Supporting Information for

Mechanisms of chemical-reaction-induced tensile deformation of Fe/Ni/Cr alloy revealed by reactive atomistic simulations

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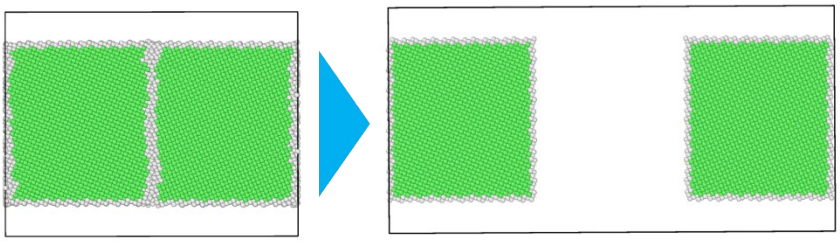
Reason of choosing ∑37(610) grain boundary

There are two reasons for us to use ∑37(610) as the research targeted: (i) the grain boundary should not be too unstable, so that a clear border line of boundary could not be maintained, (ii) on the other hand, the targeted grain boundary should also not be too stable, so that we cannot easily observe our focused phenomenon such as cracking. The grain boundary of ∑37(610) is a well-balanced choice for the above considerations: the atomic structures of ∑37(610) is very clear, while ∑37(610) is indeed relatively unstable compared with other grain boundaries. As the evidence, the surface energies of distinct grain boundaries are compared: Σ5(210), Σ13(320), Σ17(410), Σ29(520), Σ37(610), Σ41(540), Σ53(720), and Σ61(650). As shown in Figure S1, the surface energy of grain boundary ($E_{surface}$) is defined as the energy change per unit area when a grain boundary is separated: $E_{surface} = (E_1 - E_2)/2S$, where E_1 and E_2 are total energy of the connected and separated grain boundary respectively, and S is the sectional area. The higher $E_{surface}$ is, the grain boundary is more stable. According to the $E_{surface}$ results, the grain boundary of Σ37(610) has very low $E_{surface}$ although it is not the lowest one, and thus it is picked up to conduct tensile simulations.



Grain boundary	Σ5 (210)	Σ13 (320)	Σ17 (410)	Σ29 (520)	Σ37 (610)	Σ41 (540)	Σ53 (720)	Σ61 (650)
E_{surface} (J/m^2)	88473.3	96399.8	58392.6	102043.4	48010.3	38505.5	52191.1	120011.7

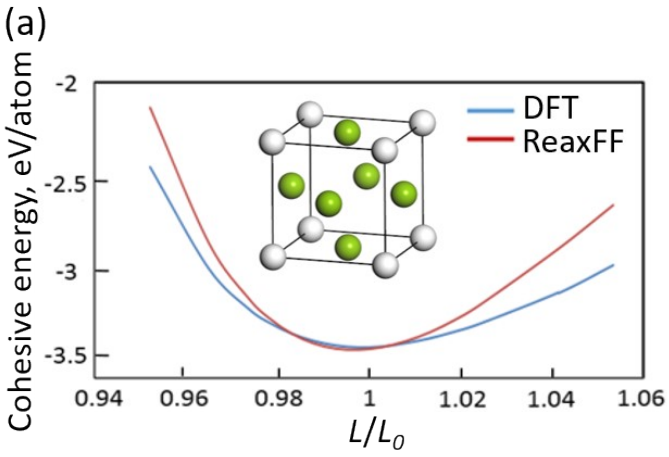
Figure S1. Surface energy of Fe/Ni/Cr alloy grain boundaries.

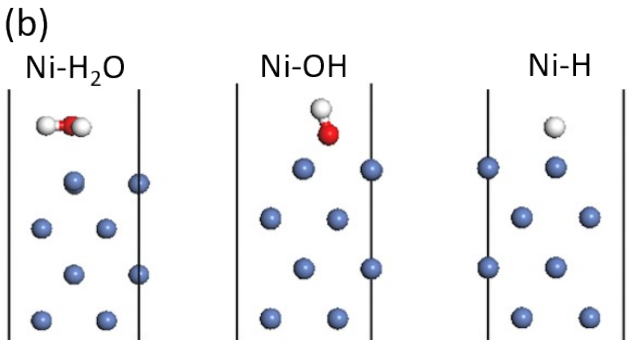
Development of ReaxFF parameters

The ReaxFF parameters are developed by comparing the interactions with density functional theory (DFT) calculations. In DFT calculations, we use the generalized gradient approximation (GGA)-type Perdew-Burke-Ernzerhof (PBE) functional with a DND basis set. While, the effective core potential method is used to treat the core electrons.

To develop the interatomic ReaxFF parameters of Ni/Cr, a Ni_3Cr primitive cell is picked up, and the cohesive energy per atom is calculated by DFT and ReaxFF with various uniform expansion/compression of volume. The ReaxFF parameters of Ni/Cr were determined to reproduce the DFT results. Figure S2a shows the obtained energy curve of Ni_3Cr obtained by DFT and ReaxFF. The horizontal axis is the ratio of the changed lattice length (L) to the original lattice length (L_0), and the vertical axis is the cohesive energy per atom. The two energy curves show a good agreement, indicating a good accuracy of our developed ReaxFF parameters.

Then, to develop the ReaxFF parameters for Ni/H_2O system, the adsorption energies of H_2O, OH, anad H on a Ni (110) surface are calculated by DFT and ReaxFF, and the ReaxFF parameters are determined to ensure the similar adsorption energies by DFT and ReaxFF as possible as we can. Figure S2b shows the calculation models and corresponding adsorption results. Using our developed ReaxFF parameters, the adsorption energies calculated by DFT and ReaxFF are in reasonable agreements.





	ReaxFF, eV	DFT, eV
Ni-H_2O	-0.89	-0.58
Ni-OH	-0.57	-0.49
Ni-H	0.51	0.53

Figure S2. Comparison between ReaxFF and density functional theory (DFT) calculations. (a) Cohesive energy of Ni_3Cr primitive cell where green and white are Ni and Cr atoms respectively. (b) Adsorption energies of H_2O, OH, and H on Ni surface while blue, red, and white are Ni, O, and H atoms respectively.

Tensile behaviors of Fe/Ni/Cr alloy with distinct element proportions

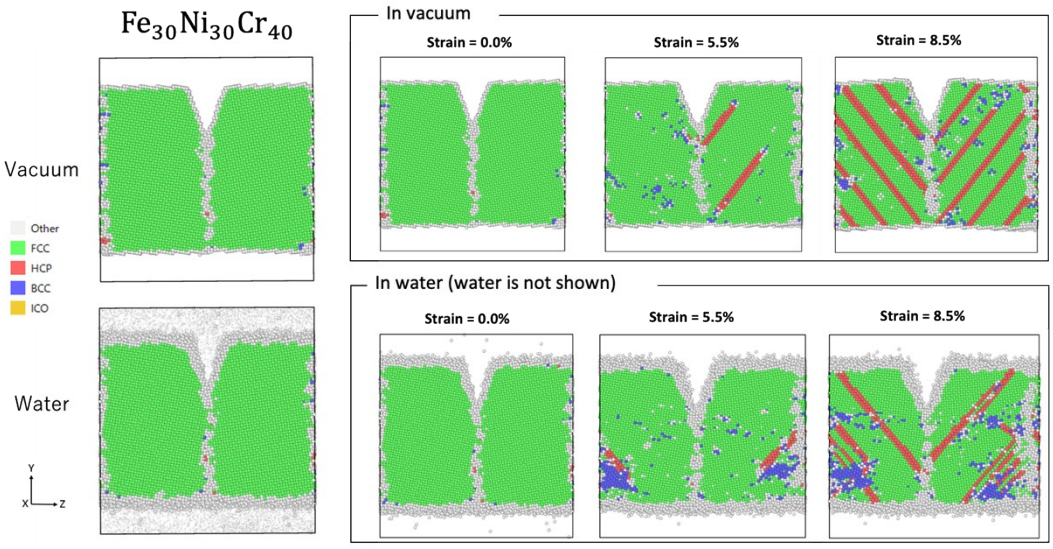


Figure S3. Atomic structures during the tensile deformation of $Fe_{30}Ni_{30}Cr_{40}$ alloy.

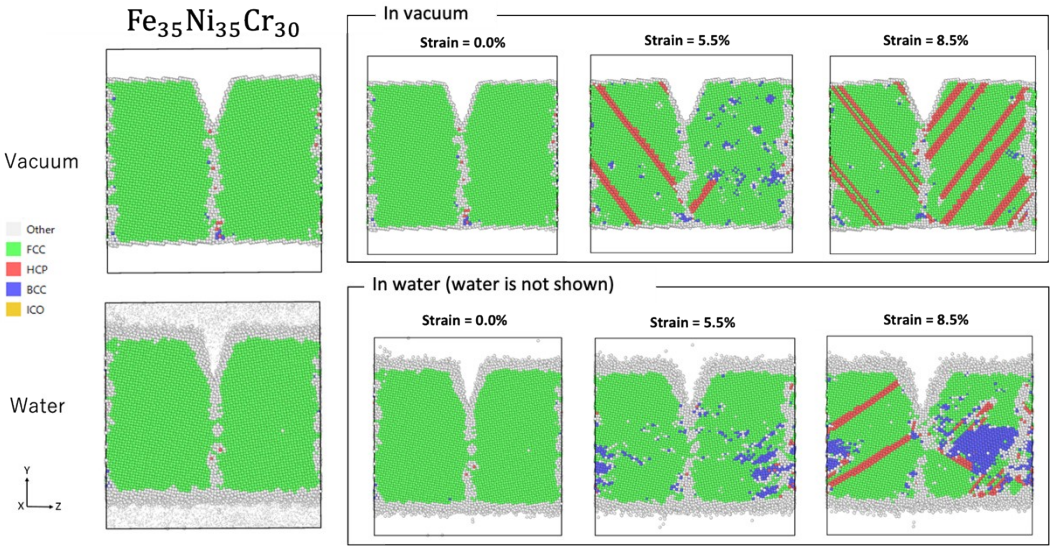


Figure S4. Atomic structures during the tensile deformation of $Fe_{35}Ni_{35}Cr_{30}$ alloy.

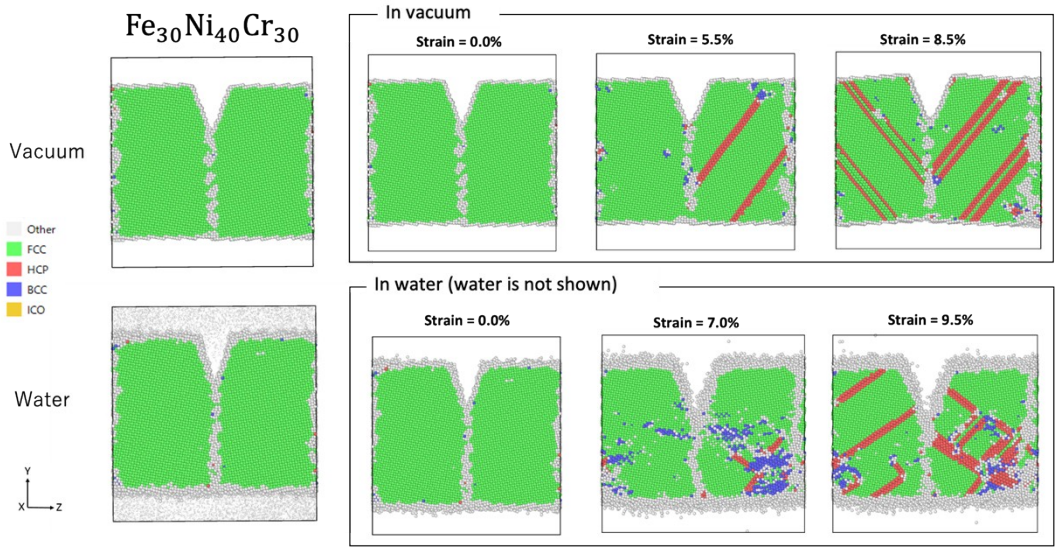


Figure S5. Atomic structures during the tensile deformation of $Fe_{30}Ni_{40}Cr_{30}$ alloy.