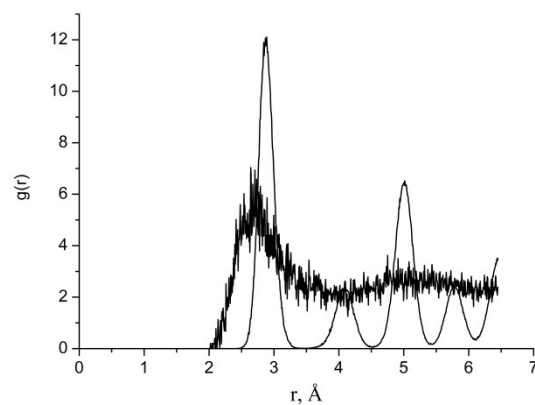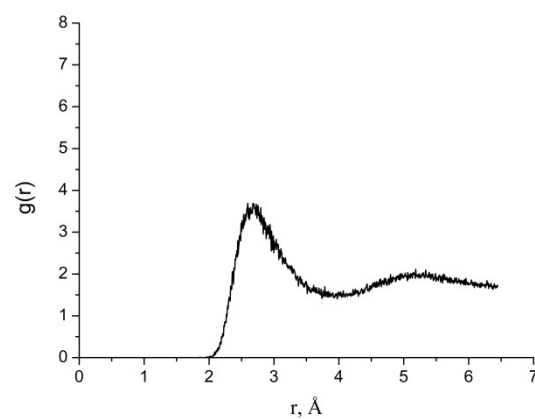## Supplementary Materials

(a)

(b)

Figure 1S. Calculated radial distribution function (RDF) for Au cluster with R=20uc, (a) -at t=0.1 ps and (b) – at t=10 ps. In figure (a) all the results correspond to Au cluster with R=50uc showing partial melting that is then completed in (b). Calculated radial distribution function (RDF) (50) for Au cluster with radius R=50uc obtained at t= 10 ps.
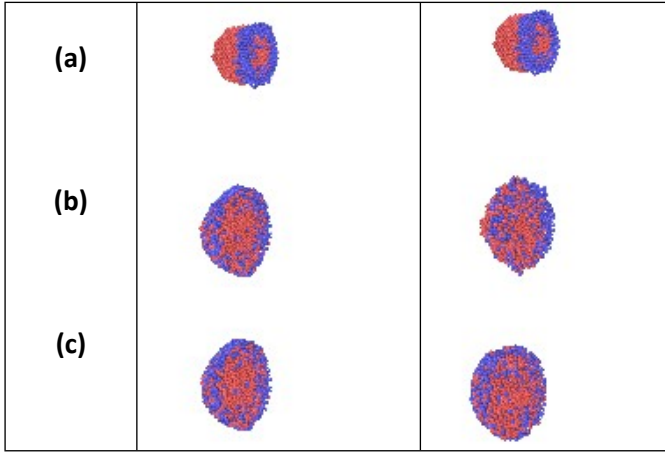
Figure 2S. Snapshots obtained in the heating of Janus AuNi nanoparticles with heating shown above. Slice view calculated at 5, 80, and 100 ps, the left column is for $T_{max}=400$ and the right one is for $T_{max}=2700K$.

Equation (S1)

$$G^{XS} = X_{Ni}X_{Au}(24,140X_{Au} + 38,280X_{Ni} - 14,230X_{Au}X_{Ni})\left(1 - \frac{T}{2660}\right)J$$

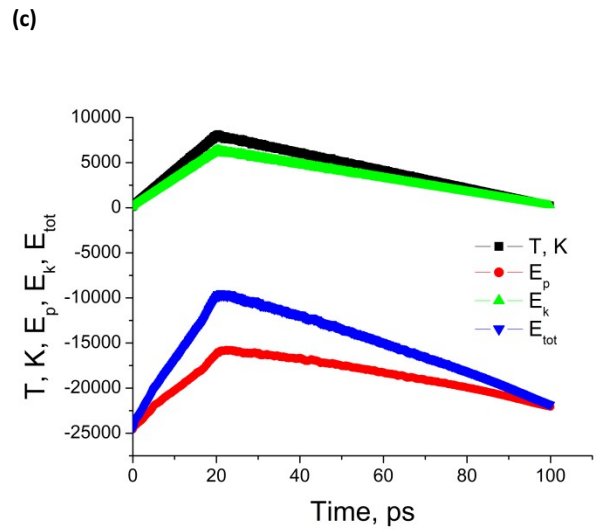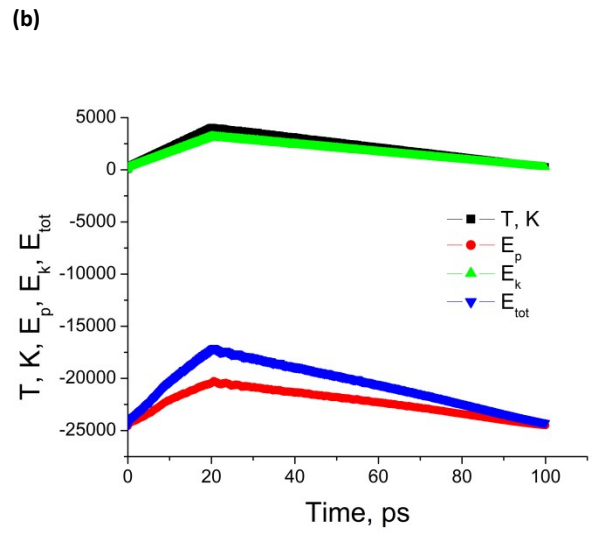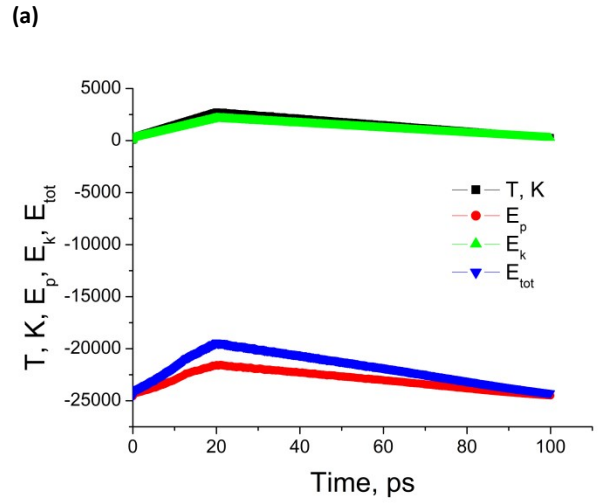where X_{Ni} and X_{Au} are Ni and Au molar fractions, respectively (63-65).

Figure 3S. Calculated temperature and NP energies vs time for AuNi Janus NP and $T_{max}=2700K$ (a), 4000 K (b), and 8000 K (c). Energy units are cal/mol.