Supplementary Information

S1. Temperature selection for a-C-to-graphene transformation

In order to define the temperature for studying the diffusion behavior, the Ni@a-C system with Ni/C atomic ratio of 1/1 is used. The temperature increases from 300 to 2400 K by stepwise heating method, and the system is diffused at each temperature for 100 ps. At each temperature, the diffusion behavior of C and Ni atoms and corresponding structural evolution of Ni@a-C system are evaluated.

Evolution of Kinetic energy (KE) and potential energy (PE)

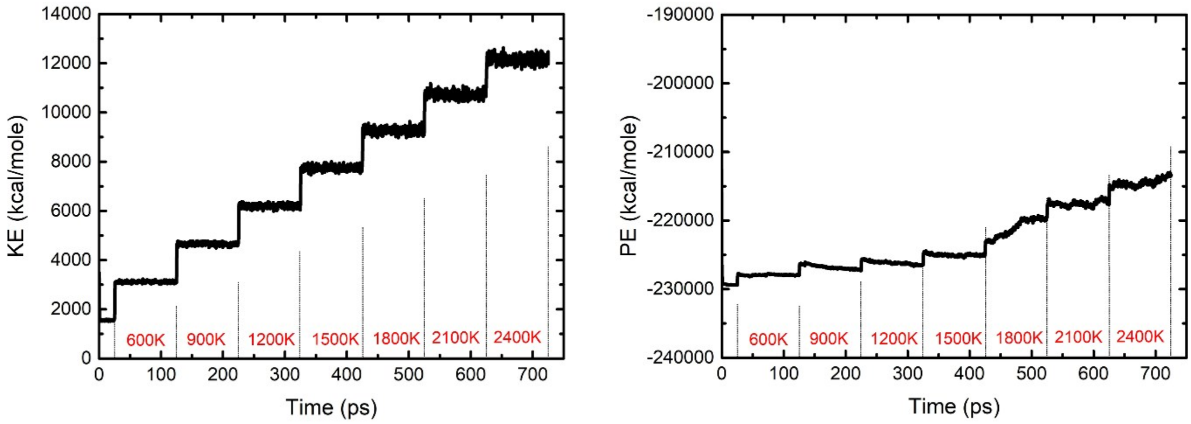


Fig. S1. Evolution of both kinetic energy (KE) and potential energy (PE) as a function of diffusion time.

Diffusion behavior and structural transformation at different temperatures

In order to evaluate the diffusion behavior at each temperature, the mean-squared displacement (MSD) is estimated as follows:

$$\mathrm{MSD} = r^2(t) = \frac{1}{N}\left\langle \sum_{i=1}^{N} \left| r_i(t) - r_i(0) \right|^2 \right\rangle \qquad (1)$$

where, *N* is the number of *i* atoms in the system, $r_i(t)$ is the position of the *i* atom at *t* moment, and $r_i(0)$ is the position of the *i* atom at *t=0* moment. From the MSD (Fig. S2) and the change in C and Ni atomic fractions along the diffusion couple direction (Fig. S3), we find that at 300K, the clear sharp interfaces separating a-C and Ni can be observed. When the temperature is located at the range of 300~1500 K, the interatomic interaction already causes some local atomic movement at the interface, the atoms at interface mix together and the sharp interface becomes a bit fuzzy (Fig. S3), so the obvious C-Ni transition layer is formed, while the MSD for Ni or C almost has no change, indicating the solid state of a-C and Ni layers. As increasing the temperature to 1500 K, the MSD curves for C and Ni atoms begin to increase obviously with diffusion time, but there are still only few C atoms diffusing into Ni layers during the time of 100 ps. With the temperature further increased to be higher than 1500 K, the MSD for C and Ni atoms increase significantly and more C atoms can further diffuse into Ni structure to form a diffusion zone, indicating the serious change in structure.

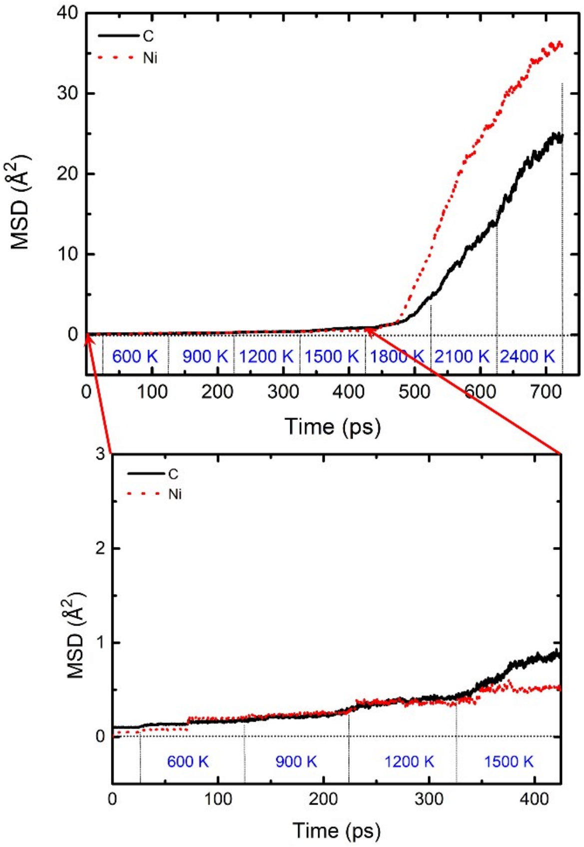


Fig. S2. MSD for C and Ni atoms as a function of diffusion time at each temperature.

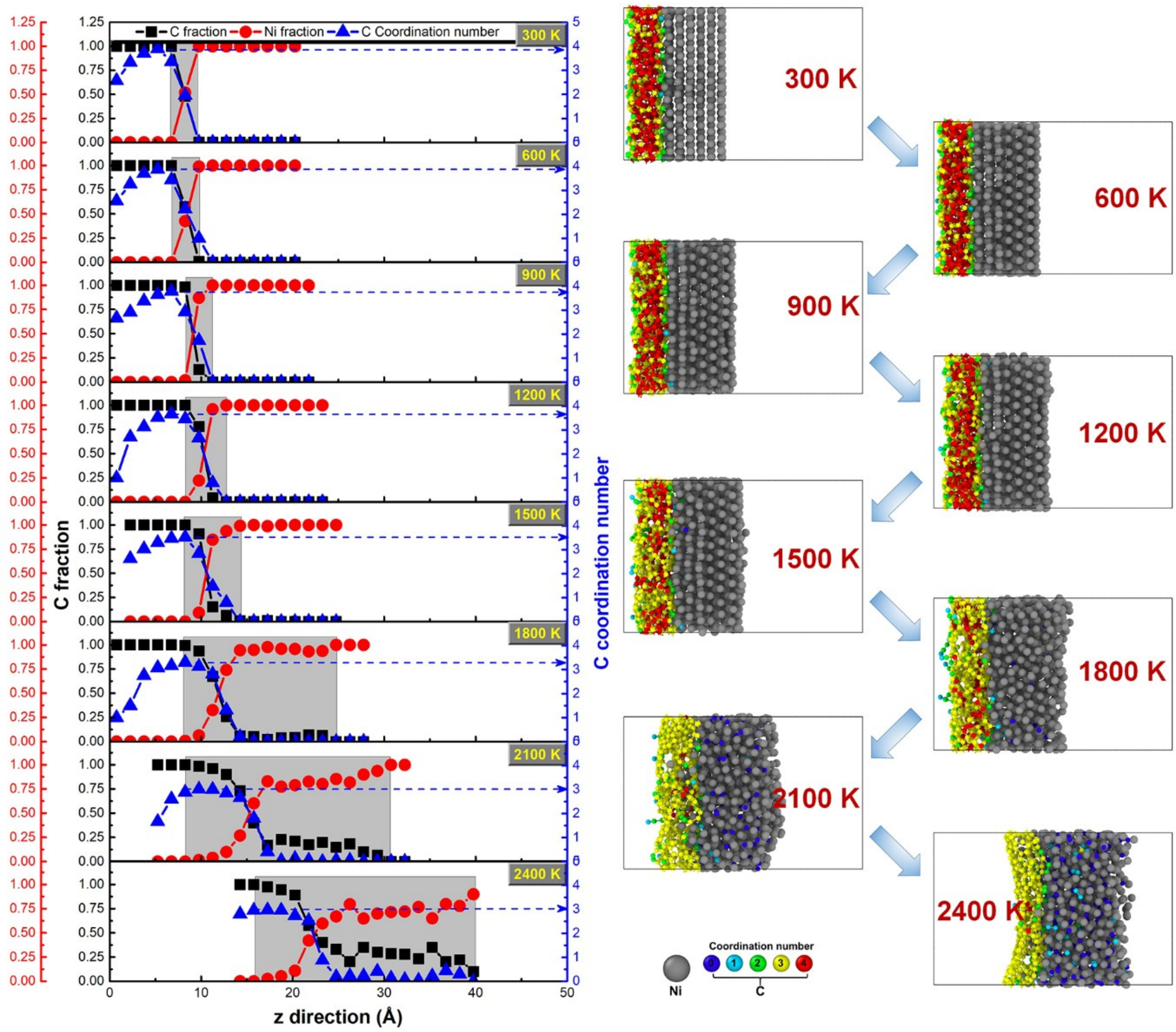


Fig. S3. Distributions of C and Ni atomic fractions along the diffusion couple direction and corresponding morphologies after diffusion time of 100 ps at each temperature.

Fig. S4 gives the change in 4- and 3-coordinated C fractions with temperature. It can be seen that when the temperature increases from 300 to 1800 K, the 3-coordinated C fraction increases significantly following the decrease of 4-coordinated C fraction; however, as the temperature further reaches to be higher than 1800 K, the dissolution of graphitic structure into Ni layer causes the reduction of 3-coordinated C fraction, which is not favorable to the formation of graphene structure. Based on the abovementioned analysis of diffusion behavior of C and Ni atoms and the evolution of hybridization structure, the temperature to study the a-C-to-graphene transformation is suggested to be 1500≤T≤1800 K. However, by further extending the diffusion time of system at 1500 or 1800 K to 1350 ps, Fig. S5 illustrates that more C atoms diffuse into Ni layer when the temperature is 1800 K, while only few C atoms diffuse into Ni layer at 1500 K. Therefore, in order to investigate the diffusion of C into Ni and the a-C-to-graphene transformation during the short molecular dynamics (MD) simulation time, the temperature of 1800K is adopted in the present work for the following calculations.

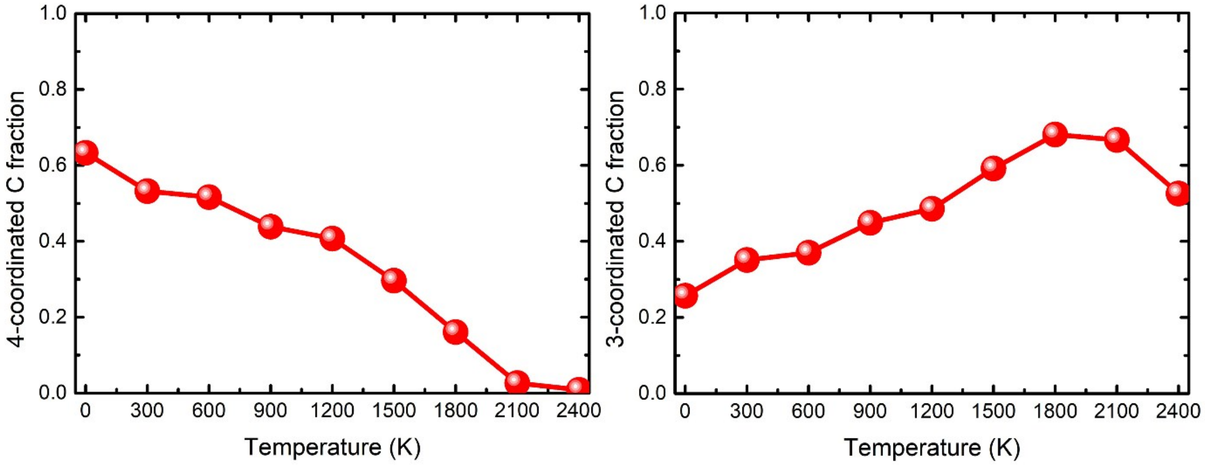


Fig. S4. Hybridization fraction of a-C structure with temperature.

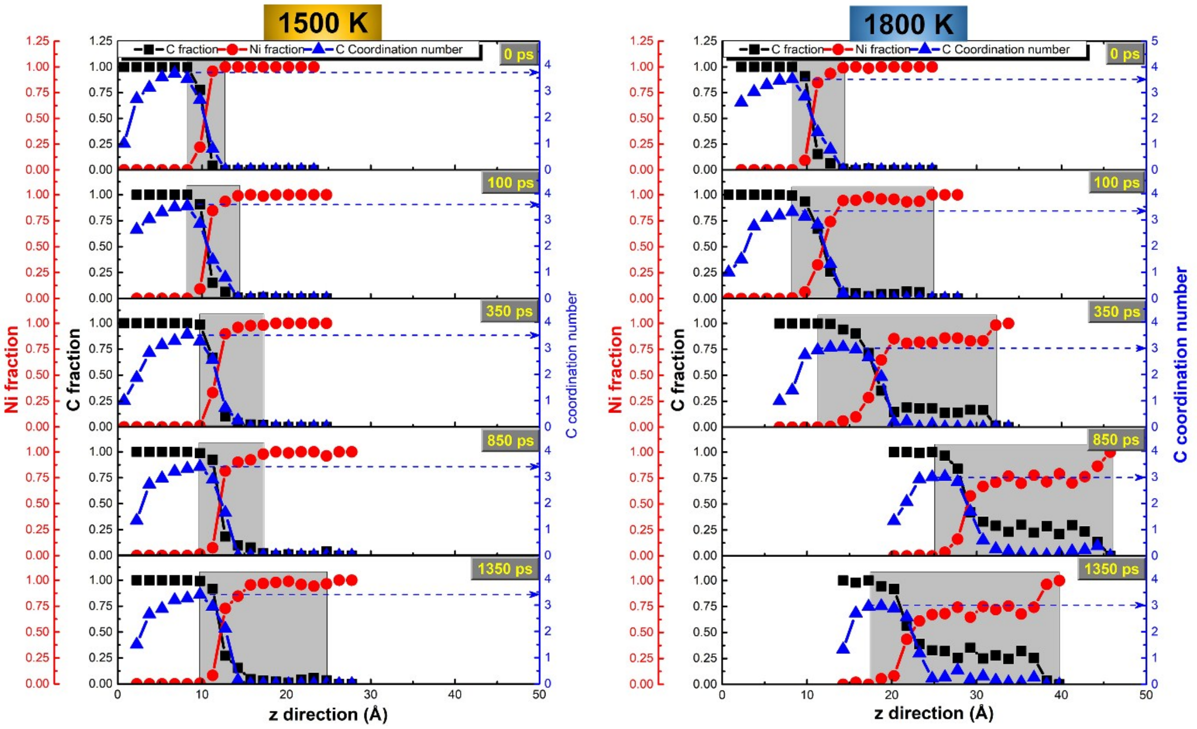


Fig. S5. Distributions of C and Ni atomic fractions along the diffusion couple direction when the system is diffused at 1500 K and 1800K for 1350 ps, respectively.

S2. Validation for ReaxFF potential

ReaxFF potential developed by Mueller *et al.*[1] is used to describe the interaction between the C and Ni atoms, which is able to model the bond forming/breaking, atom arrangement/rehybridization, chemical reactions and phase transformation accurately in carbon-based structures. Although it has been proved to be suitable for our system in previous calculation, many additional calculations for this potential including the fabrication of a-C structure (Fig. S6) using quenching method,[2] the temperature-dependence of formation energy of a-C structure (Fig. S7), the formation energies of a-C structures with different densities (Fig. S8), the energy changes of diamond, Ni crystal, and a-C induced by expansion and compression of supercell size (Fig. S9), the surface energy of Ni crystal (Fig. S10), and the formation energies of Ni-C mixing structures generated at the different stages of the quenching process (Fig. S11) are further performed by ReaxFF MD and ab-initio calculations[3,4] separately to validate the reliability of the used ReaxFF potential. They clearly display that the a-C structure is well reproduced compared to our and previous density function theory (DFT) calculations[5-8] and experiment results,[9,10] as shown in Fig. S6; the energy changes from ReaxFF are also consistent with those from ab-initio calculations in Fig. S7~S10, and the changes of both the formation energy and radial distribution function (RDF) spectra in Fig. S11 indicate that the Ni-C interaction can be accurately described compared to the ab-initio results, suggesting the validity of the force field for our simulated Ni@a-C systems.

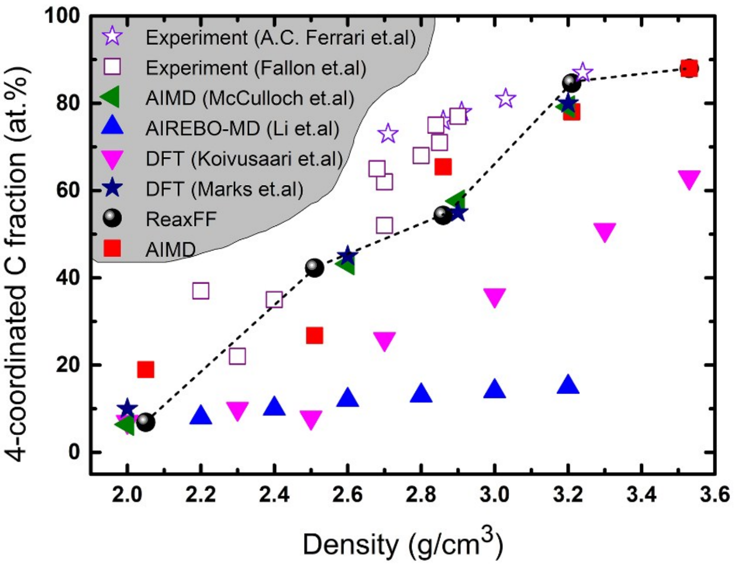


Fig. S6. Fabrication of a-C structure by ReaxFF MD simulation using quenching method. The results are compared with our and previous DFT calculations[5-8] and experiment results.[9,10]

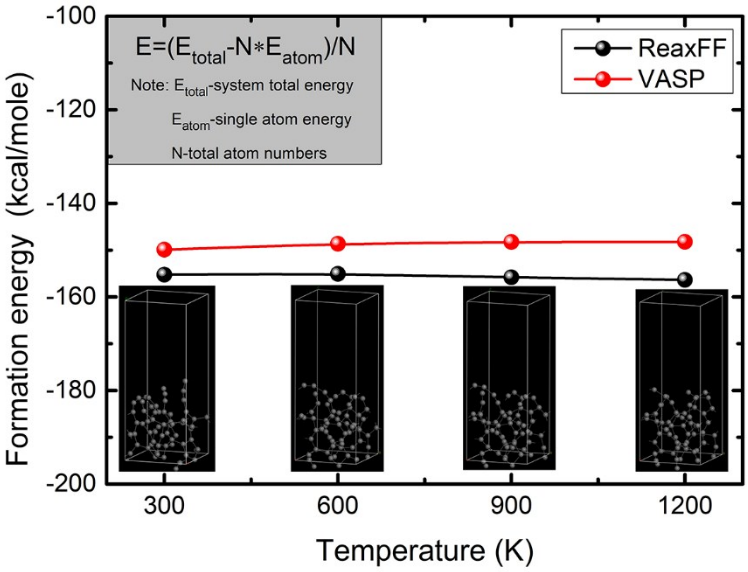


Fig. S7. Temperature-dependence of formation energy of a-C structure calculated by ReaxFF MD and ab-initio calculations, respectively.

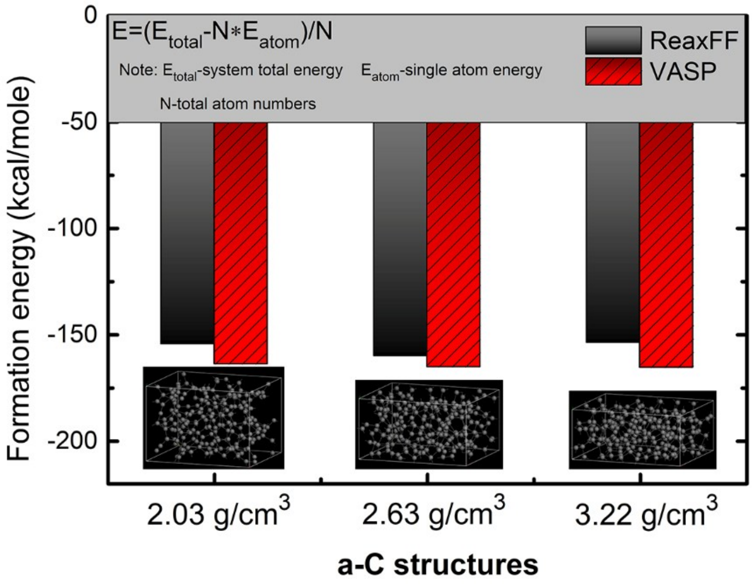
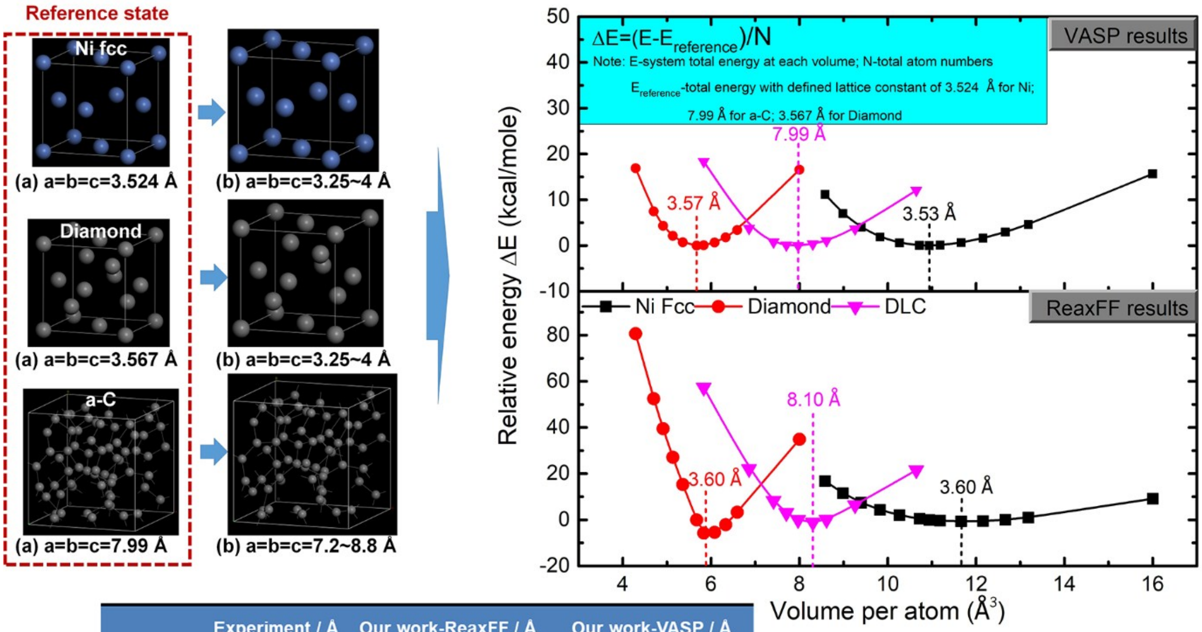
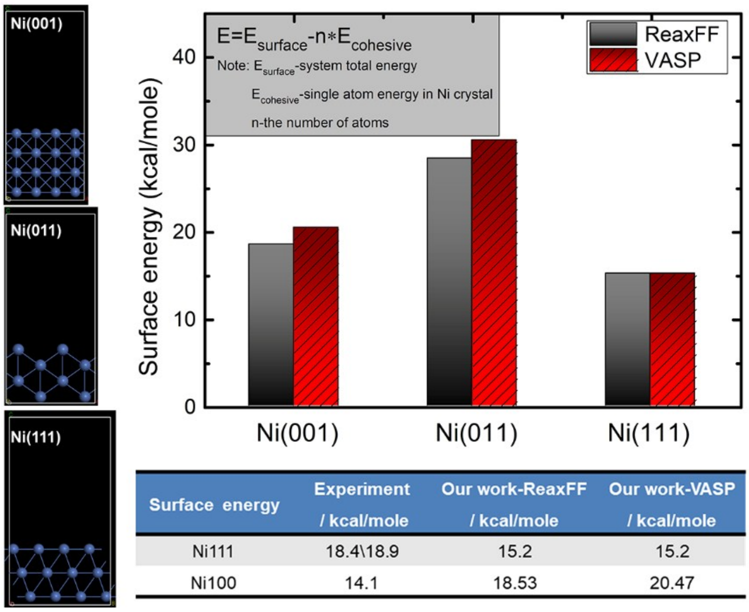


Fig. S8. Formation energies of a-C structures with different densities calculated by ReaxFF MD and ab-initio calculations, respectively.



	Experiment / Å	Our work-ReaxFF / Å	Our work-VASP / Å
Ni	3.524	3.60	3.53
Diamond	3.567	3.60	3.57

Fig. S9. Energy changes of diamond, Ni crystal, and a-C induced by expansion and compression of supercell size, which were calculated by ReaxFF MD and ab-initio calculations, respectively. The experimental values are also give for comparison.[1]



Surface energy	Experiment / kcal/mole	Our work-ReaxFF / kcal/mole	Our work-VASP / kcal/mole
Ni111	18.4\18.9	15.2	15.2
Ni100	14.1	18.53	20.47

Fig. S10. Surface energy of Ni crystal calculated by ReaxFF MD and ab-initio calculations, respectively. The experimental values are also give for comparison.[1]

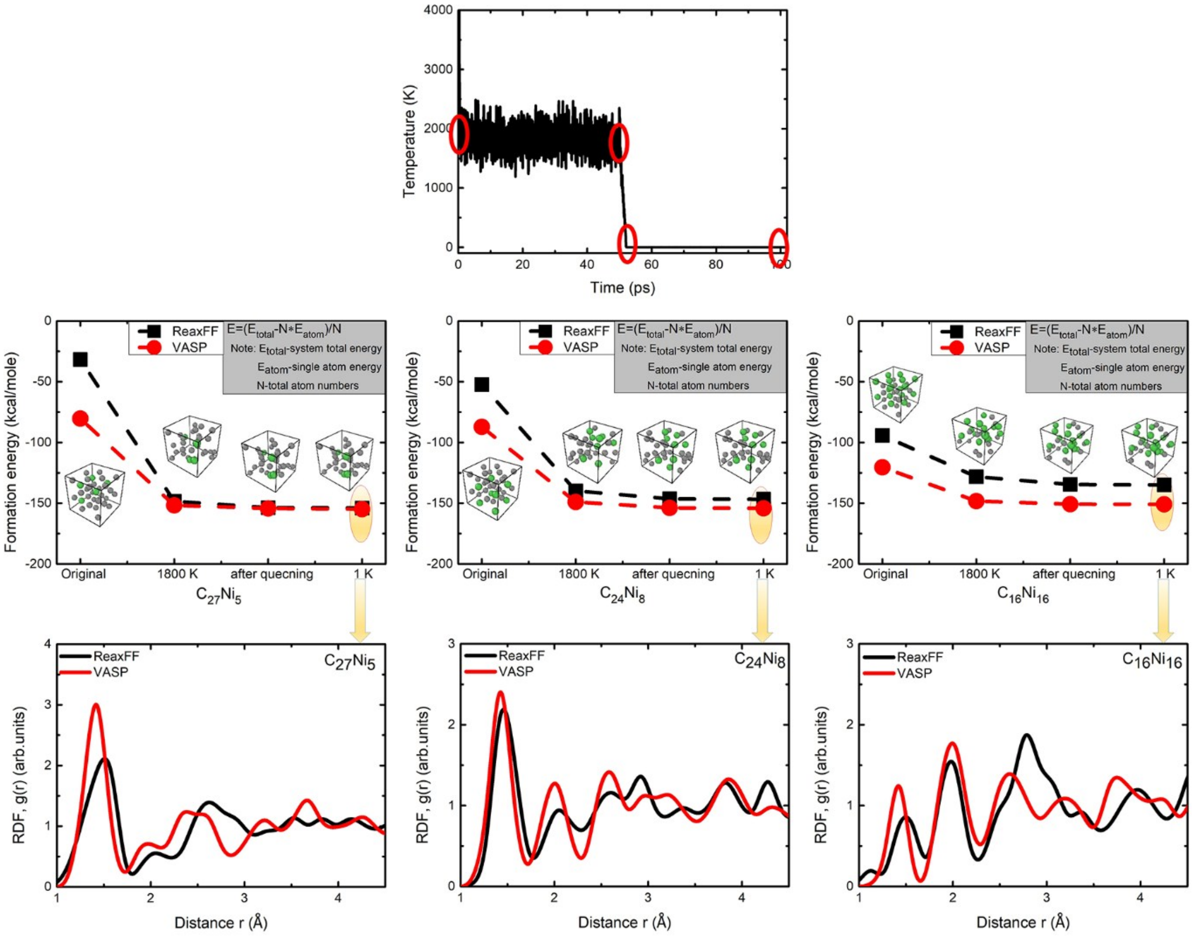


Fig. S11. Formation energies and RDF spectra of Ni-C mixing structures generated at the different stages of quenching process.

References

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