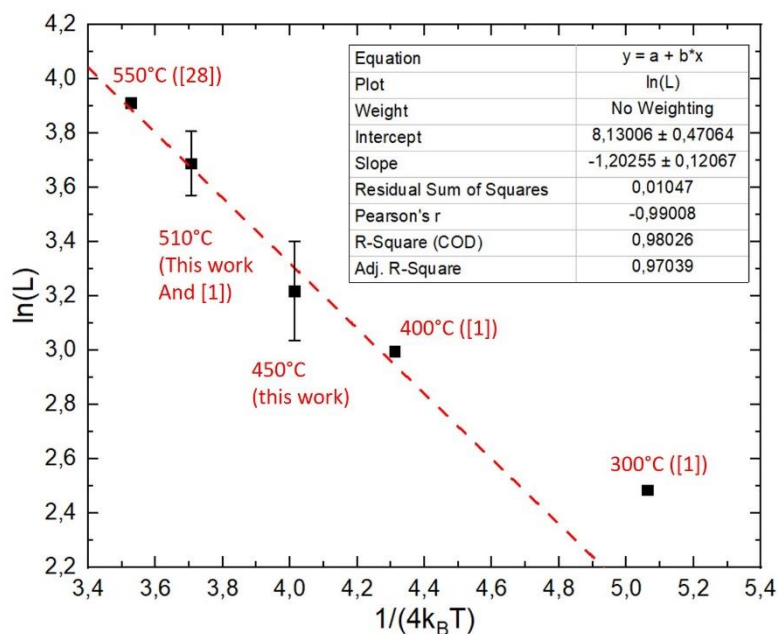
2 The depth distribution of radiation-induced defects in thin foils has been
3 investigated in several materials [1,4,19,38,52,53] to better describe irradiated
4 microstructures and understand the mechanism of the defect formation. In the literature,
5 the thickness of depleted zones was found to be about 12 nm at 300°C and 20 nm at 400°C
6 in Ni under Ar⁺ irradiation [1] for each side of the foil. Another experiment showed that
7 no loops were detected at the irradiation entry side until 50 nm in Ni irradiated by 25 keV
8 He⁺ at 550°C [29]. In electron-irradiated Ni, the depth profile of the interstitial loop size
9 was studied in a 450 nm thick foil at 450°C [19]. All studied loops were located at least
10 30 nm from the closest surface. In the present work, a loop-depleted zone is indeed shown
11 in Fig. 4(d) and is roughly 40 nm wide from each foil surface in self-ion irradiated Ni at
12 510°C. Although the amount of data is limited and irradiation conditions are different,
13 these data at least are in reasonable agreement and suggest that the width of surface
14 denuded zones depend on temperature. The width of surface denuded zones L was
15 described by a formula of Foreman [51]:

$$16 \quad L = \sqrt{\frac{D_v C_{v,\infty}}{G}} = \left(\frac{D_v}{G K_r}\right)^{\frac{1}{4}} \quad \text{Equation (3)}$$

17 where G the PD production rate and K_r the recombination coefficient are constant. L is
18 thus proportional to $D_v^{\frac{1}{4}}$ so proportional to $\exp(-E_{v,m}/(4k_B T))$. Experimental results of
19 $\ln(L)$ ($L=12/20/25/40/50$ nm) in function of $(4k_B T)^{-1}$ for 400/450/510/550°C are plotted
20 in Fig. 10. Error bars are plotted only for our experiment data since the uncertainty of L
21 is not given in the literature. Data at $T \geq 400^\circ\text{C}$ leads to a fairly good linear fitting
22 considering the error bar. The fitted slope reads 1.2 ± 0.1 eV, which gives a good

1 estimation of migration energy of vacancy from self-diffusion experiments (1.24 eV [17])
2 and by some simulations (1.32 eV [15], 0.92-1.46 eV [54]) in the literature.

3 Yet, the data at 300°C deviates from the fitting line. There are three possible
4 explanations. The first explanation is related to the uncertainty of the loop-denuded
5 thickness determination. The uncertainty and the error bar increase significantly at lower
6 temperatures. The fact that error bars were not given at 300°C and 400°C in [1] and at
7 550°C in [29] could induce a slight discrepancy of the plotted slope. The second reason
8 might be a potential influence of injected gaseous impurities (Ar^+ irradiations at
9 300/400°C in [1]) on the mobility of vacancies since it is known that impurities can impact
10 the migration of PDs [55]. The third one is the applicability of Equation (3) at low
11 temperatures. At $T < 400^\circ\text{C}$, the nature of loops nucleated at 300°C could change. Foreman
12 suggested that the denuded zone of interstitial defects should be a measure of the
13 interstitial mobility instead of the vacancy mobility [51]. Therefore, the correlation
14 between $\ln(L)$ and $(4k_B T)^{-1}$ might no longer be related to the diffusion coefficient of
15 vacancies but the one of interstitials. This will lead to a much smaller slope ($E_m^i \sim 0.15\text{eV}$
16 [16,17]), which consists with the reduction of slope towards lower temperatures shown
17 in Fig. 10.



1
2 Fig. 10. Correlation between the width of loop-depleted zones and temperatures. Linear
3 fitting is performed using the data at 550/510/450°C. The error bar of data in this work
4 results from the uncertainty of the 5 nm thickness of depleted zone.

5 4.3 Surface effects on loop type

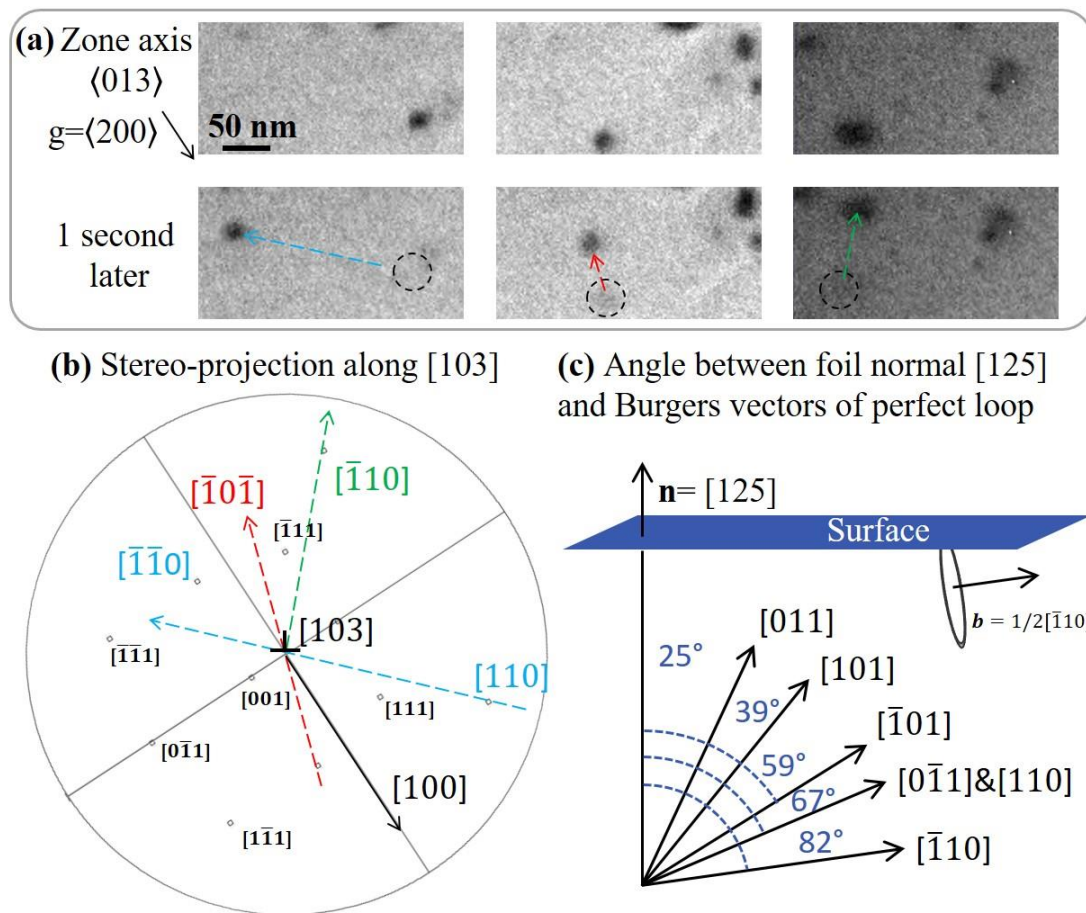
6 In Fig. 5(e), unequal distribution of Burgers vectors of perfect loops is observed
7 in irradiated Ni. To explain this phenomenon, two mechanisms should be considered: (i)
8 a preferential absorption of some loops by surfaces and (ii) a preferential formation of
9 perfect loops with some specific Burgers vectors.

10 The first one is very likely involved since many loops are indeed found to move
11 straight and then disappear probably at surfaces as shown in Fig. 2(f). Fig. 11(a) shows
12 the straight movement of three additional loops at 510°C. These TEM micrographs were
13 taken along zone axis $\langle 013 \rangle$ with $\mathbf{g} = \langle 200 \rangle$ during the irradiation. This quick straight
14 movement is very likely the slip. To analyze the direction of their movement, stereo-
15 projection along $[013]$ is shown in Fig. 11(b). The motion takes place always along $\langle 110 \rangle$
16 directions. It is in agreement with the fact that slip of perfect dislocation in fcc materials
17 occurs on $\{111\}$ planes in $\langle 110 \rangle$ directions [56].

1 It is interesting to wonder whether loops slip along the direction of their Burgers
 2 vector. It is difficult to analyze the Burgers vector and their habit plane during the
 3 irradiation. Thus, a post-characterization is performed in irradiated Ni (510°C/0.06 dpa)
 4 to study the Burgers vector and habit plane of loops. Fig. 12 shows a typical rhombus-
 5 shape perfect loop (no fault contrast inside the loop). Its Burgers vector is $1/2[01\bar{1}]$ (Fig.
 6 12(b-c)). The two segments of the rhombus (D1 and D2) are $[321]$ and $[\bar{3}21]$. The loop
 7 plane normal \mathbf{n}_{loop} is obtained by $\mathbf{n}_{\text{loop}}=[321]\times[\bar{3}21]=[0\bar{1}2]$ (conventionally \mathbf{n}_{loop} pointing
 8 always upward). Perfect loops with similar segment directions and loop planes were
 9 detected in Al-based alloys in the literature [57]. Fig. 12(d) shows schematically the loop
 10 structure. The two segments $[321]$ and $[\bar{3}21]$ are respectively in $(\bar{1}11)$ and (111) planes
 11 which are slip planes in fcc structure. The common slip direction on these planes is $[01\bar{1}]$
 12 which is collinear with the Burgers vector of the loop. Moreover, the exact habit plane
 13 and the Burgers vector give an unambiguous determination of its nature which is vacancy-
 14 type since $\mathbf{n}_{\text{loop}} \cdot \mathbf{b} < 0$ [48].

15 Here we tend to assume that the motion of perfect loops follows their Burgers vector.
 16 To understand the unequal distribution of perfect loops in post-characterization (Fig. 2(f)),
 17 the foil normal and different Burgers vectors of perfect loops are plotted in Fig. 11(c).
 18 The angle between the foil normal and each Burgers vector is shown. The $[011]$ direction
 19 has the smallest angle with the foil normal while $[01\bar{1}]$ and $[110]$ are far from this
 20 direction. We could expect that loops having shorter path lengths along the motion
 21 direction to the foil surface are more readily to be lost than loops of other Burgers vectors.
 22 This is in agreement with the experimental observations of most families except $1/2[\bar{1}10]$
 23 perfect loops. It may be understood geometrically as the $[\bar{1}10]$ direction is almost parallel
 24 to the surface (see Fig. 11(c)). If perfect loops tend to rotate their loop habit plane to

1 become edge loops i.e. Burgers vector perpendicular to loop plane, the habit plane of
 2 $1/2[\bar{1}10]$ loops will be almost perpendicular to the surface. The interception of $1/2[\bar{1}10]$
 3 loops with surfaces is the geometrically easiest. The interception results in the absorption
 4 of loops by surfaces and finally leads to a depletion of this family. It is worth noting that
 5 such an influence of surfaces remains strong at such a thick zone where the absorption of
 6 PDs by surfaces is moderate. It shows that surface effects are significant not only on the
 7 scale of primary defects (like PDs) but also on the scale of larger defects (like dislocation
 8 loops).



9
 10 Fig. 11. Analysis of motion direction of dislocation loops in irradiated Ni at 510°C. (a)
 11 TEM micrographs showing the movement of three loops with the motion direction
 12 indicated by dashed arrows; images taken along zone axis $\langle 013 \rangle$ with $g = \langle 200 \rangle$; (b)
 13 stereo-projection along $[103]$ with motion directions indicated; (c) angles between the
 14 foil normal $[125]$ and Burgers vectors of perfect loops.

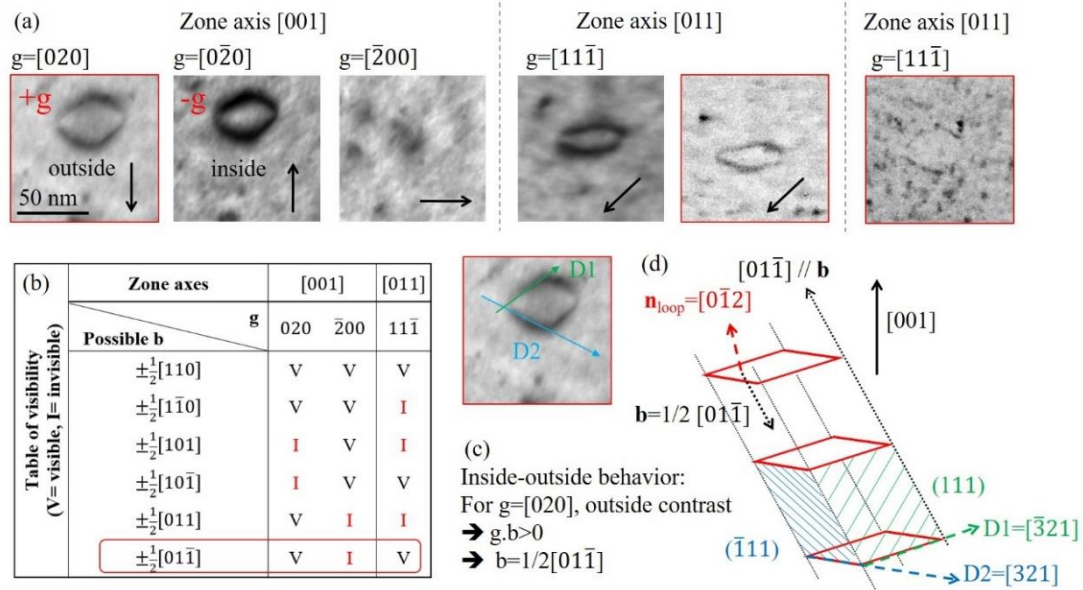


Fig. 12 Analysis of Burgers vector and habit plane of a rhombus perfect loop in Ni irradiated at 510°C up to 0.06 dpa. (a) TEM micrographs showing the visibility of the loop with the same scale bar in the first one. (b) Table of visibility showing the Burgers vector. (c) Inside-outside contrast analysis determining the sign of the Burgers vector. (d) Directions of line segments and the habit plane of the rhombus loop; the segment directions are determined from TEM micrographs with red frame.

4.4 Growth of vacancy loops in Ni at high temperatures

This work shows that dislocation loops formed in Ni at 510°C under irradiation are vacancy-type and segmented. This observation is in perfect agreement with our previous study that showed for the first time a strong correlation between the morphology of Frank loops and their nature: interstitial Frank loops are non-segmented while vacancy ones are segmented [33]. During the in-situ experiments at 510°C, Frank loops (such as the one in Fig. 2(f)) are segmented. Therefore, the dislocation loops that grow under irradiation (Fig. 2(a-c)) should be vacancy-type. Our characterization show a great application of the loop morphology-nature correlation that, during complicated in-situ experiments, the nature of dislocation loops can be easily determined based on their form.

However, it is widely accepted that, in Ni, only interstitial loops can grow into a large size under irradiation while vacancy loops would rather shrink due to the positive

1 bias of dislocation loops for both natures [58–60]. The origin and the thickness-
2 dependency of the growth of vacancy loops in our studies and the previous work [33] has
3 not been theoretically understood. Conventional rate theory based on the dislocation bias
4 model seems insufficient to explain the observed growth of vacancy loops. Conventional
5 rate theory implies that all produced point defects are free and are available for
6 annihilation at sinks. As far as we are concerned, a production bias may be a key point to
7 be taken into account. Woo and Singh suggested a production bias model in which
8 vacancies and interstitials produced in cascades are not equally accessible to different
9 sinks (dislocations and voids) [59]. Here we establish a simple system of rate equations
10 assuming that a fraction $(1 - \eta)$ of interstitials form I-clusters within the cascade, so are
11 unavailable to loops while all vacancies remain single PDs :

$$12 \quad \frac{dC_v}{dt} = G - K_r(D_i + D_v)C_iC_v - k_v^2D_vC_v \quad \text{Equation (4)}$$

$$13 \quad \frac{dC_i}{dt} = \eta G - K_r(D_i + D_v)C_iC_v - k_i^2D_iC_i \quad \text{Equation (5)}$$

14 where, G is the creation rate by irradiation, $K_r = 4\pi r_c/\Omega$ is the recombination coefficient
15 deduced from Waite's theory [61] with r_c the recombination radius assumed to be the
16 lattice parameter (0.352 nm) and Ω the volume of Ni atom, and k_α^2 is the sink strength for
17 species α . The sink strength is the sum over the different objects (dislocation lines, loops,
18 surfaces) in microstructure. At steady state, we can deduce from Eqs. (4) and (5):

$$19 \quad k_i^2D_iC_i = k_v^2D_vC_v - G + \eta G \quad \text{Equation (6)}$$

20 The growth rate of a vacancy loop reads:

$$21 \quad v = \frac{\Omega}{b} (Z_{l,v}D_vC_v - Z_{l,i}D_iC_i) \quad \text{Equation (7)}$$

22 Using Eqs. (D3), Equation (6) is written as:

$$23 \quad v = \frac{\Omega}{b} \left(Z_{l,v} - Z_{l,i} \frac{k_v^2}{k_i^2} \right) D_vC_v + \frac{\Omega Z_{l,i}}{b k_i^2} G(1 - \eta) \quad \text{Equation (8)}$$

1 To enable the growth of vacancy loops, it is required that $v > 0$, thus we deduce
 2 based on Equation (8) that :

$$3 \quad 1 - \eta > - \frac{D_v C_v}{G} \left(k_i^2 \frac{Z_{l,v}}{Z_{l,i}} - k_v^2 \right) \quad \text{Equation (9)}$$

4 We assume that surfaces are neutral sink for PDs ($Z_{surf,i} = Z_{surf,v} = Z_s$) and the bias
 5 factor is the same for dislocation lines and loops ($Z_{line,v} = Z_{loop,v} = Z_v$, $Z_{line,i} = Z_{loop,i} = Z_i$). Thus,
 6 the sink strength is written as:

$$7 \quad k_v^2 = k_{surf,v}^2 + k_{loop,v}^2 + k_{line,v}^2 = Z_s k_{surf}^2 + Z_{l,v} k_{loop}^2 + Z_{l,v} k_{line}^2$$

$$8 \quad k_i^2 \frac{Z_v}{Z_i} = (Z_s k_{surf}^2 + Z_{l,i} k_{loop}^2 + Z_{l,i} k_{line}^2) \frac{Z_v}{Z_i} = \frac{Z_{l,v} - Z_{l,i}}{Z_{l,i}} k_{surf}^2 + k_v^2 = -B_l k_{surf}^2 + k_v^2$$

9 Where $k_{surf,v}^2$, $k_{loop,v}^2$ and $k_{line,v}^2$ are sink strength depending only on the geometry and
 10 density/size of the sink. Finally, Eqs. (D6) is simplified as:

$$11 \quad 1 - \eta > \frac{D_v C_v}{G} B_l k_{surf}^2 \quad \text{Equation (10)}$$

12 Where D_v is the diffusion coefficient of vacancies, C_v the concentration of vacancies, G
 13 production rate of PDs, $B_l = (Z_i - Z_v)/Z_i$ the loop bias factor and k_{surf}^2 the sink
 14 strength of free surfaces. At 510°C in a 260 nm thick zone, $D_v = 4.6 \times 10^{-14}$ m²/s deduced
 15 from [17], $C_v = 8 \times 10^{-6}$, $G = 2.7 \times 10^{-4}$ dpa/s, $B_l = 0.4$ typically for a loop with a radius of 2
 16 nm (Fig. 3.16 in [60]), $k_{surf}^2 = 12/t^2$ where t is the thickness [62], we finally deduce
 17 that $1 - \eta > 12\%$. These calculations show that if at least ~10% interstitials are “lost”,
 18 the growth of vacancy loops could be favored. A plausible explanation for this loss of
 19 interstitial clusters is that they move in long-distance 1D motion and escape immediately
 20 from the production zone, as suggested by [15]. Equation (8) shows that the loop growth
 21 rate depends on the sink strength of surfaces therefore the thickness, which is observed
 22 in this work.

23 **5. Conclusions**

1 The influence of free surfaces on radiation damage in pure nickel is investigated
2 using in-situ irradiations within the JANNUS-Orsay platform [39] at different
3 temperatures and thin foil thicknesses. Our observations show that:

4 1) A drastic influence of thickness on the in-situ microstructural evolution and
5 post-irradiated microstructure of Ni in the studied temperature range (400-
6 700°C) is highlighted. This so-called surface effects on loop density and size
7 are firstly attributed to the absorption of point defects by surfaces. Our work
8 clearly demonstrates the existence of a critical thickness, depending on
9 temperature, above which the loop growth rate, loop density and average loop
10 size become independent on the thickness. The critical thickness for
11 510/450/400°C are respectively 220/100/80 nm.

12 2) Our vacancy concentration calculations in Ni (based on Foreman formulations
13 [51]) could be an excellent tool to predict an adequate thickness for in-situ
14 irradiations in various materials. They confirm the thickness dependency of the
15 microstructure and the calculated critical thicknesses are in good agreement
16 with those determined in our work.

17 3) Free surfaces affect the fine distribution of loop Burgers vectors even above the
18 critical thickness. Some perfect loops families with a Burgers vector of
19 $\mathbf{b}=1/2\langle 110 \rangle$ preferentially glide to free surfaces and are absorbed, leading to a
20 heterogeneous distribution of glissile perfect loops. This preferential absorption
21 of perfect loops is related to the orientation of the Burgers vector of the loop
22 and the thin foil normal. The family with the Burgers vectors closer to the foil
23 normal is the most depleted. On the contrary, the distribution of Frank loops,

1 sessile defects with $\mathbf{b}=1/3\langle 111 \rangle$, is not affected by free surfaces: they are
2 equally distributed among the four families.

3 4) For the first time, the migration energy of vacancies in Ni is experimentally
4 determined considering the temperature dependence of the loop-depleted zones.
5 The loop-depleted zones on each side of thin foils are indeed dependent of the
6 irradiation temperature. Their width are about 40 nm at 510°C and 30 nm at
7 450°C. The fit of our experiment data on the loop depleted zone with the ones
8 extracted from the literature gives a migration energy of 1.20 eV, which is in
9 good agreement with the recognized values in the literature [15,17,54].

10 5) A rate theory model including production bias term demonstrates that a loss of
11 10% interstitials, at 510°C, theoretically favors the growth of vacancy loops. It
12 well explains the first observation at 510°C of grown Frank loops and perfect
13 loops all identified as vacancy-type in self-ion irradiated Ni. It contrasts with
14 the conventional rate theory based on the dislocation bias model in which only
15 interstitial loops can grow.

16 17 **Acknowledgement**

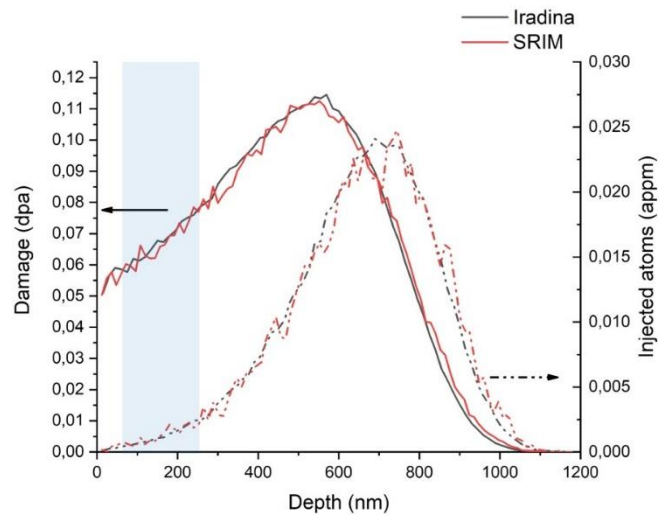
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23 24 **Declaration of interests**

1 The authors declare that they have no known competing financial interests or personal
2 relationships that could have appeared to influence the work reported in this paper.

3

4 **Appendix A. SRIM calculation of radiation damage**



5

6 Fig. A. 1 The damage profile of 2 MeV Ni²⁺ ions calculated by SRIM 2013 and IRADINA
7 for a fluence of 9×10^{13} ions.cm⁻² with a displacement threshold energy of 40 eV
8 [25,42,63].

9

1 **Appendix B. Visibility table for the determination of Burgers vectors**

2

3 Table B. 1 Visibility table of all families of Frank loops and perfect loops in function of
4 diffraction vectors used for Fig. 6.

Table of visibility (V=visible, I=invisible)									
Zone axes		[011]		[001]			[101]	BV analysis	
Possible b		$\bar{1}\bar{1}1$	$11\bar{1}$	020	$0\bar{2}0$	200	$\bar{2}\bar{2}0$		$\bar{2}02$
Frank loops	$\pm\frac{1}{3}[111]$	V	V	V	V	V	V	I	Loop A
	$\pm\frac{1}{3}[\bar{1}11]$	V	V	V	V	V	I	V	
	$\pm\frac{1}{3}[1\bar{1}1]$	V	V	V	V	V	I	I	
	$\pm\frac{1}{3}[11\bar{1}]$	V	V	V	V	V	V	V	
Perfect loops	$\pm\frac{1}{2}[110]$	V	V	V	V	V	V	V	Loop B
	$\pm\frac{1}{2}[1\bar{1}0]$	I	I	V	V	V	I	V	
	$\pm\frac{1}{2}[101]$	I	I	I	I	V	V	I	
	$\pm\frac{1}{2}[10\bar{1}]$	V	V	I	I	V	V	V	
	$\pm\frac{1}{2}[011]$	I	I	V	V	I	V	V	
	$\pm\frac{1}{2}[01\bar{1}]$	V	V	V	V	I	V	V	

5

6 **Appendix C. One-dimensional rate equations of PD concentrations**

7 The concentration profile of point defects along depth direction can be described
8 by one-dimensional steady-state diffusion equations for a given thickness and a given
9 temperature:

10
$$-D_v \frac{\partial^2 C_v}{\partial z^2} = G - K_r C_v C_i (D_v + D_i) \quad \text{Equation (3)}$$

11
$$-D_i \frac{\partial^2 C_i}{\partial z^2} = G - K_r C_v C_i (D_v + D_i) \quad \text{Equation (4)}$$

12 Where C_v and C_i are atomic fraction of vacancies and interstitials, D_v and D_i are
13 diffusion coefficients of of vacancies and interstitials, G production rate of point defects
14 in dpa/s, $K_r = 4\pi r_c / \Omega$ the recombination constant (r_c : recombination radius).

1 Equation (3) and (4) lead to $D_v C_v = D_i C_i$. Considering $D_i \gg D_v$, Equation (3) can
2 be normalized and rewritten as:

3
$$\frac{\partial^2 Y}{\partial X^2} = \frac{t^2 G}{D_v C_{v,\infty}} (Y^2 - 1) \quad \text{Equation (5)}$$

4 Where $Y = C_v/C_{v,\infty}$ with $C_{v,\infty} = \sqrt{G/(K_r D_v)}$ the vacancy concentration in bulk
5 materials without surface effects and $X = z/t$ with t the thickness of specimen.

6 Equation (5) gives the profile of vacancy concentration. A similar equation was
7 given by Foreman [51]. The form of the profile depends on the thickness, irradiation flux,
8 and the temperature. To solve Equation (5), the diffusion coefficient is calculated by
9 Arrhenius law with parameters as given in [17] and boundary conditions $C_v(z = 0) = 0$
10 ($C_{v,eq}$ at thermal equilibrium negligible to $C_{v,\infty}$) and $C'_v(z = 0.5) = 0$ (by symmetry).

11

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