Supplementary material

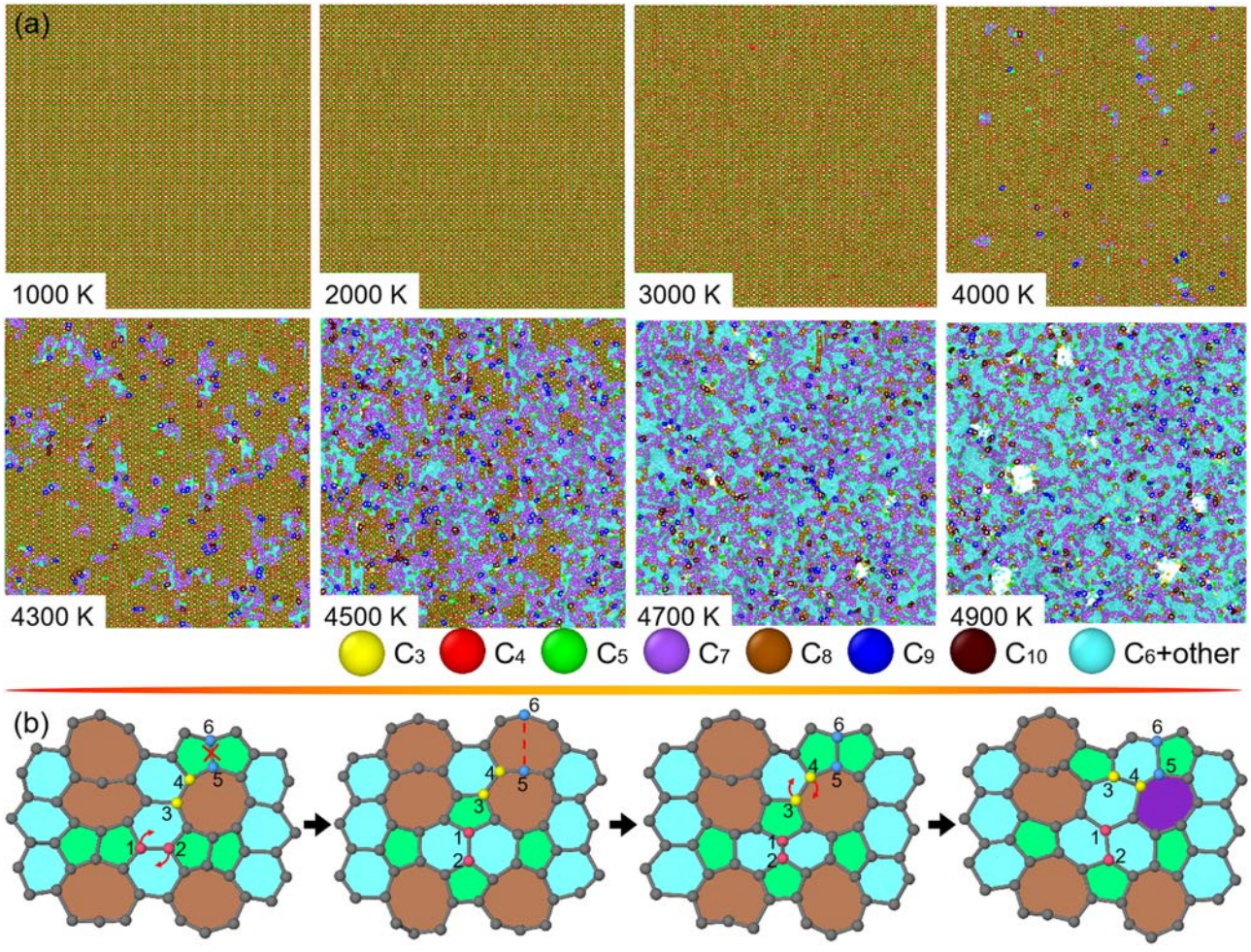

Figure 1: Structural development in HOP-graphene subjected to heating. (a) Top-viewed snapshots of HOP-graphene as the heating *T* changes from around 1,000–4,900 K. For eye-catching the thermally-induced structural transformation, carbon atoms in polygonal rings from trigon to decagon are differently-colored. (b) Atomic illustration of nucleation of hexagons from pentagons, hexagons and octagons in a local region of HOP-graphene. For clarification, some carbon atoms are number-ranked and arrows are marked for revealing the origins of microstructural transitions.

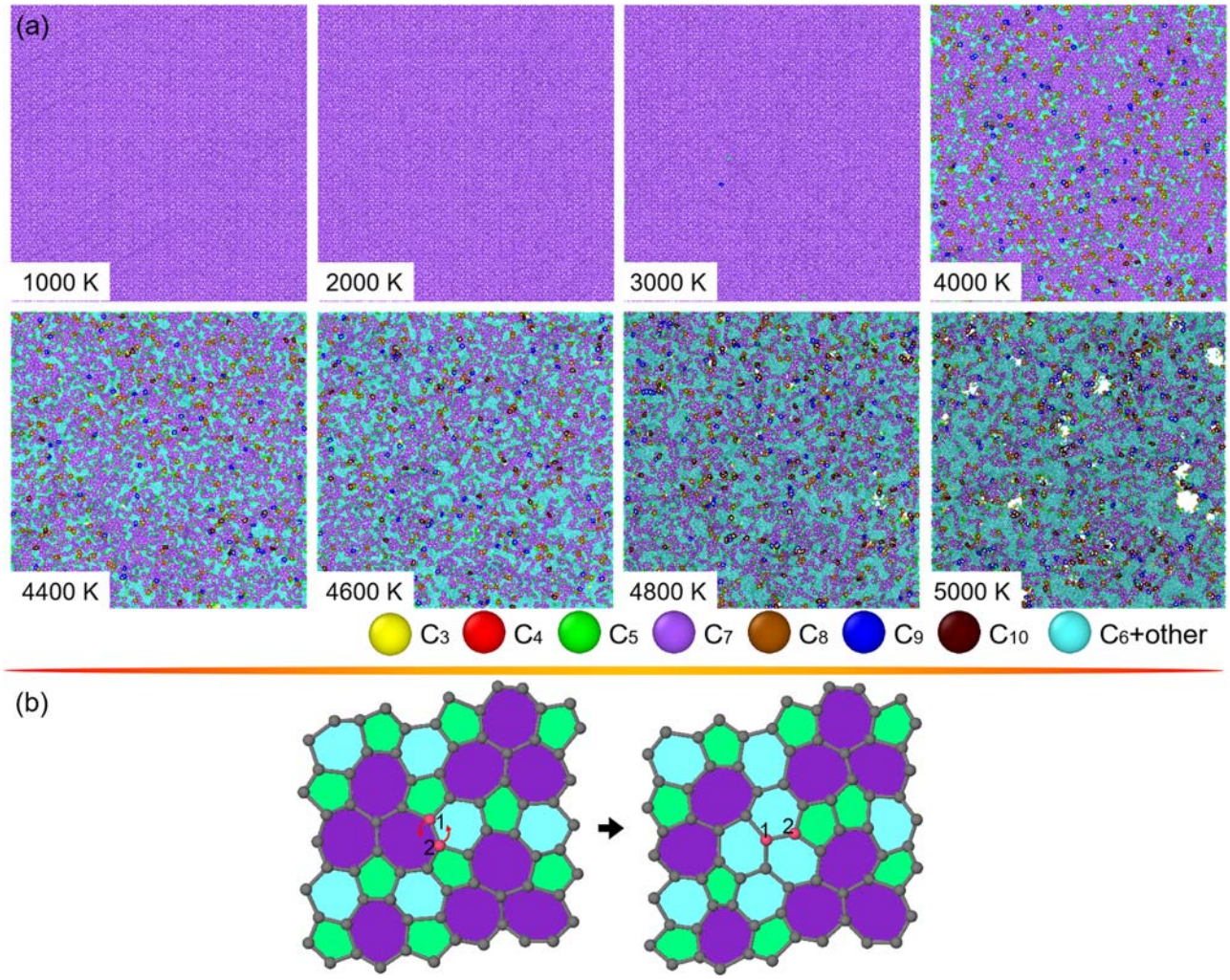

Figure 2: Structural development in H-haeckelite subjected to heating. (a) Snapshots of H-haeckelite when the heating *T* changes from around 1,000–5,000 K. For eye-capturing the thermally-induced structural transformation in the H-haeckelite, carbon atoms in polygonal rings from trigon to decagon are differently-highlighted. (b) Atomic illustration of nucleation of hexagons from pentagons, hexagons and heptagons in a local region of H-haeckelite. For clarification, some carbon atoms are number-ranked and arrows are marked for revealing the origins of microstructural transitions.

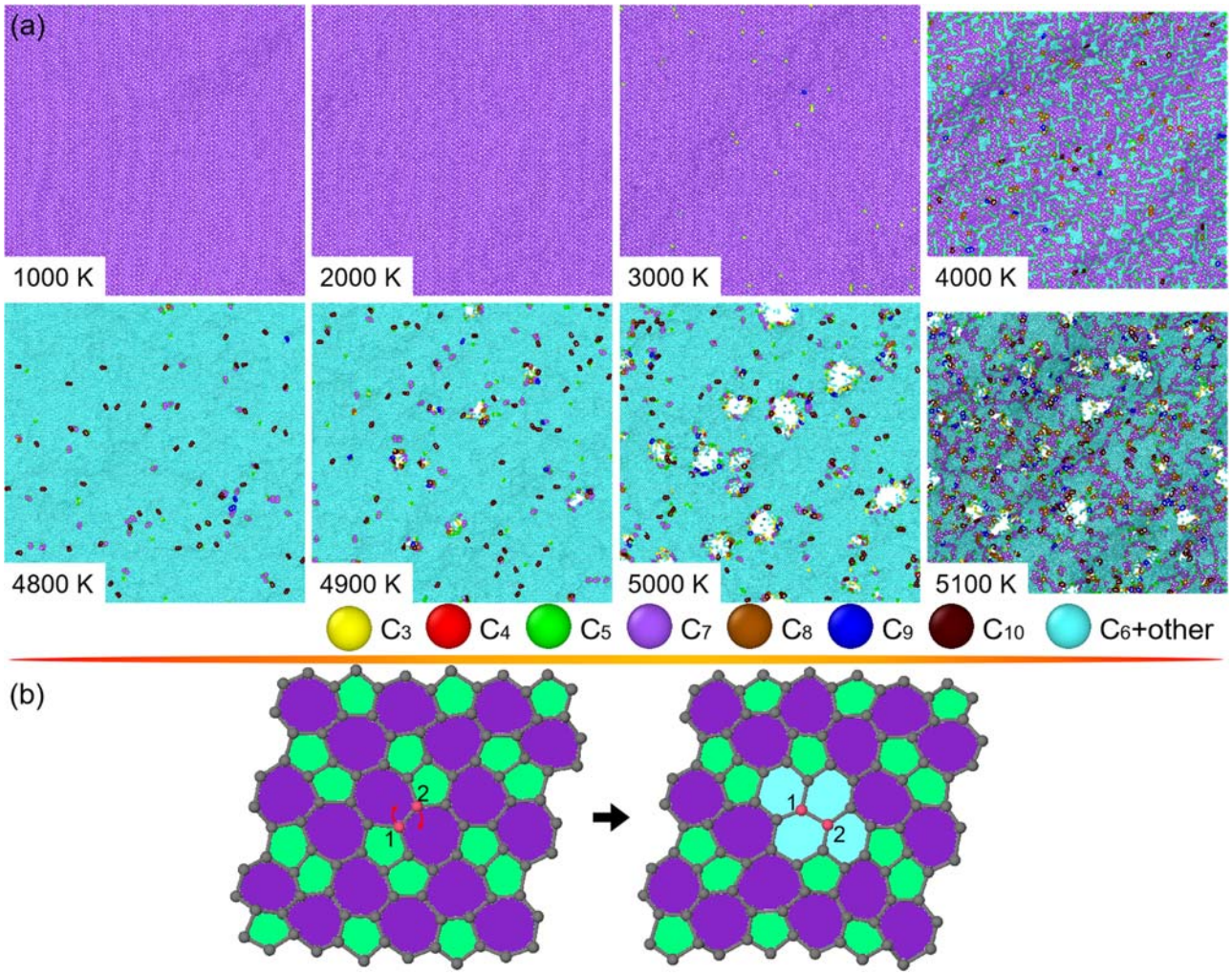

Figure 3: Structural changes in R-haeckelite upon heating. (a) Top-views of R-haeckelite as the heating *T* changes from around 1,000–5,000 K. For eye-catching the thermally-induced structural transformation in the R-haeckelite, carbon atoms in polygonal rings from trigon to decagon are differently-highlighted. (b) Atomic illustration of nucleation of hexagons from pentagons, hexagons and heptagons in a local region of R-haeckelite. For clarification, some carbon atoms are number-ranked and arrows are marked for revealing the origins of microstructural transitions.

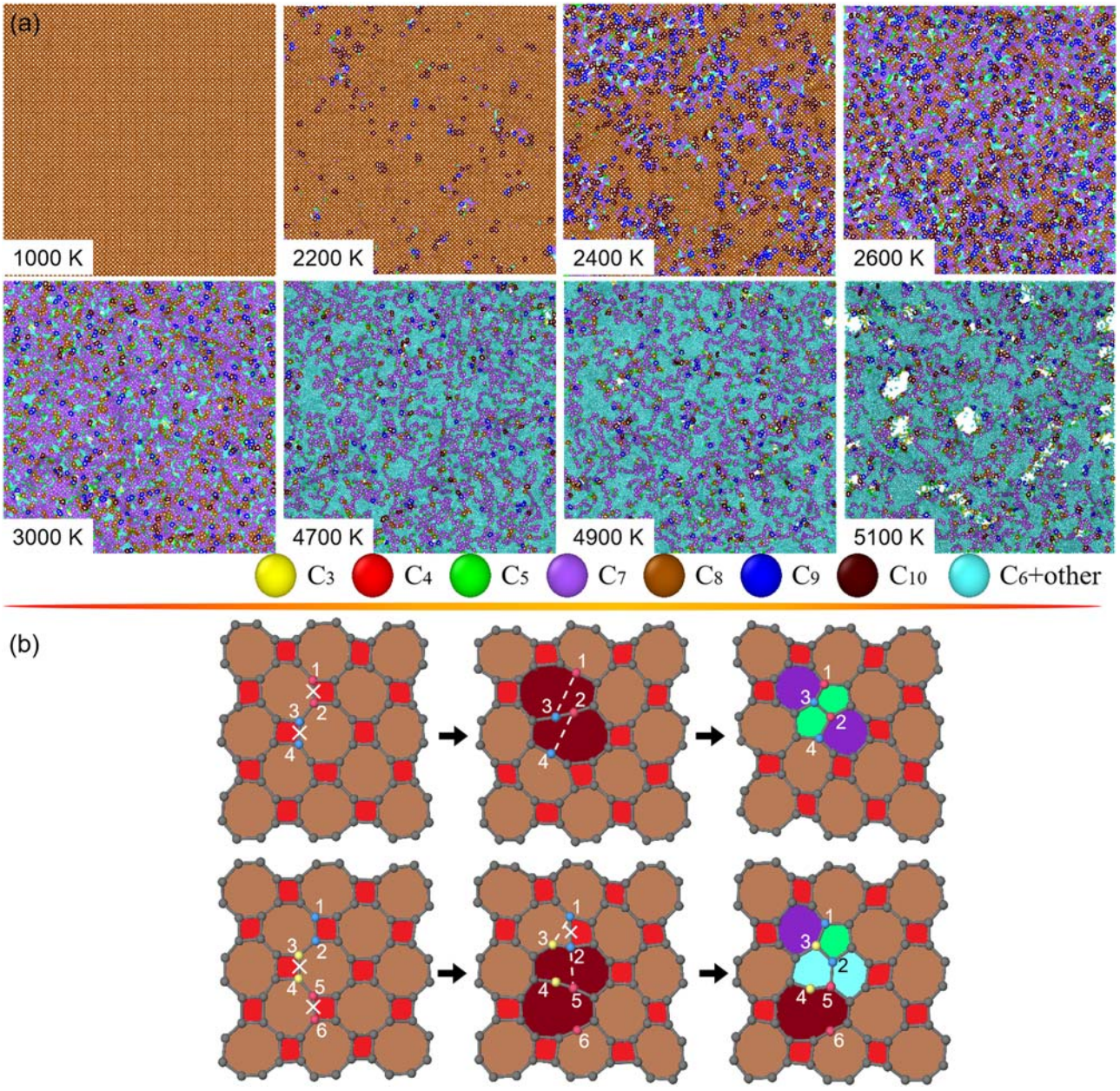

Figure 4: Structural changes in T-graphene upon heating. (a) Top-views of T-graphene as the heating *T* increases from around 1,000–5,100 K. For observing the thermally induced structural transformation carbon atoms in polygonal rings from trigon to decagon are color-stressed. (b) Atomic illustration of hexagon nucleation in a local region of T-graphene. Particularly, some carbon atoms are number-ranked and "X" symbol are marked for uncovering the origins of microstructural transitions.