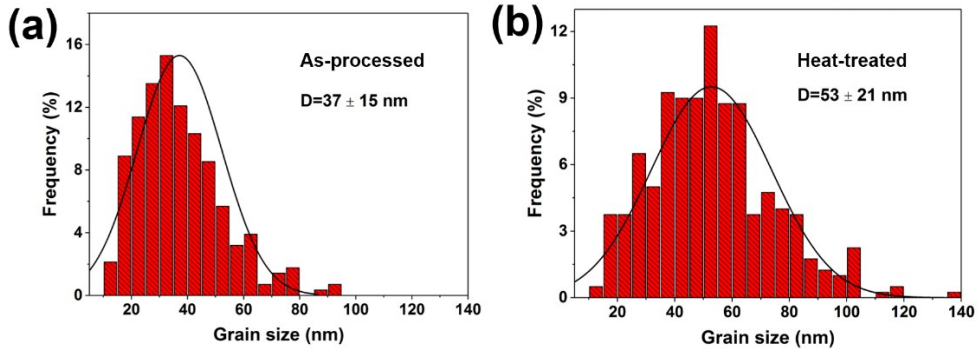
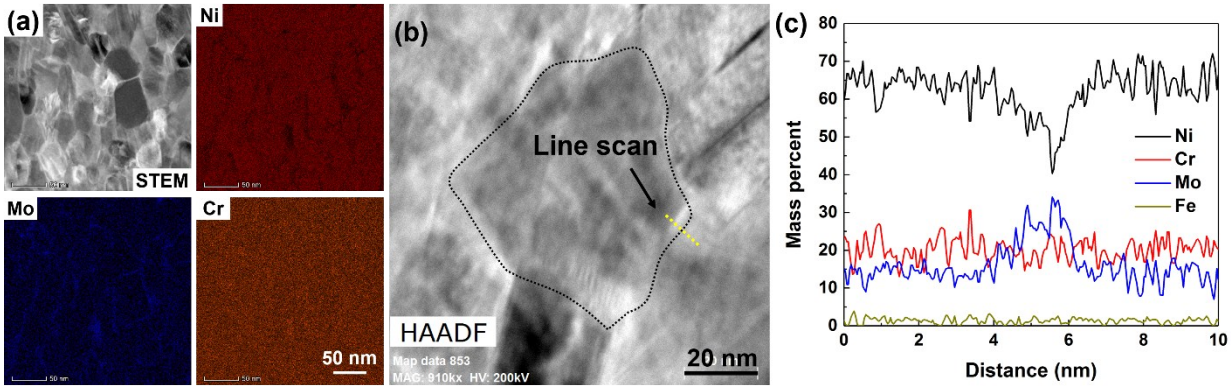
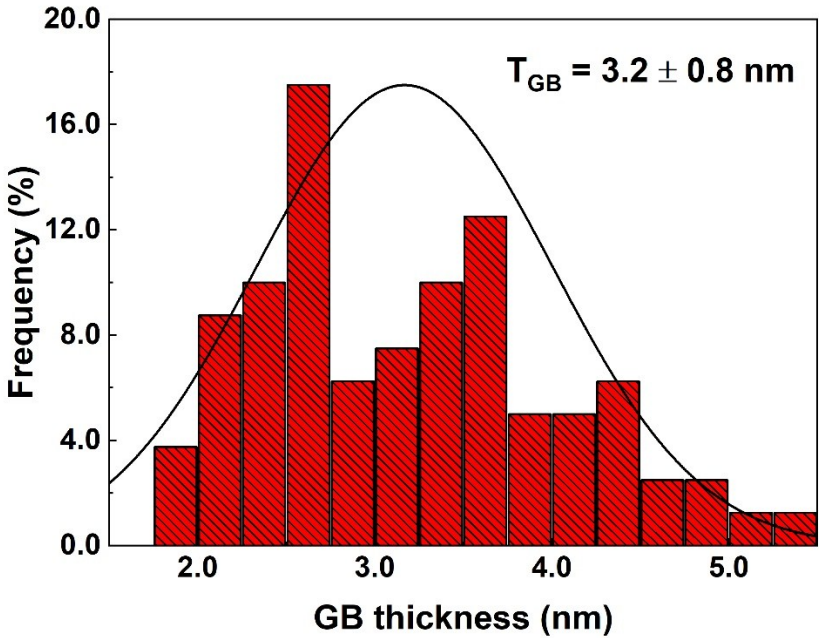
Supplementary



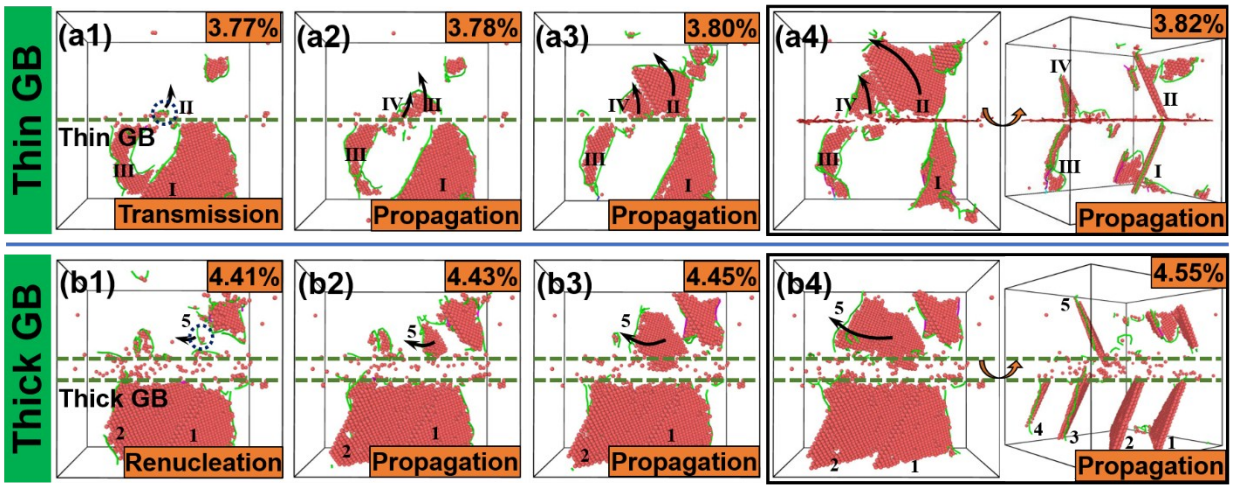
Suppl Figure 1. Grain size distribution of as-processed and heat-treated Ni alloy in the depth of ~8 μm from treated surface.



Suppl. Figure 2. (a) The STEM image and corresponding EDS maps showing the distribution of major chemical elements (Ni, Mo, Cr) of heat-treated Ni alloy sample. (b and c) The STEM image and corresponding EDS line scanning results along the line that crossing thick GBs in the STEM image in Fig.b showing the enrichment of Mo and depletion of Ni in the boundary area.



Suppl. Figure 3. The distribution of GB thickness for the heat-treated specimen.



Suppl. Figure 4. Evolution of atomistic configurations showing (a) the transmission of Shockley partials across the thin GB and thereafter propagate upwards, (b) the renucleation of a new Shockley partial on the other side of thick GB and thereafter propagate parallelly with the thick GB. Fig.a4 reveals that the two Shockley partials transmitted through the thin GB follow the Σ5 twinning orientation relationship with those in the lower grain. Whereas the new Shockley partials nucleated randomly on the upper grain for thick GB sample after yielding in Fig.b4.

Suppl video 1. Video captured during the *in situ* compression test of the thin GB sample. The video has been accelerated by 5 times.

Suppl video 2. Video captured during the *in situ* compression test of the thick GB sample. The video has been accelerated by 5 times.

Suppl video 3. Video of the MD simulation results of the thin GB sample viewing from plan view and edge-on view direction.

Suppl video 4. Video of the MD simulation results of the thick GB sample viewing from plan view and edge-on view direction.