Effects of Fe concentration on the ion-irradiation induced defect evolution and hardening in Ni-Fe solid solution alloys

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Abstract

Understanding alloying effects on the irradiation response of structural materials is pivotal in nuclear engineering. To systematically explore the effects of Fe concentration on the irradiation-induced defect evolution and hardening in face-centered cubic Ni-Fe binary solid solution alloys, single crystalline Ni-xFe (x=0-60 at%) alloys have been grown and irradiated with 1.5 MeV Ni ions. The irradiations have been performed over a wide range of fluences from 3×10^{13} to 3×10^{16} cm⁻² at room temperature. Ion channeling technique has shown reduced damage accumulation with increasing Fe concentration in the low fluence regime, which is consistent to the results from molecular dynamic simulations. No irradiation-induced compositional segregation was observed in atom probe tomography within the detection limit, even in the samples irradiated with high fluence Ni ions. Transmission electron microscopy analyses have further demonstrated that the defect size significantly decreases with increasing Fe concentration, indicating a delay in defect evolution. Furthermore, irradiation induced hardening has been measured by nanoindentation tests. Ni and the Ni-Fe alloys have largely different initial hardness, but they all follow a similar trend for the increase of hardness as a function of irradiation fluence.

Keywords: ion irradiation, solid solution alloy, nanoindentation, ion channeling, molecular dynamic simulation

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1. Introduction

Unveiling defect evolution in materials under irradiation and designing structural alloys with high radiation-resistance are fundamental to nuclear engineering.[1-4] The development of fusion and next-generation fission reactors demands materials that can withstand irradiations to higher fluence than do the current designs, and the challenges associated with radiation-induced material degradations, such as embrittlement, corrosion and swelling, have not been fully resolved.[2-4] Among the versatile approaches developed in the past decades, one of the favorable strategies in designing radiation-resistant alloys is to trap defects by dense sinks such as grain boundaries and secondary phases; nanocrystalline metals and oxide-dispersion-strengthened (ODS) steels are two of the major examples following this concepts.(e.g., [4-7]) While great progress has been made, this strategy faces challenges in materials processing and the structural stability at elevated temperature.

Other than modifying the microstructure, the concept of modifying the compositional complexity of alloys [7, 8] has recently drawn much interest due to the development of single-phase concentrated solid solution alloys (SP-CSA, including high-entropy alloys).[9-15] Different from traditional alloys, the SP-CSAs are structurally simple but compositionally complex (containing more than two principal alloying elements). They have exhibited extraordinary mechanical properties such as excellent fracture toughness at low temperature,[15, 16] which have inspired a range of potential applications. Moreover, recent studies have demonstrated that their irradiation response can be largely modified by tuning the compositional complexity without changing the microstructures.[8, 17] Scientifically, this family of alloys have motivated fundamental studies of the major compositional effects on the irradiation response by minimizing the variables in microstructures and dilute solute species. All these benefits render this family of alloys a new territory for studying irradiation damage of structural alloys.

The number, type and concentration of alloying elements are three basic components of compositional complexity. It has been shown that the type of alloying elements could be more

critical than the number of elements on the irradiation responses, as well as several mechanical and physical properties.[17-20] For example, studies on a family of Ni-containing face-centered cubic (fcc) equiatomic alloys revealed that addition of Fe can delay defect evolution more effectively under room temperature irradiation when compared to alloying with Co.[8, 21] Under high temperature, [17, 22] Fe additions in Ni can greatly reduce the volume swelling, more effective than the addition of Co and Cr. As a result, Ni-Fe based alloys are expected to be an interesting system in the development of the SP-CSA.

In this study, we will evaluate the effects of the concentration of principal elements on the irradiation response. Ni-Fe binary alloys in single-crystalline form are selected as a model system, with Fe concentration varied from 0 to 60 at%. The alloys are irradiated with self-ions (1.5 MeV Ni ions) at room temperature, spanning a wide range of fluences from 3×10^{13} to 3×10^{16} cm⁻². The defect evolution after irradiation is first characterized by using an ion channeling technique based on the Rutherford backscattering spectrometry (RBS-C) measurements. RBS-C is a single-crystalline technique to characterize the depth-resolved lattice damage in crystals.[23] It has been commonly used on studying the irradiation-induced damage in ceramics, but less applied in metallic (especially commercial) alloys, which is somewhat attributed to the fact that these alloys are usually polycrystalline. The availability of single crystalline samples enables RBS-C to systematically and fast examine the damage evolution in this series of modeling alloys. RBS-C can nevertheless hardly characterize the defect configurations and the possible irradiation-induced compositional segregation or phase separation, especially in those samples experienced high fluence irradiations. Therefore, we use atom probe tomography (APT) and transmission electron microscopy (TEM) to examine the compositional homogeneity and defect microstructures. Molecular dynamic (MD) simulations are performed to further understand the defect accumulation processes, and nanoindentation tests are used to measure the irradiation-induced hardening.

2. Experiments and methods

2.1 Single crystal growth and sample preparation

Pure nickel and/or iron (>99.9% pure) were carefully weighted and mixed by arc melting with the buttons flipped and re-melted five times to ensure good mixing before drop casting into a copper mold. Single crystals were then grown using a floating-zone directional solidification method from the polycrystalline drop-cast ingots.[24] The diameter of the molten zone was carefully reduced to produce a neck that prevented the slower growing grains from propagating.[25, 26] X-ray diffraction confirms the fcc structure for all the compositions up to 60 at% Fe. All the single crystals were re-oriented to (100) surface using backscattered Laue diffraction, and then cut into disks with thickness of ~1 mm using an electro-discharge machine (EDM). The samples were electrochemically polished to remove the damage layers of the machining.

2.2 Ion irradiation

The Ni and Ni-Fe alloys were irradiated with 1.5 MeV Ni ions using a tandem accelerator.[27] The irradiation fluence ranges from 3×10^{13} to $3 \times 10^{16} \text{ cm}^{-2}$. The flux was controlled at $\sim 1 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ for all the fluences except for $3 \times 10^{16} \text{ cm}^{-2}$, which required a flux of $\sim 8 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ to keep the irradiation within a favorable time. Assuming a displacement energy of 40 eV for both elements, corresponding to the above fluence range, the peak irradiation doses (appear at ~350 nm depth from the surface) range from 0.04 to 40 displacements per atom (dpa), according to the simulation using the Stopping and Range of Ions in Matter (SRIM-2013) code[28] in the “Kinchin-Pease formalism” mode. The dose rate is then approximately $0.0013 \text{ dpa s}^{-1}$ for all the fluences but for $3 \times 10^{16} \text{ cm}^{-2}$ (with a dose rate of $\sim 0.01 \text{ dpa s}^{-1}$). The dose rate effects are small in Ni and NiFe alloys within the regime of this study.[29] The samples were tilted $\sim 7^\circ$ off normal to minimize the channeling irradiation effects. All the irradiations were performed at room temperature.

2.3 Defect and microstructure characterizations

The irradiation induced defect evolution was first characterized using the RBS/channeling technique with 3.5 MeV He⁺ ions. In the RBS measurements, the backscattered He ions after collision with target atoms are detected by using a silicon detector set in a scattering angle of 155°. The energy of detected He ions along with the known stopping power provides the depth where the backscattering occurs. The incident He beam was aligned to the <100> axis of the samples.

APT was employed to measure the sub-nm scale elemental distributions of Ni-20Fe, Ni-50Fe and Ni-60Fe (both before and after irradiations at a fluence of $5 \times 10^{15} \text{ cm}^{-2}$) using a local electrode atom probe (LEAP 4000 XHR) under voltage mode, with a pulse fraction of 20% at 50 K and at a detection rate of 0.005 atoms per pulse.[30, 31] A FEI Nova 200 dual-beam focused ion beam (FIB) instrument was used to perform lift-outs and annular milling to fabricate the needle-shaped APT specimens. About 200 nm Pt electron beam deposition was first performed to protect the sample surface, followed by a ~1 micron Pt deposition at the FIB. The sample tips were taken at the depth of ~100-300 nm, corresponding to a dose range of ~4.5-6 dpa.

TEM samples were prepared by the FIB lift-out method using FEI helios Nanolab. Bright-field TEM images were taken under two-beam condition at $g = [200]$, using a JEOL 3011 TEM operated at 300 keV. The image pixels and exposure time were 1024×1024 and 2 s, respectively.[32] A flash-polishing technique has been employed in the sample preparation, as discussed in detail in Ref. [32], which can effectively remove the FIB-induced damage that may overlap with the primary ion-induced damage.

2.4 Nanoindentation

In order to measure the hardness change as a function of defect irradiation fluence, a Nanoindenter XP was utilized for the nanohardness measurements, using a Berkovich triangular pyramid indenter. Tests were conducted in the continuous stiffness mode (CSM) [33] at a

constant $\dot{P}/P = 0.05 \text{ s}^{-1}$. At least 24 indentations were performed on each sample to allow statistical analysis.

2.5 MD Simulations

The simulation procedure has been discussed in details in Ref. [34], and thus only key features are stated here. The Embedded atom method (EAM) based, the Bonny potential was used to model the defect evolution of Ni, Ni-20Fe, Ni-50Fe, and Ni-60Fe. The effects of prolonged radiation damage were simulated by overlapping of 5 keV single cascades. To achieve a damage dose equivalent to 0.133 dpa (NRT), 400 cumulative recoil simulations were performed. The MD simulation code LAMMPS [35] was used to create the random mixture of elements to mimic the SP-CSAs fabricated for experimental studies, and a variable time step MD code PARCAS was used for the cascade simulations.[36, 37] The simulation cell size was $115 \times 115 \times 115 \text{ \AA}^3$ containing 132,000 atoms. Each simulation runs for 30 ps at 300 K. Periodic boundary conditions were applied in all directions. The temperature was scaled at the periodic boundaries using Berendsen temperature control. In PARCAS, the electronic stopping is modeled as a friction force for atoms with kinetic energies higher than 1 eV and calculated using the SRIM code.

3. Results

3.1 Pristine single crystals

The compositional homogeneity and the initial crystal quality have been examined by Laue backscatter x-ray diffraction, APT and RBC-S, respectively. Fig. 1a and b representatively show the reconstructed atom probe volumes of the pristine Ni-50Fe samples, which demonstrate that both Ni and Fe are homogeneously distributed in all the alloys with no segregation. The actual composition of Ni (50.5 at%) and Fe (49.5 at%) determined by APT are close to the nominal composition, and no impurities other than trace O, N, and Ga (induced from APT sample

preparation) were detected in all the samples. Thus, the nominal composition is used for all the materials unless otherwise stated.

The statistics of the elemental distribution are further quantified by a frequency distribution analysis. The relative deviation from randomness is evaluated by the normalized chi-squared (χ^2) parameter, known as the Pearson coefficient (μ), [38] where 0 represents randomness and 1 represents complete association in the occurrence of the solute atoms. In all the tested alloys (Ni-20Fe, Ni-50Fe and Ni-60Fe), both Ni and Fe elements show μ values very close to 0, i.e., practically complete randomness, which demonstrates that these alloys consist of a single phase solid solution. The deviation from 0 is most likely due to variations in the APT data due to crystalline poles.

The ion channeling spectra of the pristine samples (Fig. 1c) show that the channeling yield close to surface is less than 5% of the “random” yield for all the materials, indicating high crystal quality. Furthermore, all the samples have similar channeling spectra throughout the depth of interest, demonstrating comparable initial crystal qualities.

3.2 Defect evolution under ion irradiation

Fig. 2 representatively shows the channeling spectra for the Ni-Fe alloys with Fe concentration from 0 to 60 at%, under Ni ion irradiation to various fluences. In RBS-C spectra, the backscattering yield indicates the probability of close-encounter between the incident He ions and the lattice atoms.[23] In general, when the incident ions are aligned to the low-index crystal axes of a high-quality single crystal, the backscattering yield is very low (shown as the pristine level); the backscattering yield increases with increasing damage of the crystal or lattice distortion, towards the upper limit of the “random” level.[23]

As shown in Fig. 2a-d, the comparison in irradiation response between different materials changes in different fluence regimes. Under low fluence irradiations such as 6×10^{13} and 1×10^{14} cm⁻² (Fig. 2a), where the peak dose is below 0.14 dpa, Ni-35Fe, Ni-50Fe and Ni-60Fe do not

exhibit significant difference in backscattering yield beyond the uncertainty of the experimental measurements. However, the backscattering yield of these three materials is obviously lower than that of Ni-20Fe, which is further lower than elemental Ni. With increasing fluence, e.g. to $2 \times 10^{14} \text{ cm}^{-2}$, the difference among Ni-35Fe, Ni-50Fe and Ni-60Fe becomes considerable such that the backscattering yield decreases with increasing Fe concentration. At this dose, the difference between Ni and Ni-20Fe becomes negligible, probably because the damage in Ni begins to saturate.[21, 39] As shown in Fig. 2b, this trend remains similar until a fluence of $3 \times 10^{14} \text{ cm}^{-2}$, when the spectrum for Ni-35Fe roughly overlaps with that of Ni, and the damage level of Ni-20Fe starts to be higher than Ni. As the irradiation fluence reaches $8 \times 10^{14} \text{ cm}^{-2}$ the spectra of all the materials appear similar, with Ni having a slightly lower yield (Fig. 2c). Under high fluence irradiation of $3 \times 10^{15} \text{ cm}^{-2}$ (Fig. 2d), the spectrum of pure Ni is apparently lower than the alloys, which is opposite to the trend in the low fluence regime. And the difference between Ni-50Fe and Ni-60Fe becomes small again. This reversed relationship between Ni and the alloys was further observed in a high fluence irradiation up to $3 \times 10^{16} \text{ cm}^{-2}$ in Ni and Ni-50Fe.

The fluence dependent channeling spectra after irradiations are depicted in Fig. 3 for both Ni (Fig. 3a) and Ni-50Fe alloy (Fig. 3b), which illustrate the difference in damage evolution of these two materials systems. At low fluences, the damage build-up in Ni is clearly faster than in NiFe. With further irradiation, the spectra for Ni are almost overlapped for the fluences of 2, 3, and $4 \times 10^{14} \text{ cm}^{-2}$, while the build-up continues in Ni-50Fe until $\sim 8 \times 10^{14} \text{ cm}^{-2}$. This means that the damage reaches saturation in Ni at about $2 \times 10^{14} \text{ cm}^{-2}$, while the saturation in NiFe occurs at about $8 \times 10^{14} \text{ cm}^{-2}$. In short, with increasing Fe concentration to 60 at%, the damage build-up speed is decreased in the low fluence regime and the saturation fluence generally increases. This suggests that alloying with Fe delays the defect evolution process in the Ni-Fe system.

3.3 Irradiation induced hardening

To evaluate the mechanical property change after irradiation in this system, another batch of samples with Fe concentration up to 60 at% were irradiated from 3×10^{13} to $5 \times 10^{15} \text{ cm}^{-2}$, and

subsequently tested by nanoindentation. The hardness of the pristine samples is shown in Fig. 4a, which increases from ~1 to 3 GPa when the Fe concentration increases from 0 to 20-35 at%. With more Fe content until 60 at%, the hardness of Ni-Fe alloys is reduced down to 2.2 GPa. This trend qualitatively agrees to the room temperature tensile yield strength.[40] Fig. 4b summarizes the hardness evolution of Ni and the alloys as a function of irradiation fluence. While each material has a different initial hardness, the evolution of hardness in each material follows a similar trend; the hardness increases faster in the early stage, below $2 \times 10^{14} \text{ cm}^{-2}$, and the change becomes slower in the high fluence region. Note that the hardening is significant even for the lowest fluence, $3 \times 10^{13} \text{ cm}^{-2}$, corresponding to a dose of ~0.04 dpa.

The absolute hardness changes for all the tested materials are plotted in Fig. 4c as a function of irradiation fluence. Although directly correlating the indentation hardness to the bulk mechanical (e.g. tensile) properties is non-trivial, it has been shown empirically that the change of indentation hardness (ΔH) is approximately proportional to the change in tensile yield strength $\Delta \sigma_y$ obtained in tensile tests.[22, 41] As a result, the irradiation hardening data could be analyzed based on the well-established strengthening relationship [42],

$$\Delta H \propto \Delta \sigma_y = \alpha M G b (N d)^{1/2}, \quad (1)$$

where M is the Taylor factor, which is ~ 3 for fcc lattice, α is a material dependent constant for the average barrier strength of loops, G is the shear modulus, b is the Burgers vector, N is the defect density and d is the defect size. In order to eliminate the effect of known difference in G [43] between the different materials, the hardening data are normalized and shown in Fig. 4d.

To evaluate the hardening as a function of dose or fluence, the saturation effects on the defect evolution must be considered. One of the commonly used descriptions [44, 45] for the defect density is

$$N = A_0 [1 - \exp(-BD)], \quad (2)$$

where A_0 and B are constants and D is the dose or fluence. In addition, the fluence exponents of less than 0.5 are commonly observed in the literature.[22] Thus, here we describe the hardness change as a function of fluence using the equation:

$$\Delta H = A[1 - \exp(-BD)]^C \quad (3)$$

As seen in Fig. 4d, the hardening data of all these materials overlap within the range of experimental uncertainties, despite their large difference in initial hardness and the different damage evolution behavior found by the ion channeling technique. All the data reasonably fall into the same curve under Eq. (3), with the fluence exponent C of ~ 0.24 .

The hardness profiles obtained from continuous stiffness method are plotted in Fig. 5. In each plot, the hardness under the irradiations spanning three orders of fluences is presented. The “bands” are composed by the error bar from the statistics of a number of (>24) indentation locations at each depth. The indentation size-effects are minimized since the hardness profiles of each pristine sample are subtracted, while the pristine hardness profiles are already reasonably flat (not shown). The contact displacement shallower than 60 nm is cut off to avoid the artifacts of the surface and indentation tip bluntness. Note that the stress field influenced region in the nanoindentation is more than 5 times deeper than the indentation depth of ~ 200 nm shown in Fig. 5, [46] hence the hardness represents the mechanical properties of surfaces within $\sim 1\mu\text{m}$ in depth.

Below the fluence of $4 \times 10^{14} \text{ cm}^{-2}$, the hardness over the whole depth range increases with increasing fluence. With further irradiations, the hardness in the first few hundred nanometers is saturated, indicating saturation in the product of defect density and size according to Eq. (1). Previous TEM analyses [32] have shown that, under Au ion irradiations at room temperature, the dislocation loop size increases while the loop density decreases at the high fluences in both Ni and Ni-xFe alloys (details in microstructure will be discussed in Sec. 4). The present mechanical property data, thus, qualitatively agree with the microstructure evolution. Other than the hardness

values, the hardness profiles also qualitatively indicate the evolution of damage depth distribution. In Ni, for example, the depth for the maximum hardness becomes deeper with increasing fluences. Such extension in damage range indicates the migration of defects, which has been frequently reported in fcc single crystals [47, 48] and analyzed in detail in Ref.[21, 32]

4. Discussion

In the radiation dose region from a fraction to a few dpa, TEM observations [8, 32] have shown that Ni-50%Fe has much fewer large defects (e.g. >10 nm), but more small defects compared with pure Ni. This feature has been captured by MD simulations, [29, 34] one of which has also shown that the damage accumulation in Ni is faster than that in Ni-20Fe. Here we further demonstrate in Fig. 6 the MD simulation result of the number of surviving displaced atoms (the atoms that eventually stay at off-lattice positions) in Ni, Ni-20Fe, Ni-50Fe and Ni-60Fe at different irradiation dose levels. Although little difference was observed for all of these materials at very low dose (e.g. ~0.003 dpa), the damage accumulation obviously slows down with increasing Fe concentration in the higher dose regime of ~0.13 dpa (the peak dose corresponding to the irradiation fluence of $1 \times 10^{14} \text{ cm}^{-2}$). Note that the difference between Ni-50Fe and Ni-60Fe is negligible within the error range of the simulations. This result qualitatively agrees with the results of RBS-C study, as shown in Fig. 2a. In addition, the (interstitial-type) defects in Ni-20Fe are found generally smaller than those in pure Ni but larger than in Ni-50Fe. In other words, larger interstitial-type defects tend to form with decreasing Fe concentration. This result strongly indicates that the interstitial mobility decreases with increasing Fe concentration.

Quantitative interpretation of channeling results in metals and alloys is challenging in the high fluence/dose regime. The backscattering yield evaluates the overall lattice distortion through the view of incident He direction (i.e. <100> in this study), but it hardly considers the evolution of the defect nature over a large range of irradiation doses. For example, the decrease in

backscattering yield after the saturation dose has been frequently observed in high dose irradiated radiation-tolerant ceramics such as MgO[49] and ZrO₂[50]. In these cases, the decrease in channeling yield could be somewhat attributed to that the defect structure becomes more organized in the high dose regime, e.g. small dislocation loops aggregating into dislocation networks, releasing irradiation-induced strain, decreasing the dechanneling cross section and consequently the backscattering yield. In the case of metals, the defects also experience the evolution from the black dot defects to large dislocation loops.[39, 51-53] While the delay effect by increasing Fe concentration in the defect evolution is clear based on the channeling results, the direct comparison on microstructures and defect configurations is not available solely from the channeling spectra. Furthermore, compositional segregation is another important potential irradiation effect in alloys,[22] which, in our case, requires the evaluations from complementary techniques due to the similar atomic mass between Ni and Fe and the limit of mass and spatial resolution of RBS.[54]

As a result, APT was utilized to examine the distribution of Ni and Fe elements for Ni-20Fe, Ni-50Fe and Ni-60Fe after being irradiated at a fluence of $5 \times 10^{15} \text{ cm}^{-2}$. The APT atom maps of Ni-60Fe are representatively shown in Fig. 7. Similar to the case of pristine samples, the elemental distributions are homogeneous and the frequency distribution analyses (Fig. 7d, 7e) prove that no segregation or phase separation occurred in any of these compositions during irradiation, even at such a relatively high fluence in the present study; the materials maintained a homogeneous solid solution phase. The slight deviation of the observed Ni and Fe to the binomial distribution, as shown in Fig. 7d and 7e, is due to the observation of a pole in the APT data, and in reality the observed and binomial are expected to match almost 100% without a pole in the data. The chemical homogeneity is also maintained for the Ni-20Fe and Ni-50 Fe samples after irradiation.

To further understand the defect configurations and sample microstructures. TEM analyses have been performed in Ni, Ni-20Fe, and Ni-50Fe after Ni ion irradiation to a fluence of $5 \times 10^{15} \text{ cm}^{-2}$

at a depth of ~350 nm corresponding to the area with a peak dose of ~6.5 dpa. The TEM images (as shown in Fig. 8) clearly show that the size of dislocation loops (identified as interstitial-type) decreases significantly with increasing Fe concentration. In pure Ni, large dislocation loops (> 30 nm) are frequently observed, but in the alloys, especially in Ni-50Fe, most of the defects are the so-called “black dot defects”, which have been identified as either small interstitial loops of a few nm or vacancy type stacking fault tetrahedra (SFT).[32] Quantitatively, the distribution of defect size for Ni, Ni-20Fe and Ni-50Fe are shown and compared in Fig. 9, and the average size of defects in Ni, Ni-20Fe and Ni-50Fe are ~9, 4, and 3 nm, respectively. Apparently, the aggregation of interstitial-type dislocation loops slows down with the addition of Fe ions, which indicates an increased irradiation resistance. This result agrees with the previous TEM results for low dose irradiations.[8, 32] Moreover, this result can well interpret the RBS-C results, consistent to the discussions above, that with increasing fluences, the growth of dislocation loops and reduction of associated lattice strain make the channeling yield decrease, similar to the cases of various ceramics. Furthermore, we found that, not surprisingly, while the average defect size of Ni is greater than the alloys, the defect density increases with increasing Fe concentration: the defect densities of Ni, Ni-20Fe and Ni-50Fe have been counted as 0.4, 1.2, and $1.9 \times 10^{23} \text{ m}^{-3}$, respectively.

The compositional effects on the defect evolution observed in channeling studies were, however, not observed beyond the error bar in the mechanical property tests using nanoindentation. One of the possible reasons is that we only normalized the hardening effects to the shear modulus, but not to the barrier strength α , which is a material dependent factor. It is possible that α changes with Fe content. Another possible reason is that, according to the TEM observations (for high dose irradiations in this study and low dose irradiations in literatures[8]), the defect size decreases with increasing Fe concentration but the defect density increases with increasing Fe concentration. Since the hardening effects consider both defect size and density (Eq. 1), such combined effects can make the hardening less sensitive to the delay in the microstructure

evolution in the present case. In the practical sense, current experiments indicate that the hardening under room temperature self-ion irradiation of Ni is not affected significantly with increasing Fe concentration.

5. Summary and conclusions

Compositional effects on the defect evolution and hardening in fcc Ni-xFe binary solid solution alloys under irradiation have been systemically studied by increasing the Fe composition from 0 to 60 at%. Single crystals have been successfully grown and irradiated with 1.5 MeV Ni ions. Combining experimental investigations with computational simulations, the following conclusion can be drawn:

1. Under irradiations at low fluences, the overall irradiation-induced damage observed along the $\langle 100 \rangle$ channel is high for pure Ni, and decreases with increasing Fe concentration. Such resistance in defect accumulation has also been captured by MD simulations that the addition of Fe efficiently decreases the number of surviving displaced atoms in Ni-xFe alloys.
2. No irradiation-induced compositional segregation was observed in APT examinations, even in the samples irradiated with high fluence of Ni ions.
3. TEM observations have demonstrated that, under high fluence irradiations, the defect size significantly decreases with increasing Fe concentration, but the defect density in Ni is smaller than that in the Ni-xFe alloys.
4. Although the defect evolution is delayed by the addition of Fe, the compositional dependence is not significant in the hardening processes under room-temperature irradiations, presumably attributed to the combined effects on the size and density of defects.
5. Both channeling and continuous hardness profiles have shown that the damage ranges increase with increasing fluences for Ni and all the tested Ni-xFe alloys.

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Figure captions

Fig. 1. Atom probe tomography shows a homogenous distribution of (a) Ni and (b) Fe elements in the single crystals (Ni-50Fe is representatively shown); (c) ion channeling spectra show that all the tested materials have similar initial crystal quality.

Fig. 2. Ion channeling spectra of Ni-xFe with x from 0 to 60 at% after 1.5 MeV Ni ion irradiations to the fluence of (a) 1×10^{14} (b) 3×10^{14} (c) 8×10^{14} and (d) $3 \times 10^{15} \text{ cm}^{-2}$.

Fig. 3. Ion channeling spectra of (a) Ni and (b) Ni-50Fe after irradiations from 6×10^{13} to $3 \times 10^{16} \text{ cm}^{-2}$. The captions of fluence are applied to both figures. The bottom x-axes are the outputs from the detector, while the top x-axes are the converted depth as shown in Fig. 2.

Fig. 4. (a) Nanoindentation hardness of the pristine samples, as a function of Fe concentration in fcc Ni-Fe alloys; (b) nanoindentation hardness of fcc Ni-xFe irradiated with 1.5 MeV Ni ions to the fluence up to $5 \times 10^{15} \text{ cm}^{-2}$; (c) and (d) show the absolute, and normalized hardness change derived from (b), respectively.

Fig. 5. The changes of hardness as a function of depth in (a) Ni, (b) Ni-35Fe, and (c) Ni-50Fe, under 1.5 MeV Ni ion irradiations to the fluence of 3×10^{13} , 4×10^{14} and $5 \times 10^{15} \text{ cm}^{-2}$.

Fig. 6. The number of surviving displaced atoms as a function of Fe concentration in the Ni-xFe alloys, at different irradiation dose levels of 0.003, 0.07 and 0.13 dpa, according to MD simulations.

Fig. 7. The APT atom maps of (a) all ions, (b) Ni, and (c) Fe for Ni-60Fe after Ni ion irradiation with a fluence of $5 \times 10^{15} \text{ cm}^{-2}$. The frequency distribution analyses of both (d) irradiation-free and (e) post-irradiation samples indicate no obvious chemical variance after irradiation. The sampling of experimental data is 100 ions per bin.

Fig. 8. Microstructure observed by TEM of (a) Ni, (b) Ni-20Fe, and (c) Ni-50Fe after Ni ion irradiation with a fluence of $5 \times 10^{15} \text{ cm}^{-2}$. Some typical SFT and dislocation loops are marked with red circles and blue arrows, respectively. A decrease in the dislocation size is clearly visible for Ni-xFe alloys with higher Fe contents.

Fig. 9. Defect size distribution in Ni, Ni-20Fe and Ni-50Fe after Ni ion irradiation with a fluence of $5 \times 10^{15} \text{ cm}^{-2}$, observed by TEM.

Pristine Ni-50Fe

20 nm

(a) Ni

50 nm

(b) Ni-20Fe

50 nm

(c) Ni-50Fe

50 nm

Fraction (%)

Defect size distribution
by TEM

Ni
Ni-20Fe
Ni-50Fe

Ion irradiation induced defect evolution and hardening in Ni-Fe solid solutions

Ni-20Fe 50 nm

Ni-50Fe 50 nm